INVESTIGATIONS OF THE COHERENCE PROPERTIES OF SCATTERED LIGHT

by

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ABSTRACT

We consider the statistical properties of electromagnetic radiation scattered by many particle systems. Using techniques which have been developed to treat the coherence properties of the radiation field in quantum mechanical terms, we obtain statistical properties of the light scattered by inelastic processes such as Raman scattering and scattering by density fluctuations. We emphasize the first and second order temporal correlation functions of the radiation field and find that, for the processes considered, they are related to each other by an equation which is very similar to that which is expected to hold if the radiation field is taken to be a classical narrow band Gaussian process. The suggestion that the statistical properties of matter near a phase transition critical point are not Gaussian lead us to construct a theory of the statistical properties of a Heisenberg-Ising model ferromagnet. We obtain equations of motion which describe the non-equilibrium evolution in time of the system towards thermal equilibrium by considering the ferromagnet coupled to a thermal reservoir. We find that the dynamical behavior is different for different reservoirs, of which we consider two types in detail, but that the steady state (thermal equilibrium) properties are independent of the specific nature of the reservoir. We show that our steady state solutions lead to the Landau form for the magnetic free energy near the Curie temperature, and to the molecular field equation of state and the Bragg-Williams form of the free energy for all temperatures. Our density operator equation of motion method avoids the explicit statistical counting procedures employed in conventional derivations of the free energy.

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INTRODUCTION

The atomic properties of matter have been investigated principally by studying the interaction of matter with electromagnetic fields. The earliest studies revealed that atoms of pure elements emit and absorb electromagnetic radiation at a sequence of discrete frequencies, each sequence or pattern being characteristic of the particular element under study and different from the patterns of other elements. Spectroscopy thus provided the empirical data upon which atomic theory was later constructed, and even today the study of the emission and absorbtion of radiation by atomic systems continues to provide information about the structure of complex atoms and molecules.

A complementary method in the study of matter by means of its interaction with electromagnetic radiation is the observation of the scattering, rather than the emission or absorbtion, of radiation. The properties of individual atoms or molecules as well as the structure of aggregates of particles - gases, liquids, and solids - may be studied by light scattering. With regard to the former category, the Raman effect, first observed in 1928[1], provides the basis of the experimental techniques which center on the study of the spectrum of the scattered light. The presence in the scattered light of frequencies other than those in the incident light which illuminates the scatterer yields information about the quantum states of the target. Thus, many atomic and molecular species have been studied in this way [2].

Since the inelastic scattering of light is generally very weak, observation of the effect is enhanced by increasing the number of atomic or molecular scatterers. Thus, Raman scattering studies were frequently done with liquid targets. However, in addition to scattering light via the Raman effect at frequencies characteristic of its constituent atoms or molecules, a liquid (gas and solid as well) scatters light because it is not perfectly homogeneous. That is, fluctuations of the density of atoms of the fluid over distances of the order of an optical wave length cause fluctuations of the dielectric constant which in turn cause a light beam to be scattered away from the forward direction. Studies of the fluctuation scattering of light [3] go back to Tyndall and Lord Rayleigh in the latter part of the 19th century, although Brillouin was one of the first to relate the scattering of light by density fluctuations to a model of the properties of the scatterer. First inspired by the Debye model of a solid, and later by a study of hypersonic waves in liquids, Brillouin predicted [4] that a liquid or solid should scatter light by means of thermal sound waves which continuously exist in any body at a non-zero temperature. These sound waves would appear to an incident light wave as a regular alternation of the density of the body and thus would act like a moving diffraction grating.

The scattered light would be Doppler shifted in frequency from the incident light by an amount proportional to the velocity of the sound wave in the body. Thus, Brillouin proposed that the structure of a liquid could be studied by observing the light inelastically scattered from it.

There were very great experimental obstacles to the observation of Brillouin scattering. Besides the very weak character of the scattering, which thus required very intense sources of illumination, the frequency shifts expected were extremely minute. Hence, the source of radiation had to have a very narrow spectrum in order to be able to distinguish the Brillouin scattering from the elastically scattered light which was always present - due in part to dust impurities in the sample and in part to intrinsic elastic scattering by the fluid, an effect treated by L.D. Landau and G. Placzek in 1934[5]. It was not until quite recently that major advances in both light sources and detectors made the observation of fluctuation scattering in liquids relatively easy.

The laser provided a light source of greatly increased intensity relative to discharge tubes. At the same time, the laser also provided an extremely narrow spectrum of light, so that two of the main experimental difficulties were overcome at once. However, a great deal of work on lasers[6,7,8,9] revealed inadequacies in the description of electromagnetic fields hitherto used. It was discovered that the characterization of the radiation field by its power (amplitude) spectrum was not sufficient to completely describe the state of the radiation emitted by a laser. In addition, new electronic methods of detecting radiation, such as the photomultiplyer tube, emphasized the quantum nature as well as the fluctuation or noise properties of the optical field. To deal with these problems, a new general description of physically realizable radiation fields was evolved [10].

The method developed characterizes the radiation field by giving the values of average properties of the field. Besides the average electric field strength, a complete description of the field involves specification of average values of the product of two field strengths (evaluated at different space and time points), three field strengths, and so on. The radiation field is thus looked upon as a stochastic system. The statistical properties of such a system are fully known only in terms of joint multitime probability distributions or equivalently, the expectation values of products of variables computed with respect to the joint probability densities. This statistical description of the electromagnetic field is analogous to the characterization of a many-body system by an infinite hierarchy of multiparticle Greens functions or correlation functions.

Considerable work has been done on the statistical properties of laser radiation. In particular, it is known that a laser produces light which is not Gaussian in its statistics [6,8]. Therefore, the question arises as to how the statistical properties of laser radiation are modified by scattering processes of the types mentioned above. Is the characterization of the scattered light by its frequency spectrum an adequate description, or can additional useful information about the target be extracted by measuring higher order correlation functions of the scattered field? There is a great deal of information about the scatterer contained in measurements of the intensity distribution and frequency spectrum of scattered radiation [11,12] and one may well wonder if measuring higher order correlation functions would provide more. We shall attempt to answer in part this question in Part A of this work by presenting calculations of the coherence properties of light scattered by systems which model the processes mentioned above. In Section I of Part A we define the statistical properties of optical fields and demonstrate that a certain class of density operators leads to field correlation functions which obey a Gaussian-like factorization relation. In Section II of Part A we study a model of Raman scattering by density operator techniques. In Section III of Part A we calculate temporal correlation functions of the light scattered by density fluctuations in a particular model.

The suggestion [13] that the validity of thermodynamic fluctuation theory, which generally results in Gaussian statistics for fluctuating thermodynamic variables in a system in thermal equilibrium, may be in doubt near a phase transition critical point of a many particle system leads us to consider the statistical properties of such a system in Part B. We choose to investigate the Heisenberg-Ising model of a ferromagnet [14] and construct a density operator theory of this system in order to obtain from a microscopic calculation its statistical properties. In Section I of Part B we obtain an equation of motion for the density operator of the spin system which describes the time evolution of the ferromagnet through non-equilibrium states toward thermal equilibrium by considering the system to be in contact with a thermal reservoir. In Section II of Part B we introduce a c-number quasiprobability density distribution function [15,16] and obtain its equation of motion from the density operator equation of motion by techniques [17] developed originally to treat properties of the electromagnetic field interacting with atoms in a laser. In Section III of Part B we treat the equation of motion by an approximation expected to be valid in the critical region and show that although the dynamical properties of the system depend upon the details of the reservoir to which it is coupled - we consider two particular kinds of thermal reservoirs in this Section - the steady-state solution is independent of those details.

In particular, we obtain a non-Gaussian form for the steady state probability distribution of the magnetization which leads us to identify the free energy of the magnet as being of the form predicted by the Landau theory of a second order phase transition [18]. In Section IV of Part B we use a discrete representation to show that the exact steady state solution of our equation of motion yields the Bragg-Williams form [19] of the magnetic free energy for all temperatures. We obtain this result without resorting to any of the usual statistical counting procedures commonly employed in deriving it.

PART A: STATISTICS OF SCATTERED LIGHT

Part A. Section I. Optical Coherence and Gaussian Statistics

According to classical electromagnetic theory, the electric field $\vec{E}(\vec{r},t)$ of a radiation field may be written in general as a linear super position of normal modes of the form [20]

$$\vec{E}(\vec{r},t) = \sum_{\vec{k}} C_{\vec{k}}\, \vec{u}_{\vec{k}}(\vec{r})\, e^{-i\omega_{\vec{k}} t} + \sum_{\vec{k}} C^{*}_{\vec{k}}\, \vec{u}^{*}_{\vec{k}}(\vec{r})\, e^{i\omega_{\vec{k}} t} \tag{1.1a}$$

$$\equiv \vec{E}^{(+)}(\vec{r},t) + \vec{E}^{(-)}(\vec{r},t) \tag{1.1b}$$

where the mode functions $\vec{u}_{\vec{k}}(\vec{r})$ and frequencies $\omega_{\vec{k}}$ satisfy the wave equation

$$\left(\nabla^2 + \frac{\omega_{\vec{k}}^2}{c^2}\right) \vec{u}_{\vec{k}}(\vec{r}) = 0 \tag{1.2a}$$

with the condition

$$\vec{\nabla} \cdot \vec{u}_{\vec{k}}(\vec{r}) = 0 \tag{1.2b}$$

and appropriate boundary conditions. The positive (+) and negative (-) frequency components of the field are defined by (1.1b) and are complex conjugates of each other. Hence, the electric field is entirely determined by specifying the values of the set of complex constants $\{C_{\vec{k}}\}$.

However, the sources of either natural or man-made radiation fields are atomic in nature. Each tiny atomic radiator contributes to the total radiated field, but it is impossible to arrange them such that they all radiate together in a coherent fashion. To a greater or lesser extent, the individual atomic radiators emit their fields randomly, different groups of atoms of a macroscopic source radiating for short periods of time. Therefore, the fields in nature cannot be wholly specified at each instant of time. Rather, they exhibit fluctuations or noise properties and measuring devices that respond to some function of the electric field, say f($\vec{E}$), record average values of that function $\langle f(\vec{E})\rangle$:

$$\langle f(\vec{E})\rangle = \int f\left[\vec{E}(\{C_{\vec{k}}\})\right] P(\{C_{\vec{k}}\}) \prod_{\vec{k}} d^2 C_{\vec{k}} \qquad (1.3)$$

where $d^2 C_{\vec{k}} = d(\mathrm{Re}\, C_{\vec{k}})\, d(\mathrm{Im}\, C_{\vec{k}})$ and the statistical properties of the field are contained in the probability distribution of the set of expansion coefficients $\{C_{\vec{k}}\}$:

$$P(\{C_{\vec{k}}\}) = P(C_{\vec{k}_1}, C_{\vec{k}_2}, \cdots) \qquad (1.4)$$

The quantum-mechanical description of electromagnetic radiation regards the electric field as an operator. It possesses a mode expansion analogous to (1.1a) where the expansion coefficients $\{C_{\vec{k}}\}$ are replaced by operators:

$$\vec{E}^{(+)}(\vec{r},t) = i \sum_{\vec{k}} \left(\tfrac{1}{2}\hbar\omega_{\vec{k}}\right)^{\frac{1}{2}} a_{\vec{k}}\, \vec{U}_{\vec{k}}(\vec{r})\, e^{-i\omega_{\vec{k}} t} \qquad (1.5a)$$

$$\vec{E}^{(-)}(\vec{r},t) = \left[\vec{E}^{(+)}(\vec{r},t)\right]^{+} \tag{1.5b}$$

The positive and negative frequency parts of the electric field operator are Hermitian conjugates of each other while the photon annihilation and creation operators ($a_{\vec{K}}$, $a^{+}_{\vec{K}}$) obey Bose commutation relations:

$$[a_{\vec{K}}, a^{+}_{\vec{K}'}] = \delta_{\vec{K},\vec{K}'} \; ; \; [a_{\vec{K}}, a_{\vec{K}'}] = [a^{+}_{\vec{K}}, a^{+}_{\vec{K}'}] = 0 \tag{1.6}$$

The statisitical nature of the electromagnetic field is now properly specified by a density operator ρ.

As pointed out by Glauber [20], radiation detectors commonly in use function by absorbtion of photons. For this reason it is useful to define a set of electric field correlation functions as they are precisely the field quantities determined by conventional experiments. The general nth order electric field correlation function is defined by

$$G^{(n)}(x_1 \cdots x_n, x_{n+1} \cdots x_{2n}) \equiv \mathrm{Tr}\left\{\rho \vec{E}^{(-)}(x_1) \cdots \vec{E}^{(-)}(x_n)\vec{E}^{(+)}(x_{n+1}) \cdots \vec{E}^{(+)}(x_{2n})\right\} \tag{1.7}$$

where $x = \{\vec{r}, t\}$. Note that $G^{(n)}$ in general depends upon the 2n polarization vectors of the fields, but this dependence has been suppressed in (1.7) as we shall not be significantly concerned with these properties of the correlation functions. We shall frequently refer to the first order (or amplitude) correlation function given by

$$G^{(1)}(t,t') = Tr\{\rho E^{(-)}(t) E^{(+)}(t')\} \qquad (1.8)$$

and the second order (intensity) correlation function given by

$$G^{(2)}(t,t';t',t) = Tr\left\{\rho E^{(-)}(t) E^{(-)}(t') E^{(+)}(t') E^{(+)}(t)\right\} \qquad (1.9)$$

We have suppressed the spacial arguments in (1.8) and (1.9) as we will primarily be interested in the temporal coherence properties of the radiation field (we imagine all field operators in the correlation functions to be evaluated at the same space point).

The significance of the first and second order correlation functions is this: frequently it is argued that the statistical properties of the radiation field ought to be Gaussian. That is, classically one supposes that the stochastic properties of the field may be obtained by treating the electric field as a c-number (that is, non-operator) Gaussian random process. For such a process, it may be shown [21] that all correlation functions higher than the first order may be expressed in terms of the first order correlation function. In particular, one has that

$$\langle |E(t)|^2 |E(t+\tau)|^2 \rangle = \left(\langle |E(t)|^2 \rangle\right)^2 + |\langle E(t+\tau) \overset{*}{E}(t) \rangle|^2 \qquad (1.10)$$

The method of light beating (or self beat) spectroscopy [22,23,24] in fact directly measures the intensity spectrum of the radiation field, that is $G^{(2)}$, rather than the amplitude spectrum $G^{(1)}$. One needs a relation like (1.10) in order to interpret the results of such experiments in terms of $G^{(1)}$, or its Fourier transform, about which most theoretical predictions are made. Indeed, most self beat experimenters assume that the radiation field is adequately described as a Gaussian process and thus use (1.10) directly. It is clear that the analysis of such experiments crucially depends upon this assumption.

A basic theorem of statistics is usually invoked to justify the assumption that the radiation field of scattered light is Gaussian. If one is observing light scattered by density fluctuations, the total scattered field at the site of the detector is the sum of fields scattered by density fluctuations throughout the (macroscopic) scattering volume. It is argued that, by the central limit theorem of statistics[25], whatever the statistics of the density fluctuations in volumes of the order of an optical wavelength on a side, the actual scattering volume contains so many such regions that the sum of the electric fields scattered by these fluctuations must have Gaussian statistics. In addition, one can show from thermodynamic fluctuation theory that the fluctuations of the medium themselves have Gaussian statistics [13]. The valid application of thermodynamic fluctuation theory to the large fluctuations near a critical point is however by no means certain.

The techniques of self beat spectroscopy are particularly useful in investigations of critical phenomena and phase transitions [13,24]. However, it is just in such phenomena that one expects density fluctuations to be of long range. Should the range of fluctuations extend over the entire scattering volume, the incident beam would be scattered as by a mirror, and one would expect the scattered light to have statistical properties very much like those of the incident beam. Since the source of illumination for such experiments is invariably a laser, and since it is known [6,8] that the radiation emitted by a laser is not well described by Gaussian statistics, one might question the validity of analyzing the results by making the Gaussian assumption (1.10).

Hence, it is clear that the question of the statistics of the scattered light is of importance in practice as well as of intrinsic interest. It shall be studied by some model calculations in the succeeding two sections, and the question of critical fluctuations will be discussed in Part B. However, before proceeding to specific models of the scattering process, we demonstrate a result which is the quantum mechanical analogue of (1.10).

A very similar relation between the first and second order electric field correlation functions, as defined by (1.8) and (1.9), can be shown to hold in general whenever the density matrix of the radiation field factors into a product of single mode (diagonal) density matrices. That is, assuming that

$$\langle\{n\}|\rho|\{n'\}\rangle = \rho_{\{n\}\{n\}}\,\delta_{\{n\}\{n'\}} \tag{1.11}$$

(where $\{n\}=\{n_1, n_2, \cdots\}$ labels the occupation numbers of the various modes) and

$$\langle\{n\}|\rho|\{n\}\rangle = \rho_{\substack{n_1 n_2\cdots\\ n_1 n_2\cdots}} = \prod_{j=1}^{\infty} \rho^{(j)}_{n_j n_j} \tag{1.12}$$

then the following relation exists between the first and second order electric field correlation functions:

$$G^{(2)}(t,t';t',t) = G^{(1)}(t,t')\,G^{(1)}(t',t) + G^{(2)}(t,t;t,t) - G^{(1)}(t,t)\,G^{(1)}(t',t') \tag{1.13}$$

For a stationary process [20], the correlation functions depend only on the variable $\tau = t'-t$ so that the Fourier transform of this relation gives

$$G^{(2)}(\omega) = 2\pi\,\delta(\omega)\left[G^{(2)}(t,t;t,t) - G^{(1)}(t,t)\,G^{(1)}(t',t')\right] + \frac{1}{2\pi}\,G^{(1)}(\omega)\otimes G^{(1)*}(\omega) \tag{1.14}$$

where the intensity spectrum $G^{(2)}(\omega)$ and the amplitude spectrum $G^{(1)}(\omega)$ are defined by

$$G^{(2)}(\omega) = \int_{-\infty}^{\infty} d\tau\, e^{-i\omega\tau}\, G^{(2)}(0,\tau;\tau,0) \tag{1.15}$$

$$G^{(1)}(\omega) = \int_{-\infty}^{\infty} d\tau\, e^{-i\omega\tau}\, G^{(1)}(\tau, 0) \tag{1.16}$$

and the convolution product is defined by

$$G^{(1)}(\omega) \otimes G^{(1)*}(\omega) = \int_{-\infty}^{\infty} d\omega'\, G^{(1)}(\omega')\, G^{(1)*}(\omega' - \omega) \tag{1.17}$$

For Gaussian fields, such as black-body radiation described by a Planck density operator, one has in addition that

$$G^{(2)}(t,t;t,t) = 2\left|G^{(1)}(t,t)\right|^{2} \tag{1.18}$$

so that relation (1.14) becomes identical to the one which holds for a classical Gaussian process, (1.10). In general, for non-Gaussian fields which obey the diagonality and factorization conditions, (1.11) and (1.12), the coefficient of the δ- function term in (1.14) differs from the one for Gaussian light, but the convolution of the amplitude spectrum with itself always constitutes the second term of that relation. For example, for a radiation field composed of a single monochromatic mode with Poisson statistics - that is, a single mode pure Glauber coherent state [20] of the field - the coefficient of the δ- function term vanishes identically. In the time domain, this means that the normalized intensity correlation function $g^{(2)}(t,t';t',t)$ defined by

$$g^{(2)}(t,t';t',t) = G^{(2)}(t,t';t',t)\left[G^{(2)}(t,t;t,t)\right]^{-1} \qquad (1.19)$$

decreases from a maximum value of one at $\tau = t'-t = 0$ to zero at $\tau \to +\infty$ (not to the asymptotic value of one half as it does for Gaussian light).

These results are derived in Appendix I. The relation (1.13), or the equivalent one in the frequency domain, (1.14), will be referred to in what follows as the Gaussian factorization relation between the first and second order correlation functions.

Part A. Section II. Statistics of Raman Scattering

Inelastic scattering of light by Raman processes is a useful method of investigating the properties of atoms and molecules [26]. In this section we shall study the statistical properties of the Raman scattered light by calculating the density matrix of the radiation field. We utilize a simple model of the process as a phenomenological description to avoid explicitly introducing details (which of course depend upon the specific nature of the scatterer) that are unimportant for the purposes of our study.

Therefore, we imagine a set of N two level atoms illuminated by an incident beam of frequency ω_0. We shall focus our attention on the light inelastically scattered into a particular direction and label by ω the different modes (all having propagation vectors in the same direction) of the scattered field. Thus, we take the Hamiltonian H of our system to be

$$H = H_R + H_A + V \qquad (2.1)$$

where H_R is the Hamiltonian of the free radiation field; H_A is the Hamiltonian for N (non-interacting) two level atoms whose states will be denoted by $|\alpha\rangle$ and $|\beta\rangle$, where the energy separation of these states (taken to be the same for all N atoms) is $E_\alpha - E_\beta = \hbar\Omega \geq 0$; and we take the interaction to be [27]

$$V = \hbar g \sum_{j=1}^{N} \sum_{\omega} \left\{ a_0 e^{i\vec{k}_0 \cdot \vec{r}_j} a_\omega^+ e^{-i\vec{k}_\omega \cdot \vec{r}_j} \sigma_j^+ + h.c. \right\} \tag{2.2}$$

Here the index zero refers to the incident beam and the atomic operators σ_j, σ_j^+ connect the upper and lower states in the following way:

$$\sigma_i |\xi\rangle = \delta_{\xi,\beta_i} |\beta_i\rangle ; \quad \sigma_i^+ |\xi\rangle = \delta_{\xi,\alpha_i} |\alpha_i\rangle ; \quad \xi = \alpha_i, \beta_i \tag{2.3}$$

The atomic states are taken to be orthogonal for different atoms and normalized to unity:

$$\langle \alpha_i | \beta_j \rangle = 0 ; \quad \langle \alpha_i | \alpha_j \rangle = \langle \beta_i | \beta_j \rangle = \delta_{ij} \tag{2.4}$$

We include all numerical constants and matrix elements (including sums over virtual intermediate states) in the single effective coupling constant g, which we note has the dimensions of a frequency.

Next we transform to the interaction picture to obtain:

$$\begin{aligned} V(t) &= e^{\frac{i}{\hbar}(H_R + H_A)t} \, V \, e^{-\frac{i}{\hbar}(H_R + H_A)t} \\ &= \hbar g \sum_{j=1}^{N} \sum_{\omega} \left\{ a_0 a_\omega^+ \sigma_j^+ e^{i(\vec{k}_0 - \vec{k}_\omega)\cdot \vec{r}_j} e^{-i(\omega_0 - \omega - \Omega)t} + h.c. \right\} \end{aligned} \tag{2.5}$$

The density operator for the total system (atoms and radiation) in the interaction picture, $\rho(t)$, obeys the equation of motion

$$i\hbar \dot{\rho}(t) = [V(t), \rho(t)] \tag{2.6}$$

It is convenient to formally integrate this equation

$$s(t) = s(-\infty) + \frac{1}{i\hbar}\int_{-\infty}^{t} dt' \left[V(t'), s(t')\right] \tag{2.7}$$

and insert the result into the right side of (2.6) to obtain

$$\dot{s}(t) = \frac{1}{i\hbar}\left[V(t), s(-\infty)\right] - \frac{1}{\hbar^2}\int_{0}^{t} dt' \left[V(t), \left[V(t'), s(t')\right]\right] \tag{2.8}$$

The statistical properties of the radiation field may be obtained from the reduced density operator of the field, ρ, defined by

$$\rho(t) = \mathrm{Tr}_{ATOMS}\, s(t) \tag{2.9}$$

We next write the density operator for the combined system in the form

$$s(t) = \rho(t)\otimes\rho^{M} + \Delta \tag{2.10}$$

where the matter density operator ρ^M is assumed not to vary with time and to have the following properties:

$$\rho^{M} = \prod_{j=1}^{N} \rho^{(j)} \;;\; \langle \xi_j | \rho^{(j)} | \xi_j' \rangle \propto \delta_{\xi_j, \xi_j'} \;;\; \mathrm{Tr}_{ATOMS}\, \rho^{M} = 1 \tag{2.11}$$

That is, ρ^M factors into a product of diagonal single atom matrices. Hence, taking the trace of (2.8) over atomic states yields

$$\dot{\rho}(t) = -\frac{1}{\hbar^2} \mathrm{Tr}_{\mathrm{ATOMS}} \int_{-\infty}^{t} dt' \left[V(t), \left[V(t'), \rho(t') \otimes \rho^{M}\right]\right] \tag{2.12}$$

where we have neglected the contribution of Δ since we assume that the interaction is weak enough to be adequately treated to second order. That Δ is of higher order is seen since if $V \equiv 0$ the two systems are uncorrelated and the density matrix always factors into the product of radiation and atomic density matrices.

We note that (2.12) depends on atomic variables through their correlation functions. We shall assume that the upper atomic state has a finite damping constant γ so that such atomic correlation functions decay rapidly for times t' very much different from t. Because of this damping, we argue that the equation of motion for $\rho(t)$ should depend upon ρ at the time t, rather than t' as it does in (2.12). We therefore take the following Markoffian equation of motion [28,29] for ρ :

$$\dot{\rho}(t) = -\frac{1}{\hbar^2} \mathrm{Tr}_{\mathrm{ATOMS}} \int_{-\infty}^{t} dt' \left[V(t), \left[V(t'), \rho(t) \otimes \rho^{M}\right]\right] \tag{2.13}$$

We shall use equations of this type in succeeding sections and discuss their validity for the specific problems under consideration. A general derivation of the Markoffian approximation is given in Ref. (28) and (29). We shall solve (2.13) and verify that the characteristic time scale of changes of ρ is much slower than the scale of time changes (lifetimes) of the atomic system provided that the following relation between the coupling strength g and the atomic damping rate γ is satisfied:

$$\frac{g}{\gamma} \ll 1 \tag{2.14}$$

The four terms which come from expanding the double commutator in (2.13) may be written as

$$\dot{\rho}(t) = \sum_{q=1}^{4} C_q \tag{2.15a}$$

where

$$C_1 = -\frac{1}{\hbar^2} \mathrm{Tr}_{ATOMS} \int_{-\infty}^{t} dt' \, V(t) V(t') \rho(t) \otimes \rho^M \tag{2.15b}$$

$$C_2 = \frac{1}{\hbar^2} \mathrm{Tr}_{ATOMS} \int_{-\infty}^{t} dt' \, V(t) \rho(t) \otimes \rho^M V(t') \tag{2.15c}$$

$$C_3 = C_2^* \; ; \quad C_4 = C_1^* \tag{2.15d}$$

The contribution of C_1 is obtained as follows: taking matrix elements and substituting for the V's from (2.5) gives (we take $\Delta\omega \equiv \omega_0 - \omega - \Omega$, $\Delta\omega' \equiv \omega_0 - \omega' - \Omega$)

$$
\begin{aligned}
\left(C_1\right)_{n_0\{n_s\}} &\equiv \langle n_0\{n_s\}|C_1|n_0\{n_s\}\rangle \\
&= -g^2\,\mathrm{Tr}_{\text{ATOMS}}\,\langle n_0\{n_s\}\{\xi_\ell\}|\sum_{j,m=1}^{N}\sum_{\omega,\omega'}\int_{-\infty}^{t}dt'\Big\{a_0 a_\omega^{+}\sigma_j^{+} \\
&\quad e^{i(\vec{k}_0-\vec{k}_\omega)\cdot\vec{r}_j}\,e^{-i\Delta\omega t} + a_0^{+}a_\omega\sigma_j\,e^{-i(\vec{k}_0-\vec{k}_\omega)\cdot\vec{r}_j} \\
&\quad e^{i\Delta\omega t}\Big\}\Big\{a_0 a_{\omega'}^{+}\sigma_m^{+}\,e^{i(\vec{k}_0-\vec{k}_{\omega'})\cdot\vec{r}_m}\,e^{-i\Delta\omega' t'} \\
&\quad + a_0^{+}a_{\omega'}\sigma_m\,e^{-i(\vec{k}_0-\vec{k}_{\omega'})\cdot\vec{r}_m}\,e^{i\Delta\omega' t'}\Big\}\rho(t)\otimes\rho^{M}|n_0\{n_s\}\{\xi_\ell\}\rangle
\end{aligned}
\tag{2.16}
$$

where n_0 refers to the occupation number of the incident beam, $\{n_s\}$ to the scattered beam occupation numbers, and $\{\xi_\ell\}$ specifies a set of states of the N atoms. Since ρ^M is diagonal in the atomic states, equation (2.11), only two of the four possible terms of the above product contribute; also, since ρ^M factors into a product of single atom density matrices, the sums over all atomic variables other than those specified by the indices on the pair of atomic operators within the matrix element contribute a factor of one. Thus,

$$
\left(C_1\right)_{n_0\{n_s\}} = -g^2\int_{-\infty}^{t}dt'\sum_{j=1}^{N}\sum_{\omega,\omega'}\langle n_0\{n_s\}|\Big\{P_{\alpha_j}\,a_0 a_\omega^{+}a_0^{+}a_{\omega'}\,e^{-i(\vec{k}_\omega-\vec{k}_{\omega'})\cdot\vec{r}_j}\,e^{-i\Delta\omega t}\,e^{i\Delta\omega' t'}
$$

$$+\rho_{\beta_j} a_0^{+} a_\omega a_0 a_{\omega'}^{+} e^{i(\vec{K}_\omega - \vec{K}_{\omega'})\cdot\vec{r}_j} e^{i\Delta\omega t} e^{-i\Delta\omega' t'} \Big\} \rho(t) | n_0 \{n_s\} \rangle \qquad (2.17)$$

where

$$\rho_{\alpha_j} = \langle \alpha_j | \rho^{(j)} | \alpha_j \rangle \qquad (2.18)$$

and similarly for ρ_{β_j}. If we assume that these occupation probabilities are the same for all atoms so that they do not depend on the index j, then the sum over j in (2.17) can be done if we assume that the atomic locations $\vec{r}_j$ are fixed to a regular spacial lattice. The sum over j thus contributes delta functions of the difference between the propagation vectors $\vec{K}_\omega$ and $\vec{K}_{\omega'}$. Since we assumed that we are looking at the light scattered into a single direction, and since the magnitudes of the propagation vectors are directly proportional to the frequencies ω and ω', this makes all terms for which $\omega \neq \omega'$ vanish. Hence, doing the sum over ω' gives

$$(C_1)_{n_0\{n_s\}} = -g^2 \int_{-\infty}^{t} dt' \sum_{\omega} \Big\{ \rho_\alpha \, \langle n_0\{n_s\}| a_0 a_\omega^+ a_0^+ a_\omega \, e^{-i\Delta\omega(t-t')} \rho(t)|n_0\{n_s\}\rangle$$

$$+ \rho_\beta \, \langle n_0\{n_s\}| a_0^+ a_\omega a_0 a_\omega^+ \, e^{i\Delta\omega(t-t')} \rho(t)|n_0\{n_s\}\rangle \Big\} \qquad (2.19)$$

$$(C_1)_{n_0\{n_s\}} = -g^2 \sum_{\omega} \Big\{ \rho_\alpha (n_0+1) n_\omega \, \rho^{(t)}_{n_0\{n_1 n_2 \cdots n_\omega \cdots\}} \int_{-\infty}^{t} dt' \, e^{-i\Delta\omega(t-t')} \qquad (2.20)$$

$$+ \rho_\beta n_0 (n_\omega+1) \, \rho^{(t)}_{n_0\{n_1 n_2 \cdots n_\omega \cdots\}} \int_{-\infty}^{t} dt' \, e^{i\Delta\omega(t-t')} \Big\}$$

Taking damping into account by adding a small imaginary part to the atomic separation energy and thus denoting

$$\Delta\omega^{\pm} = \omega_0 - \omega - (\Omega \pm i\gamma) \qquad (2.21)$$

the time integrations in (2.20) may be carried out to yield

$$(C_1)_{n_0\{n_1 n_2 \cdots\}} = -g^2 \sum_{\omega} \left\{ \frac{\rho_\alpha (n_0+1) n_\omega}{i\Delta\omega^+} - \frac{\rho_\beta n_0 (n_\omega+1)}{i\Delta\omega^-} \right\} \rho^{(t)}_{n_0\{n_1 n_2 \cdots n_\omega \cdots\}} \qquad (2.22)$$

The contribution from C_2 , (2.15c), may be calculated in a similar way to obtain

$$(C_2)_{n_0\{n_s\}} = g^2 \sum_{\omega} \left\{ \frac{P_\beta (n_0+1) n_\omega}{i\Delta\omega^+} \rho^{(t)}_{n_0+1\,\{n_1 n_2 \cdots n_\omega-1 \cdots\}} - \frac{P_\alpha n_0 (n_\omega+1)}{i\Delta\omega^-} \rho^{(t)}_{n_0-1\,\{n_1 n_2 \cdots n_\omega+1 \cdots\}} \right\} \qquad (2.23)$$

Thus, using (2.15d), one obtains the following equation of motion for the radiation density matrix

$$\begin{aligned} \dot{\rho}^{(t)}_{n_0\{n_1 n_2 \cdots\}} = \sum_{\omega} \Bigg\{ \frac{2g^2\gamma}{\gamma^2+(\Delta\omega)^2} \Bigg[&-\Big(P_\alpha (n_0+1) n_\omega + P_\beta n_0 (n_\omega+1)\Big) \rho^{(t)}_{n_0\,\{n_1 n_2 \cdots n_\omega \cdots\}} \\ &+ P_\beta (n_0+1) n_\omega \, \rho^{(t)}_{n_0+1\,\{n_1 n_2 \cdots n_\omega-1 \cdots\}} \\ &+ P_\alpha n_0 (n_\omega+1) \, \rho^{(t)}_{n_0-1\,\{n_1 n_2 \cdots n_\omega+1 \cdots\}} \Bigg] \Bigg\} \end{aligned} \qquad (2.24)$$

If we try a solution of the form

$$\rho^{(t)}_{n_0\,\{n_1 n_2 \cdots\}} = \prod_j \rho^{(t)}_{n_0 n_j} \qquad (2.25)$$

then (2.24) yields

$$\sum_{\vec{K}} \dot{\rho}^{(t)}_{n_0 n_K} \prod_{j\neq K} \rho^{(t)}_{n_0 n_j} = \sum_{\omega} \frac{2g^2\gamma}{\gamma^2+(\Delta\omega)^2}\Big\{ A(n_0,n_\omega)\,\rho^{(t)}_{n_0 n_\omega} + B(n_0,n_\omega)\,\rho^{(t)}_{n_0+1,\,n_\omega-1} + C(n_0,n_\omega)\,\rho^{(t)}_{n_0-1,\,n_\omega+1}\Big\}\prod_{j\neq\omega}\rho^{(t)}_{n_0 n_\omega} \qquad (2.26)$$

Since the coefficients A,B, and C only involve n_0 and n_ω, we may identify the indices $K=\omega$ and obtain a solution of (2.24) provided the submatrices satisfy the following equation (the quantities A,B, and C are evident from (2.24):

$$\dot{\rho}^{(t)}_{n_0 n_\omega} = \frac{2g^2\gamma}{\gamma^2+(\Delta\omega)^2}\Big\{ -\Big[\mathcal{P}_\alpha (n_0+1)\, n_\omega + \mathcal{P}_\beta (n_0)(n_\omega+1)\Big]\rho^{(t)}_{n_0 n_\omega} + \mathcal{P}_\beta (n_0+1)\, n_\omega\, \rho^{(t)}_{n_0+1,\,n_\omega-1} + \mathcal{P}_\alpha\, n_0 (n_\omega+1)\, \rho^{(t)}_{n_0-1,\,n_\omega+1}\Big\} \qquad (2.27)$$

If we take $\mathcal{P}_\alpha \simeq 0$, $\mathcal{P}_\beta \simeq 1$ and argue that for spontaneous (non-stimulated) scattering the occupation numbers n_ω are expected to be small, this equation becomes (note that the two terms in (2.27) thus neglected correspond to the rescattering of photons back into the incident beam, a presumably weak effect)

$$\dot{\rho}^{(t)}_{n_0 n_\omega} = \frac{2g^2\gamma}{\gamma^2+(\Delta\omega)^2}\Big[-n_0(n_\omega+1)\,\rho^{(t)}_{n_0 n_\omega} + (n_0+1)n_\omega\,\rho^{(t)}_{n_0+1,\,n_\omega-1}\Big] \tag{2.28}$$

which we may interpret as giving the rate of change of the occupation of the (n_0, n_ω) state of the radiation field due to absorbtion of incident photons and emission of scattered photons. We drop the "stimulated" terms in (2.28) by setting $n_\omega+1 \approx 1$ in the first term and $(n_\omega-1)+1 \approx 1$ in the second. This gives

$$\dot{\rho}^{(t)}_{n_0 n_\omega} = \frac{2g^2\gamma}{\gamma^2+(\Delta\omega)^2}\Big[-n_0\,\rho^{(t)}_{n_0 n_\omega} + (n_0+1)\,\rho^{(t)}_{n_0+1,\,n_\omega-1}\Big] \tag{2.29}$$

subject to the initial time boundary condition

$$\rho^{(0)}_{n_0 n_\omega} = \delta_{n_\omega,0}\,\rho^{L\,(0)}_{n_0 n_0} \tag{2.30}$$

where $\rho^{L\,(0)}_{n_0 n_0}$ is the incident beam density matrix. This is a special case of the more general initial value boundary condition

$$\rho^{(0)}_{n_0 n_\omega} = \rho^{S\,(0)}_{n_\omega n_\omega}\,\rho^{L\,(0)}_{n_0 n_0} \tag{2.31}$$

In (2.30) we are therefore assuming that the initial value of the scattered field density matrix consists of unity for the vacuum state and zero for all other occupation numbers. Since (2.29) couples density matrix elements for which $n_0 + n_\omega$ is constant, we may define a new variable N by

$$n_0 + n_\omega = N \tag{2.32}$$

Thus we define

$$\rho^{(N)}_{n_\omega} \equiv \rho_{N-n_\omega,\, n_\omega} = \rho_{n_0,\, n_\omega} \tag{2.33}$$

so that, from (2.29), $\rho^{(N)}_{n_\omega}$ satisfies the equation

$$\dot{\rho}^{(N)}_{n_\omega} = \Gamma\left[-(N-n_\omega)\,\rho^{(N)}_{n_\omega} + \big(N-(n_\omega-1)\big)\,\rho^{(N)}_{n_\omega-1}\right] \tag{2.34}$$

Before proceeding to solve this equation, we note that we may recover the quantity desired by

$$\rho_{n_0 n_\omega} = \sum_N \rho^{(N)}_{n_\omega}\, \delta_{N-n_\omega,\, n_0} \tag{2.35}$$

so that the density matrix for the radiation field alone is

$$\rho_{n_\omega} = \sum_{n_0} \rho_{n_0 n_\omega} = \sum_{n_0} \sum_N \rho^{(N)}_{n_\omega}\, \delta_{N-n_\omega,\, n_0} = \sum_{n_0} \rho^{(n_0)}_{n_\omega} \tag{2.36}$$

Now, equation (2.34) is a special case of a more general equation solved in 1957 in an early paper on photon noise in quantum amplifiers [30] by Shimoda, Takahasi, and Townes. They obtained the solution of the equation

$$\frac{d\rho_{n,m}^{(t)}}{dt} = -\left[(a+b)m + c\right]\rho_{n,m}^{(t)} + \left[a(m-1) + c\right]\rho_{n,m-1} + b(m+1)\rho_{n,m+1} \tag{2.37}$$

with the boundary condition that at t=0, $\rho_{n,m}^{(0)} = \delta_{n,m}$ (the index n is given significance only through this boundary condition) in the form of a generating function defined by

$$f_n = \sum_m \rho_{n,m}\, x^m \tag{2.38}$$

Their solution for f_n is

$$f_n = \left[\frac{1}{1+G(1-x)}\right]^{\frac{c}{a}} \left[\frac{1+(G-K)(1-x)}{1+G(1-x)}\right]^n \tag{2.39}$$

where (assuming a,b, and c in (2.37) are independent of time)

$$K = e^{(a-b)t} \; ; \quad G = \frac{a}{a-b}(K-1) \tag{2.40}$$

We note that should we wish to solve (2.37) subject to the more general boundary condition

$$\rho_{n,m}^{(0)} = Q(n)\,\delta_{n,m} \tag{2.41}$$

where Q is some function of n , the appropriate generating function would be

$$F_n = Q(n)\, f_n \qquad (2.42)$$

since this reduces to the correct term at t=0. Generalizing further to include the boundary condition

$$P_m(0) = Q(m) \qquad (2.43)$$

that is, there is a full initial distribution, the appropriate generating function would be

$$F = \sum_n Q(n)\, f_n \qquad (2.44)$$

If we make the identifications via comparison of (2.34) and (2.37)

$$a = -\Gamma \; ; \quad b = 0 \; ; \quad c = \Gamma N \; ; \quad \frac{c}{a} = -N \qquad (2.41)$$

and note that our boundary condition (2.30) implies that the correct generating function for (2.34) is given by (2.42) with $n = 0$:

$$\mathcal{F} = \sum_{n_\omega} P^{(N)}_{n_\omega}\, x^{n_\omega} \qquad (2.42)$$

$$= \rho^{L}_{NN}(0)\, f_0 \qquad (2.43)$$

However, let us define a generating function for the radiation density matrix ρ_{n_ω}:

$$\mathcal{G} = \sum_{n_\omega} \rho_{n_\omega} x^{n_\omega} \tag{2.44a}$$

$$= \sum_{n_\omega} x^{n_\omega} \sum_{N} \rho^{(N)}_{n_\omega} \tag{2.44b}$$

where the second line of (2.44) follows from (2.35). We thus have

$$\mathcal{G} = \sum_{N} \Big(\sum_{n_\omega} x^{n_\omega} \rho^{(N)}_{n_\omega} \Big) = \sum_{N} f_0 \, \rho^{L}_{NN}(0) \tag{2.45}$$

by (2.43). However, from the constants in (2.41) and (2.39) we find that

$$f_0 = \left[1 + G(1-x) \right]^{N} = (1-K)^{N} \left[\frac{K}{1-K} + x \right]^{N} \tag{2.46}$$

Using the binomial expansion theorem this becomes

$$f_0 = (1-K)^{N} \sum_{n_\omega = 0}^{N} \binom{N}{n_\omega} \left(\frac{K}{1-K} \right)^{N-n_\omega} x^{n_\omega}$$

We may formally extend the sum up to infinity since the binomial coefficients for $n_\omega > N$ vanish identically. Thus, the generating function we want becomes, from (2.45),

$$\mathcal{G} = \sum_{n_\omega = 0}^{\infty} x^{n_\omega} \sum_{N=0}^{\infty} \binom{N}{n_\omega} (1-K)^{N} \left(\frac{K}{1-K} \right)^{N-n_\omega} \rho^{L}_{NN}(0) \tag{2.47}$$

Comparing (2.47) with the definition (2.44a) we can immediately read off the scattered field density matrix:

$$\rho_{n_\omega}(t) = \sum_{N=0}^{\infty} \binom{N}{n_\omega} \left(1 - e^{-\Gamma t}\right)^{n_\omega} \left(e^{-\Gamma t}\right)^{N-n_\omega} \rho^{L}_{NN}(0) \tag{2.48}$$

This is then the complete solution obtained from (2.29) and the boundary condition (2.30).

We note that the maximum value of Γ is achieved for $\Delta\omega = 0 : \Gamma_{max} = 2g^2/\gamma$. Therefore, while the atoms in the system evolve according to factors like $e^{-\gamma t}$, the density matrix evolution involves $exp\,[-2\,\frac{g^2}{\gamma}\,t] = exp\left[-2\left(\frac{g}{\gamma}\right)^2 \gamma t\right]$. Provided that the relation (2.14) is obeyed, we see that the density matrix evolves at a much slower rate than the atoms, thus justifying our original Markoffian approximation for the density operator equation of motion.

The solution has the characteristic property that if the incident beam density matrix $\rho^{L}_{NN}(0)$ is either Gaussian (thermal) Poisson (coherent), then the scattered field density matrix will also be Gaussian or Poisson respectively.

If the incident beam is Poisson:

$$\rho^{L}_{n_0 n_0}(0) = \frac{(\bar{n}_0)^{n_0}\, e^{-\bar{n}_0}}{(n_0)!} \tag{2.49}$$

Then (taking $\sigma = 1 - e^{-\Gamma t}$)

$$\rho_{n_\omega}(t) = \sum_{n_0=0}^{\infty} \binom{n_0}{n_\omega} \sigma^{n_\omega} (1-\sigma)^{n_0 - n_\omega} \frac{(\bar{n}_0)^{n_0}\, e^{-\bar{n}_0}}{(n_0)!}$$

$$\rho_{n_\omega}(t) = \frac{\sigma^{n_\omega}(1-\sigma)^{-n_\omega-\overline{n}_0}}{(n_\omega)!} \sum_{n_0=0}^{\infty} \frac{\left[\overline{n}_0(1-\sigma)\right]^{n_0}}{(n_0-n_\omega)!}$$

$$= \frac{\sigma^{n_\omega}(1-\sigma)^{-n_\omega} e^{-\overline{n}_0}}{(n_\omega)!} \sum_{m=0}^{\infty} \frac{\left[\overline{n}_0(1-\sigma)\right]^{m+n_0}}{(m)!} \qquad (2.50)$$

$$= \frac{\sigma^{n_\omega}(1-\sigma)^{-n_\omega-\overline{n}_0}}{(n_\omega)!} e^{-\overline{n}_0} \, e^{\overline{n}_0(1-\sigma)} \left[\overline{n}_0(1-\sigma)\right]^{n_\omega}$$

$$= \frac{(\overline{n}_0\sigma)^{n_\omega} e^{-\overline{n}_0\sigma}}{(n_\omega)!}$$

which is again Poisson with mean photon number $\overline{n}_0 \sigma$.

If the incident beam is thermal, characterized by temperature T:

$$\rho^{L}_{n_0 n_0}(0) = x^{n_0}(1-x) \qquad (2.51)$$

where

$$x = e^{-\frac{\hbar\omega_0}{k_B T}} \qquad (2.52)$$

since

$$\overline{n}_0 = \sum_{n_0=0}^{\infty} n_0 x^{n_0}(1-x) = \frac{x}{1-x} \qquad (2.53)$$

we can also write the distribution as

$$\rho^{L}_{n_0 n_0}(0) = \frac{(\overline{n}_0)^{n_0}}{\left[1+\overline{n}_0\right]^{1+n_0}} \qquad (2.54)$$

Then:

$$\rho_{n_\omega}(t) = \sum_{n_0=0}^{\infty} \binom{n_0}{n_\omega} \sigma^{n_\omega} (1-\sigma)^{n_0-n_\omega} x^{n_0} (1-x)$$

$$= \frac{(-)^{n_\omega}}{n_\omega !} \left(\frac{d}{d\lambda}\right)^{n_\omega} \left\{ \sum_{n_0=0}^{\infty} (1-\lambda\sigma)^{n_0} x^{n_0} (1-x) \right\} \Bigg|_{\lambda=1} \tag{2.55}$$

$$= \frac{(-)^{n_\omega}}{n_\omega !} \left(\frac{d}{d\lambda}\right)^{n_\omega} \left\{ \frac{1-x}{1-(1-\lambda\sigma)x} \right\} \Bigg|_{\lambda=1} = \frac{(-)^{n_\omega}}{n_\omega !} \left[\frac{n_\omega ! (-)^{n_\omega} x^{n_\omega} \sigma^{n_\omega}}{[1-(1-\lambda\sigma)x]^{n_\omega+1}} \right] (1-x) \Bigg\} \Bigg|_{\lambda=1}$$

$$= \frac{(1-x)(x\sigma)^{n_\omega}}{(1-x+x\sigma)^{n_\omega+1}} = \left(\frac{x}{1-x}\sigma\right)^{n_\omega} \left[1 + \frac{x}{1-x}\sigma\right]^{-n_\omega-1}$$

which is again a thermal distribution of mean value

$$\bar{n}_\omega = \frac{x}{1-x}\sigma = \bar{n}_0 \sigma \tag{2.56}$$

We have thus obtained the statistical properties of the scattered radiation by computing the density matrix of the field. In addition to the properties mentioned above, we note that since the scattered field density matrix obeys the diagonality and factorization conditions (1.11) and (1.12), the field correlation functions are expected to obey the Gaussian factorization relations (1.13) and (1.14).

Part A. Section III. Scattering by Density Fluctuations

The use of light scattering techniques to study the properties of many particle systems has a long history [3,13,26]. In particular, light is a particularly useful method for probing density fluctuations of such systems. The significance of scattering experiments in measuring fundamental statistical mechanical properties of many particle systems was demonstrated, in the case of neutron scattering, by Van Hove [11]. His analysis was extended to light scattering by Komarov and Fisher [12].

In this section we shall study the effects of the statistical properties of density fluctuations upon the statistics of the scattered light by considering a model of the scattering process. Since we shall be concerned primarily with the effects induced by the properties of the scattering medium, we take the incident field to be a classical monochromatic light wave of frequency ω_0, amplitude E_0, and propagation vector $\vec{K}_0$. If such a light wave is incident upon a homogeneous, isotropic scattering medium, a scattered wave $\vec{E}_{\vec{K}}(\vec{R},t)$ is produced by the dielectric constant fluctuations of the medium [13]:

$$\vec{E}_{\vec{K}}(\vec{R},t) = -E_0 \left(\frac{\omega_0}{c}\right)^2 \frac{e^{i(\vec{K}\cdot\vec{R}-\omega_0 t)}}{4\pi R} \sin\varphi \, (2\pi)^{3/2} \, \delta\epsilon(\vec{q},t)\,\hat{\varphi}$$

$$= \mathcal{E}\, e^{i(\vec{K}\cdot\vec{R}-\omega_0 t)} \, \delta\epsilon(\vec{q},t)\,\hat{\varphi} \qquad (3.1)$$

Here φ is the angle between the direction of polarization of the (plane polarized) incident light and the propagation vector $\vec{K}$ of the scattered light, and $\delta\epsilon(\vec{q},t)$ is the q^{th} spacial Fourier component of the fluctuating part of the dielectric constant ($\epsilon = \langle\epsilon\rangle + \delta\epsilon(\vec{r},t)$; since, for simplicity, we are considering an isotropic medium, we take ϵ to be a scalar),where

$$\vec{K} = \vec{K}_o + \vec{q} \qquad (3.2)$$

For a one component homogeneous isotropic medium in thermal equilibrium, the density ρ and temperature T are appropriate thermodynamic variables. We may assume that the local fluctuation of the dielectric constant at some point $\vec{r}$ in the medium, $\delta\epsilon(\vec{r},t)$, is related to the local fluctuation of the density $\delta\rho(\vec{r},t)$ and the temperature $\delta T(\vec{r},t)$ at the same point:

$$\delta\epsilon(\vec{r},t) = \left(\frac{\partial\epsilon}{\partial\rho}\right)_T \delta\rho(\vec{r},t) + \left(\frac{\partial\epsilon}{\partial T}\right)_\rho \delta T(\vec{r},t) \qquad (3.3)$$

Neglecting the second term, which is frequently much smaller than the first due to the small thermodynamic derivative, we thus see from (3.1) that the scattered field $E_{\vec{K}}(\vec{R},t)$ is proportional to the q^{th} Fourier component of the density fluctuation. Therefore, in view of the form of the incident field assumed, the first and second order correlation functions of the scattered field will be proportional to the corresponding correlation functions of the density fluctuation:

$$G^{(1)}(t,0) = \left\langle E^{(-)}_{\vec{K}}(\vec{R},t)\, E^{(+)}_{\vec{K}}(\vec{R},0) \right\rangle \tag{3.4a}$$

$$= \mathcal{E}^{2} e^{i\omega_0 t} \left\langle \delta\rho^{(-)}(\vec{q},t)\, \delta\rho^{(+)}(\vec{q},t) \right\rangle \tag{3.4b}$$

$$G^{(2)}(0,t;t,0) = \left\langle E^{(-)}_{\vec{K}}(\vec{R},0)\, E^{(-)}_{\vec{K}}(\vec{R},t)\, E^{(+)}_{\vec{K}}(\vec{R},t)\, E^{(+)}_{\vec{K}}(\vec{R},0) \right\rangle \tag{3.5a}$$

$$= \mathcal{E}^{4} \left\langle \delta\rho^{(-)}(\vec{q},0)\, \delta\rho^{(-)}(\vec{q},t)\, \delta\rho^{(+)}(\vec{q},t)\, \delta\rho^{(+)}(\vec{q},0) \right\rangle \tag{3.5b}$$

The correlation functions are ensemble averages of positive ($\propto e^{-i\omega t}$) and negative frequency quantities; the density fluctuation averages are to be taken with respect to a thermal equilibrium ensemble.

We now take the following model for the density fluctuation $\delta\rho(\vec{q},t)$: we regard it as a quantum mechanical (one dimensional) linear harmonic oscillator coupled to a thermal reservoir consisting of a canonical ensemble of (noninteracting) oscillators at a temperature $T = (K_B \beta)^{-1}$. Hence, we identify the density fluctuation $\delta\rho^{(+)}(\vec{q},t)$ with the second quantized (boson) annihilation operator $a(t)$ and the fluctuation $\delta\rho^{(-)}(\vec{q},t)$ with the creation operator $a^{+}(t)$. The Hamiltonian for the entire system of interest - single oscillator mode ("system") plus reservoir plus interaction -

is taken to be

$$\mathcal{H} = H + R + V \tag{3.6}$$

$$H = \hbar \Omega a^+ a \tag{3.7}$$

$$R = \sum_{j=1}^{\infty} \hbar \omega_j b_j^+ b_j \tag{3.8}$$

$$V = \hbar \sum_{j=1}^{\infty} (\kappa_j b_j a^+ + \kappa_j^* b_j^+ a) \tag{3.9}$$

where κ_j, κ_j^* are c-number coupling constants.

The equation of motion of the density operator ρ of the entire system, in the Schrödinger picture, is

$$\frac{\partial \rho(t)}{\partial t} = \dot{\rho}(t) = \frac{1}{i\hbar} \left[H + R + V, \ \rho(t) \right] \tag{3.10}$$

The density operator in the interaction picture, $\chi(t)$, is given by

$$\rho(t) = \exp\left[-\frac{i}{\hbar} (H+R) t \right] \chi(t) \exp\left[\frac{i}{\hbar} (H+R) t \right] \tag{3.11}$$

and satisfies

$$\dot{\chi}(t) = \frac{1}{i\hbar} \left[V(t), \ \chi(t) \right] \tag{3.12}$$

where the interaction Hamiltonian in the interaction picture is given by

$$V(t) = \exp\left[\frac{i}{\hbar}(H+R)t\right] V \exp\left[-\frac{i}{\hbar}(H+R)t\right] \tag{3.13}$$

Thus

$$\begin{aligned} \mathrm{Tr}_R\, \rho(t) \equiv S(t) &= \mathrm{Tr}_R\, e^{-\frac{i}{\hbar}(H+R)t}\, \chi(t)\, e^{\frac{i}{\hbar}(H+R)t} \\ &= e^{-\frac{i}{\hbar}Ht}\, s(t)\, e^{\frac{i}{\hbar}Ht} \end{aligned} \tag{3.14}$$

where

$$s(t) \equiv \mathrm{Tr}_R\, \chi(t) \tag{3.15}$$

Assuming the interaction is turned on at t=0, we take

$$\rho(0) = S(0)\, f_0(R) = s(0)\, f_0(R) = \chi(0) \tag{3.16}$$

with

$$f_0(R) = e^{-\beta R}\left[\mathrm{Tr}_R\, e^{-\beta R}\right]^{-1} \tag{3.17}$$

The differential equation (3.12) may be formally integrated to give

$$\chi(t) = \chi(0) + \frac{1}{i\hbar}\int_0^t dt' \left[V(t'), \chi(t')\right] \tag{3.18}$$

Inserting this into the right side of (3.12) we have

$$\dot{\chi}(t) = \frac{1}{i\hbar}\left[V(t), \chi(0)\right] - \frac{1}{\hbar^2}\int_0^t dt'\left[V(t), \left[V(t'), \chi(t')\right]\right] \tag{3.19}$$

We look for a solution of the form

$$\chi(t) = s(t) f_0(R) + \Delta\chi(t) \tag{3.20}$$

where $\Delta\chi(t)$ must evidently satisfy

$$\mathrm{Tr}_R\, \Delta\chi(t) = 0$$

Since for $V = 0$ the systems are uncoupled and $\Delta\chi = 0$, this correction term must be at least of some order in V higher than the zero th. If we assume that the interaction is weak enough to be treated to second order, then this term will represent a higher order effect in (3.19) which we may neglect. Thus, to second order, we have (tracing over the reservoir in (3.19)) the following equation for the reduced density operator of the system, $s(t)$:

$$\dot{s}(t) = \frac{1}{i\hbar}\mathrm{Tr}_R\left[V(t), s(0) f_0(R)\right] - \frac{1}{\hbar^2}\int_0^t dt'\, \mathrm{Tr}_R\left[V(t), \left[V(t'), s(t') f_0(R)\right]\right] \tag{3.21}$$

Following Louisell [16,31] we develop this equation for the particular model we have chosen.

In view of our choice of Hamiltonians (3.7), (3.8), and (3.9) for our model, (3.13) yields the following expression for the interaction $V(t)$:

$$V(t) = e^{\frac{i}{\hbar}(H+R)t}\Big[\hbar\sum_j(\kappa_j b_j a^+ + \kappa_j^* b_j^+ a)\Big]e^{-\frac{i}{\hbar}(H+R)t}$$

$$\begin{aligned} &= \hbar\sum_j\Big[\kappa_j b_j(0)e^{-i\omega_j t}a^+(0)e^{i\Omega t} \\ &\qquad + \kappa_j^* b_j^+(0)e^{i\omega_j t}a(0)e^{-i\Omega t}\Big] \end{aligned} \tag{3.22}$$

$$\equiv i\hbar\left[F(t)a^+(0) - F^+(t)a(0)\right]$$

In view of (3.17), which is diagonal in the reservoir states, we see that the following correlation functions of the "forces" $F(t)$ and $F^+(t)$ vanish:

$$\langle F(t_1)F(t_2)\rangle = Tr_R\left[f_0(R)F(t_1)F(t_2)\right] = 0 \tag{3.23}$$

$$\langle F^+(t_1)F^+(t_2)\rangle = Tr_R\left[f_0(R)F^+(t_1)F^+(t_2)\right] = 0 \tag{3.24}$$

However

$$\langle F^+(t_1)F(t_2)\rangle = \sum_{j,k}\kappa_j^*\kappa_k\langle b_j^+(0)b_k(0)\rangle e^{i[(\omega_j-\Omega)t_1-(\omega_k-\Omega)t_2]} \tag{3.25}$$

does not vanish since

$$\langle b_j^+(0)\, b_K(0) \rangle = \bar{n}_j\, \delta_{jK} \tag{3.26}$$

where $\bar{n}_j$ is the mean occupation number of mode j. Thus

$$\langle F^+(t_1) F(t_2) \rangle = \sum_j' |V_j|^2\, \bar{n}_j\, e^{i(\omega_j - \Omega)(t_1 - t_2)} \tag{3.27}$$

and similarly

$$\langle F(t_1) F^+(t_2) \rangle = \sum_j' |V_j|^2 (1 + \bar{n}_j)\, e^{-i(\omega_j - \Omega)(t_1 - t_2)} \tag{3.28}$$

since

$$\langle b_j(0)\, b_K^+(0) \rangle = (1 + \bar{n}_j)\, \delta_{jK} \tag{3.29}$$

As $|t_1 - t_2| \to \infty$, both of these correlation functions must vanish. Let τ_c be the time over which these functions have a significant non-zero value. Then, integrating with respect to t_1 and t_2 over an interval Δt much larger than τ_c gives:

$$I = \int_t^{t+\Delta t} dt_1 \int_t^{t+\Delta t} dt_2 \sum_j' |V_j|^2\, \bar{n}_j\, e^{i(\omega_j - \Omega)(t_1 - t_2)}$$

$$I = \sum_j |M_j|^2 \bar{n}_j \left[\frac{e^{i(\omega_j - \Omega)(t+\Delta t)} - e^{i(\omega_j-\Omega)t}}{i(\omega_j - \Omega)} \right] \left[\frac{e^{-i(\omega_j-\Omega)(t+\Delta t)} - e^{-i(\omega_j-\Omega)t}}{-i(\omega_j - \Omega)} \right]$$

$$= \sum_j |M_j|^2 \bar{n}_j \left[\frac{1 + e^{i(\omega_j-\Omega)\Delta t} - e^{-i(\omega_j-\Omega)\Delta t} + 1}{(\omega_j - \Omega)^2} \right]$$

$$= \sum_j |M_j|^2 \bar{n}_j \frac{2\left[1 - \cos[(\omega_j - \Omega)\Delta t]\right]}{(\omega_j - \Omega)^2}$$

$$= \sum_j |M_j|^2 \bar{n}_j \frac{4 \sin^2\left[\frac{(\omega_j-\Omega)\Delta t}{2}\right]}{(\omega_j - \Omega)^2}$$

Thus:

$$\frac{1}{\Delta t} I = \Delta t \sum_j |M_j|^2 \bar{n}_j \frac{4 \sin^2\left[\frac{(\omega_j-\Omega)\Delta t}{2}\right]}{(\omega_j - \Omega)^2 (\Delta t)^2}$$

$$\simeq \int_0^\infty d\omega_j \, g(\omega_j) |M_j(\omega_j)|^2 \bar{n}(\omega_j) \frac{4 \sin^2\left[\frac{(\omega_j-\Omega)\Delta t}{2}\right]}{(\omega_j - \Omega)^2 (\Delta t)^2}$$

Where $g(\omega_j)$ is the density of reservoir states per unit frequency at frequency ω_j. If $\Delta t \gg \tau_c$, the integral is strongly peaked at $\omega_j \simeq \Omega$. Thus

$$\frac{1}{\Delta t} I \cong \frac{\gamma}{2\pi} \bar{n}(\Omega) \frac{2}{\Delta t} \int_{-\infty}^{\infty} \frac{\sin^2(\Delta t \cdot x)}{x^2} \, dx$$

$$= \frac{\gamma}{2\pi} \bar{n}(\Omega) \frac{2}{\Delta t} \pi \Delta t = \gamma \bar{n}(\Omega)$$

where

$$\gamma = 2\pi g(\Omega) |V(\Omega)|^2 \tag{3.30}$$

Therefore, without loss of generality we may take

$$\langle F^{+}(t_1) F(t_2) \rangle = \gamma \bar{n}(\Omega) \delta(t_1 - t_2) \tag{3.31}$$

and similarly

$$\langle F(t_1) F^{+}(t_2) \rangle = \gamma \left(1 + \bar{n}(\Omega)\right) \delta(t_1 - t_2) \tag{3.32}$$

Returning to the equation of motion (3.21) and using the interaction in the form given by (3.22) along with the reservoir correlation function results (3.23), (3.24), (3.31), and (3.33), we find that the reduced system density operator $s(t)$ satisfies the equation

$$\dot{s}(t) = \frac{\gamma}{2}\left[2a(0)s(t)a^{+}(0) - a^{+}(0)a(0)s(t) - s(t)a^{+}(0)a(0)\right]$$

$$+ \gamma\bar{n}(\Omega)\left[a^{+}(0)s(0)a(0) + a(0)s(t)a^{+}(0)\right. \tag{3.33}$$

$$\left. - a^{+}(0)a(0)s(t) - s(t)a(0)a^{+}(0)\right]$$

Note that the first term of (3.21) makes no contribution in view of the form of $V(t)$.

In order to compute two-time correlation functions from the solution of this equation, we use the regression theorem for Markoff processes [32,7,8,31,33]. This theorem, which is a consequence of writing the total density operator $\chi(t)$ in the product form of equation (3.20) (neglecting the $\Delta\chi(t)$ term), enables one to calculate two time correlation functions if one can find the single time evolution of the average value of some system operator. It is discussed in more detail in Appendix II. We use the regression theorem here in the following way: suppose that the solution $s(t)$ to the above equation of motion is known in the number (n) representation of oscillator states:

$$s^{(t)}_{n,n'} = \sum_{K,K'} f(n,n';K,K';t)\, s^{(0)}_{K,K'} \tag{3.34}$$

The mean value of an operator O will thus be given by (O is an operator which acts on the system variables)

$$\langle O(t)\rangle = \mathrm{Tr}_{S,R}\left[O\,\rho(t)\right]$$

$$= \mathrm{Tr}_{S'}\left\{O\,\mathrm{Tr}_R\left[\rho(t)\right]\right\} \tag{3.35}$$

$$= \mathrm{Tr}_{S'}\left\{O\,\mathrm{Tr}_R\left[e^{-\frac{i}{\hbar}(H+R)t}\,\chi(t)\,e^{\frac{i}{\hbar}(H+R)t}\right]\right\}$$

$$= \mathrm{Tr}_{S'}\left\{O\,e^{-\frac{i}{\hbar}Ht}\left[\mathrm{Tr}_R\,\chi(t)\right]e^{\frac{i}{\hbar}Ht}\right\}$$

$$= \mathrm{Tr}_{S'}\left\{O\,e^{-\frac{i}{\hbar}Ht}\left[\mathrm{Tr}_R\,e^{-\frac{i}{\hbar}Rt}\chi(t)e^{\frac{i}{\hbar}Rt}\right]e^{\frac{i}{\hbar}Ht}\right\} \tag{3.36}$$

$$= \mathrm{Tr}_{S'}\left\{O\,e^{-\frac{i}{\hbar}Ht}\,s(t)\,e^{\frac{i}{\hbar}Ht}\right\}$$

$$= \mathrm{Tr}_{S'}\left\{\left(e^{\frac{i}{\hbar}Ht}\,O\,e^{-\frac{i}{\hbar}Ht}\right)s(t)\right\}$$

$$\equiv \mathrm{Tr}_{S'}\left\{O(t)\,s(t)\right\}$$

The time dependence of $O(t)$ is the free ($V \equiv 0$) time dependence--in what follows this function of time will be understood to have been factored out of the $\mathrm{Tr}_{S'}$. Hence, we have

$$\langle O(t)\rangle = \sum_{n,n'} \sigma^{(t)}_{nn'}\, O_{n'n} = \sum_{n,n',\kappa,\kappa'} O_{n'n}\, f(n,n';\kappa,\kappa';t)\, \sigma^{(0)}_{\kappa\kappa'} \tag{3.37}$$

We now assume that the following expansion can be made:

$$\langle O(t)\rangle = \sum_{n,n',\kappa,\kappa'} \sum_{ij} O_{n'n}\, f^{ij}(n,n';t)\, (\kappa)^{i} (\kappa')^{j}\, \sigma^{(0)}_{\kappa\kappa'} \tag{3.38}$$

$$= \sum_{ij} \Big[\sum_{n,n'} O_{n'n}\, f^{ij}(n,n';t) \Big] \langle Q^{ij}\rangle \tag{3.39}$$

where

$$\langle Q^{ij}\rangle \equiv \sum_{\kappa,\kappa'} Q^{ij}_{\kappa'\kappa}\, \sigma^{(0)}_{\kappa\kappa'}\,;\ Q^{ij}_{\kappa'\kappa} = (\kappa)^{i} (\kappa')^{j} \tag{3.40}$$

The regression theorem then states that the correlation function $\langle O(t)\, \rho(0)\rangle$ is given by

$$\langle O(t)\, \rho(0)\rangle = \sum_{ij} \Big[\sum_{n,n'} O_{n'n}\, f^{ij}(n,n';t) \Big] \langle Q^{ij} \rho\rangle \tag{3.41}$$

where

$$\langle Q^{ij} \rho\rangle = \sum_{\kappa,\kappa',\kappa''} \sigma^{(0)}_{\kappa\kappa'}\, Q^{ij}_{\kappa'\kappa''}\, \rho^{(0)}_{\kappa''\kappa} \tag{3.42}$$

$$= \sum_{\kappa,\kappa',\kappa''} \sigma^{(0)}_{\kappa\kappa'}\, (\kappa')^{j} (\kappa'')^{i}\, \rho^{(0)}_{\kappa''\kappa}$$

Hence:

$$\langle O(t)\rho(0)\rangle = \sum_{\substack{ij\\ n,n'}} \sum_{K,K',K''} O_{n'n}\, f^{ij}_{(n,n';t)}\,(K')^{j}(K'')^{i}\, s^{(0)}_{KK'}\,\rho^{(0)}_{K''K} \tag{3.43}$$

$$= \sum_{\substack{n,n'\\ K,K',K''}} O_{n'n}\, f(n,n';K'',K',t)\, s^{(0)}_{KK'}\,\rho^{(0)}_{K''K} \tag{3.44}$$

$$= \sum_{\substack{n,n'\\ K',K''}} O_{n'n}\, f(n,n';K'',K',t)\,\tilde{s}^{(0)}_{K''K'} \tag{3.45}$$

where

$$\tilde{s}^{(0)}_{K''K'} \equiv \sum_{K} \rho^{(0)}_{K''K}\, s^{(0)}_{KK'} \tag{3.46}$$

Therefore, the regression theorem states that the correlation function $\langle O(t)\rho(0)\rangle$ is obtained by computing just $\langle O(t)\rangle$ assuming that the initial density matrix is $\tilde{s}^{(0)}_{K'',K'}$ rather than $s^{(0)}_{K'',K'}$. This result may be used to reduce the calculation in the following way. Suppose that the generating function of the initial distribution $s^{(0)}_{K,K'}$ is known:

$$F(x,y,0) = \sum_{K,K'} s^{(0)}_{KK'}\, x^{K}\, y^{K'} \tag{3.47}$$

Solution of the equation of motion of $s(t)$ implies that one can find an operator $\mathcal{O}(x,y,t)$ which, when acting on the initial generating function, yields the generating function for the time-evolved density matrix elements $\rho_{KK'}^{(t)}$:

$$F(x,y,t) = \mathcal{O}(x,y,t)\,F(x,y,0) \tag{3.48}$$

$$F(x,y,t) = \sum_{K,K'} \rho_{KK'}^{(t)}\, x^K y^{K'} \tag{3.49}$$

Presumably the average $\langle O(t)\rangle$ can be obtained in a simple way (by differentiation) from $F(x,y,t)$. The regression theorem then indicates that $\langle O(t)\,P(0)\rangle$ can be obtained by carrying out the same operations on $\tilde{F}(x,y,t)$, where

$$\tilde{F}(x,y,t) = \mathcal{O}(x,y,t)\,\tilde{F}(x,y,0) \tag{3.50}$$

$$\tilde{F}(x,y,0) = \sum_{K,K'} \tilde{\rho}_{KK'}(0)\; x^K y^{K'} \tag{3.51}$$

It is to be noted that this method relies on knowledge of the solution for $\rho(t)$ for arbitrary initial distributions.

The first case of interest with regard to the density correlation functions we wish to obtain involves the calculation of $\langle a^+(t)\rangle$. It can be seen that $\langle a^+(t)\rangle$ depends on the $\rho_{n,n+1}^{(t)}$ matrix elements of the system density operator. Hence, we shall first solve the equation of motion for them. Taking the (n, $n+1$) matrix elements of the above equation for s(t) yields the following equation:

$$\frac{\partial}{\partial t} s_{n,n+1}^{(t)} = \gamma(1+\bar{n})\sqrt{(n+1)(n+2)}\, s_{n+1,n+2}^{(t)} - \gamma\left[(n+1)(2\bar{n}+1) - \frac{1}{2}\right] s_{n,n+1}^{(t)} \tag{3.52}$$

$$+ \gamma\bar{n}\sqrt{n(n+1)}\; s_{n-1,n}^{(t)}$$

It can be seen that the equation couples density matrix elements along a stripe parallel to the main diagonal. Hence, it is convenient to define

$$S_{n+1}^{(t)} \equiv s_{n,n+1}^{(t)} \tag{3.53}$$

and introduce the generating function f(x,t):

$$f_{(m+1)}(x,t) = \sum_{n=0}^{\infty} S_{n+1}^{(t)} (n+1)^{-\frac{1}{2}} x^{n+1} \tag{3.54}$$

The subscript refers to the boundary condition:

$$S_{n+1}(0) = \delta_{m+1,\,n+1} \Rightarrow f_{(m+1)}(x,0) = \frac{x^{m+1}}{\sqrt{m+1}} \tag{3.55}$$

Using the following relations (we omit the subscript (m+1) on f until further notice)

$$\sum_{n=0}^{\infty} \sqrt{n+2}\, S_{n+2}\, x^{n+2} \cong \frac{\partial f}{\partial x} \tag{3.56}$$

Proof: $f = \sum_{0}^{\infty} S_{n+1} \frac{x^{n+1}}{\sqrt{n+1}} = \sum_{-1}^{\infty} S_{n+2} \frac{x^{n+2}}{\sqrt{n+2}}$; $\frac{\partial f}{\partial x} = \sum_{-1}^{\infty} S_{n+2} \sqrt{n+2}\, x^{n+1} \simeq \sum_{0}^{\infty} \sqrt{n+2}\, S_{n+2} x^{n+1}$

$$\sum_{0}^{\infty} \sqrt{n+1}\, S_{n+1}\, x^{n+1} = x \frac{\partial}{\partial x} \sum_{0}^{\infty} S_{n+1} \frac{x^{n+1}}{\sqrt{n+1}} = x \frac{\partial f}{\partial x} \qquad (3.57)$$

$$\sum_{0}^{\infty} S_n \sqrt{n}\, x^{n+1} = x^2 \frac{\partial}{\partial x} \sum_{0}^{\infty} S_{n+1} \frac{x^{n+1}}{\sqrt{n+1}} = x^2 \frac{\partial f}{\partial x} \qquad (3.58)$$

and multiplying the equation for $S^{(t)}_{n,n+1}$ by $x^{n+1}(n+1)^{-\frac{1}{2}}$ and summing on n gives the following equation for the generating function:

$$\frac{\partial f(x,t)}{\partial t} = \gamma(1+\bar{n}) \frac{\partial f}{\partial x} - \gamma\left[(2\bar{n}+1)\, x \frac{\partial f}{\partial x} - \frac{1}{2} f\right] + \gamma \bar{n}\, x^2 \frac{\partial f}{\partial x} \qquad (3.59)$$

$$\frac{\partial f(x,t)}{\partial t} + \gamma\left[(2\bar{n}+1)x - (1+\bar{n}) - \bar{n}x^2\right] \frac{\partial f}{\partial x} = \frac{\gamma}{2} f \qquad (3.60)$$

This first order linear partial differential equation may be solved by the method of characteristics[30]: The characteristic equations for the above equation are

$$\frac{dx}{-\gamma\left[1+\bar{n} - (2\bar{n}+1)x + \bar{n}x^2\right]} = \frac{dt}{1} = \frac{df}{\frac{\gamma}{2} f} \qquad (3.61)$$

The right hand pair is soluble at once:

$$\frac{df}{f} = \frac{\gamma}{2}\,dt \Rightarrow f = e^{\frac{\gamma}{2}t}\,C \tag{3.62}$$

The integration constant is determined from the boundary condition

$$f(x,0) = (m+1)^{-\frac{1}{2}}\,X_0^{\,m+1} \tag{3.63}$$

Hence

$$f = \frac{e^{\frac{\gamma}{2}t}}{\sqrt{m+1}}\,(X_0)^{m+1} \tag{3.64}$$

where X_0 is the initial value of X . The left hand pair is

$$\frac{dx}{\bar{n}x^2 - (2\bar{n}+1)x + 1+\bar{n}} = -\gamma\,dt = \left\{\left(x - \frac{1+\bar{n}}{\bar{n}}\right)^{-1} - (x-1)^{-1}\right\}dx \tag{3.65}$$

Letting $C = \ln C'$, then

$$e^{-\gamma t} = C'\left(x - \frac{1+\bar{n}}{\bar{n}}\right)(x-1)^{-1} \tag{3.66}$$

Solving for x gives

$$X = \frac{\frac{\bar{n}+1}{\bar{n}}C' - e^{-\gamma t}}{C' - e^{-\gamma t}} \tag{3.67}$$

At t=0, we have

$$x_0 = \frac{\frac{\bar{n}+1}{\bar{n}} c' - 1}{c' - 1} \Rightarrow c' = \frac{x_0 - 1}{x_0 - \frac{1+\bar{n}}{\bar{n}}} \qquad (3.68)$$

Hence, the solution for x reducing to x_0 at t=0 is given by:

$$x = \left[(\bar{n}+1)(x_0-1) - (\bar{n}x_0 - \bar{n} - 1)e^{-\gamma t}\right]\left[\bar{n}(x_0-1) - (\bar{n}x_0 - \bar{n} - 1)e^{-\gamma t}\right]^{-1} \qquad (3.69)$$

Inverting this relation we find x_0 in terms of x and t:

$$x_0 = \frac{(1-x)\left[(1+\bar{n})e^{-\gamma t} - \bar{n}\right] - 1}{(1-x)\bar{n}\left[e^{-\gamma t} - 1\right] - 1} \qquad (3.70)$$

Therefore, the complete solution for $f_{(m+1)}(x,t)$ is (restoring the subscript on $f(x,t)$)

$$f_{m+1}(x,t) = \frac{e^{\frac{\gamma t}{2}}}{\sqrt{m+1}} \left\{ \frac{(1-x)\left[(1+\bar{n})e^{-\gamma t} - \bar{n}\right] - 1}{(1-x)\bar{n}\left[e^{-\gamma t} - 1\right] - 1} \right\}^{m+1} \qquad (3.71)$$

However, we have obtained the solution which corresponds to the evolution starting with a single non-zero density matrix element $\rho^{(0)}_{m,m+1}$ at $t=0$. To obtain the generating function for $\rho^{(t)}_{n,n+1}$ which evolves from an arbitrary initial distribution of elements - say $\rho^{(0)}_{n,n+1}$ - we do the following:

$$f(x,t) = \sum_{0}^{\infty} f_{(m+1)}(x,t)\,\rho^{(0)}_{m+1} \simeq \sum_{0}^{\infty} f_{(m)}(x,t)\,\rho_m(0) \qquad (3.72)$$

since $\rho^{(0)}_{-1,0} = 0$. Suppose that the generating function $\mathcal{F}(y)$ for the initial distribution is known:

$$\mathcal{F}(y) = \sum_{q=0}^{\infty} \rho^{(0)}_{q,q+1} \frac{x^{q+1}}{\sqrt{q+1}} \qquad (3.73)$$

then

$$\rho^{(0)}_{q,q+1} = \sqrt{q+1}\,\frac{1}{(q+1)!}\left(\frac{\partial}{\partial y}\right)^{q+1} \mathcal{F}(y)\Big|_{y=0} \qquad (3.74)$$

Therefore, using the above solution for $f_{(m+1)}(x,t)$ and the shifted sum for $f(x,t)$ we obtain the generating function for the matrix elements $s^{(t)}_{n,n+1}$ which have evolved from the (arbitrary) initial distribution $\rho^{(0)}_{q,q+1}$:

$$f(x,t) = \sum_{0}^{\infty} f_{(n)}(x,t)\,\rho_n(0) \qquad (3.75)$$

$$= \sum_{0}^{\infty} \frac{e^{-\frac{\gamma t}{2}}}{\sqrt{n}} (x_0)^n \sqrt{n}\,\frac{1}{n!}\left(\frac{\partial}{\partial y}\right)^n \mathcal{F}(y)\Big|_{y=0} \qquad (3.76)$$

$$= e^{\frac{\gamma t}{2}} \exp\left[x_0 \frac{\partial}{\partial y}\right] \mathcal{F}(y)\Big|_{y=0} \qquad (3.77)$$

$$= \Theta(x,t)\,\mathcal{F}(y) \qquad (3.78)$$

where X_0 is the above function of x and t.

We indicate how the regression theorem above allows us to use this result in the special case of the first order correlation function we want.

$$\langle a^+(t)\rangle = \sum_{n,n'}' a^+_{n'n}\, s^{(t)}_{nn'} = \sum_{n,n'}' \sqrt{n+1}\, \delta_{n',n+1}\, s^{(t)}_{n,n'} \tag{3.79}$$

$$s^{(t)}_{n,n+1} = \sum_{K,K'}' f(n,n+1;K,K',t)\, s^{(0)}_{KK'} \tag{3.80}$$

The equation for $s^{(t)}_{n,n+1}$ couples only elements on the stripe parallel to the main diagonal, so that we need not sum over all K, K':

$$s^{(t)}_{n,n+1} = \sum_{K}' f(n,n+1;K,K+1;t)\, s^{(0)}_{K,K+1} \tag{3.81}$$

so

$$\langle a^+(t)\rangle = \sum_{n,K} \sqrt{n+1}\, f(n,n+1;K,K+1;t)\, s^{(0)}_{K,K+1} \tag{3.82}$$

Let us write this in the same form that we had for the general case:

$$\langle a^+(t)\rangle = \sum_{n,n',K,K'} \delta_{n',n+1} \sqrt{n+1}\, f(n,n';K,K';t)\, s^{(0)}_{KK'}\, \delta_{K',K+1} \tag{3.83}$$

$$= \sum_{i,j}' \sum_{\substack{nn'\\KK'}} \delta_{n',n+1} \sqrt{n+1}\, f^{ij}(n,n';t)\,(K)^i (K')^j s^{(0)}_{KK'}\, \delta_{K',K+1} \tag{3.84}$$

$$= \sum_{i,j} \Big(\sum_{n,n'}' \delta_{n',n+1} \sqrt{n+1}\, f^{ij}(n,n';t) \Big) \langle Q^{ij}\rangle \tag{3.85}$$

where

$$\langle Q^{ij}\rangle = \sum_{\kappa,\kappa'} Q^{ij}_{\kappa'\kappa}\, s^{(0)}_{\kappa\kappa'} = \sum_{\kappa,\kappa'} (\kappa)^i (\kappa')^j \delta_{\kappa',\kappa+1}\, s^{(0)}_{\kappa\kappa'} \tag{3.86}$$

The regression theorem then states that

$$\langle a^+(t)a(0)\rangle = \sum_{ij}\Big\{\sum_{n,n'}{}' \delta_{n',n+1}\sqrt{n+1}\, f^{ij}(n,n';t)\Big\}\langle Q^{ij}a\rangle \tag{3.87}$$

where

$$\langle Q^{ij}a\rangle = \sum_{\kappa,\kappa',\kappa''} s^{(0)}_{\kappa\kappa'}\, Q^{ij}_{\kappa'\kappa''}\, a_{\kappa''\kappa} \tag{3.88}$$

$$= \sum_{\kappa,\kappa',\kappa''} s^{(0)}_{\kappa\kappa'}\, (\kappa'')^j (\kappa')^i \delta_{\kappa',\kappa''+1}\sqrt{\kappa}\,\delta_{\kappa'',\kappa-1} \tag{3.89}$$

Thus:

$$\langle a^+(t)a(0)\rangle = \sum_{ij}\sum_{\substack{n\,n'\\ \kappa\,\kappa'\kappa''}} \delta_{n',n+1}\sqrt{n+1}\, f^{ij}(n,n';t)\,(\kappa'')^i(\kappa')^j s^{(0)}_{\kappa\kappa'}\,\delta_{\kappa',\kappa''+1}\sqrt{\kappa}\,\delta_{\kappa'',\kappa-1} \tag{3.90}$$

$$= \sum_{\substack{n,n'\\ \kappa',\kappa''}} \delta_{n',n+1}\sqrt{n+1}\, f(n,n';\kappa'',\kappa';t)\sum_{\kappa}\sqrt{\kappa}\,\delta_{\kappa'',\kappa-1}\, s^{(0)}_{\kappa\kappa'}\,\delta_{\kappa',\kappa''+1} \tag{3.91}$$

Now:

$$\sum_{\kappa}\sqrt{\kappa}\;\delta_{\kappa'',\kappa-1}\, s^{(0)}_{\kappa\kappa'}\,\delta_{\kappa',\kappa''+1} = \sqrt{\kappa''+1}\; s^{(0)}_{\kappa''+1,\kappa'}\;\delta_{\kappa',\kappa''+1} \tag{3.92}$$

Hence, summing n',K' gives

$$\langle a^+(t)a(0)\rangle = \sum_{n,K''}' \sqrt{n+1}\, f(n,n+1;K'',K''+1;t)\sqrt{K''+1}\, s^{(0)}_{K''+1,K''+1} \tag{3.93}$$

$$= \sum_{n,K''}' \sqrt{n+1}\, f(n,n+1;K'',K''+1)\, \tilde{s}^{(0)}_{K''+1,K''+1} \tag{3.94}$$

Thus, this equation, when compared to the first expression above for $\langle a^+(t)\rangle$, indicates that the correlation function is obtained by just calculating $\langle a^+(t)\rangle$ with the changed initial distribution $\tilde{s}(0)$:

$$\tilde{s}^{(0)}_{K''+1,K''+1} \equiv \sqrt{K''+1}\, s^{(0)}_{K''+1,K''+1} \tag{3.95}$$

Suppose that we know the generating function for the distribution $s^{(0)}_{K''+1,K''+1}$:

$$h(y) = \sum_0^\infty{}' s^{(0)}_{n+1,n+1}(n+1)^{-\frac{1}{2}} y^{n+1} \tag{3.96}$$

Suppose also that we know the generating function for $\tilde{s}(0)$:

$$\tilde{h}(y) = \sum_0^\infty \tilde{s}^{(0)}_{n+1,n+1}(n+1)^{-\frac{1}{2}} y^{n+1} = \sum_0^\infty{}' \sqrt{n+1}\, s^{(0)}_{n+1,n+1}(n+1)^{-\frac{1}{2}} y^{n+1} \tag{3.97}$$

$$= \sum_0^\infty s^{(0)}_{n+1,n+1}\, y^{n+1} \simeq \sum_0^\infty s^{(0)}_{n,n}\, y^n \tag{3.98}$$

We are interested in the particular case that at t=0 the system is in thermal equilibrium with $\bar{n}$ occupation number. Hence, $\mathcal{A}^{(0)}_{n,n}$ is a thermal distribution:

$$\tilde{h}(y) = \sum_{0}^{\infty} \frac{\bar{n}^{n}}{(1+\bar{n})^{n+1}} y^{n} = \frac{1}{1+\bar{n}(1-y)} \qquad (3.99)$$

Previously we have found the operator $\mathcal{O}(x,t)$ which generated the distribution $\mathcal{A}^{(t)}_{n,n+1}$ needed to compute $\langle a^{+}(t)\rangle$: since

$$\langle a^{+}(t)\rangle = \sum_{0}^{\infty} \sqrt{n+1}\, \mathcal{A}^{(t)}_{n,n+1} = x\frac{\partial}{\partial x}\left(\sum_{0}^{\infty} \mathcal{A}^{(t)}_{n,n+1} (n+1)^{-\frac{1}{2}} x^{n+1}\right)\Big|_{x=1} \qquad (3.100)$$

$$= x\,\frac{\partial f(x,t)}{\partial x}\Big|_{x=1} = x\frac{\partial}{\partial x}\left\{\mathcal{O}(x,t)\,\mathcal{F}(y)\right\}\Big|_{x=1} \qquad (3.101)$$

the regression theorem indicates we need to carry out the same operations to obtain the correlation function, except $\tilde{h}(y)$ replaces $\mathcal{F}(y)$: so

$$\langle a^{+}(t)a(0)\rangle = x\frac{\partial}{\partial x}\left[\mathcal{O}(x,t)\tilde{h}(y)\right]\Big|_{x=1} = x\frac{\partial}{\partial x}\left\{e^{\frac{\gamma t}{2}}\left[\exp\left(x_0\frac{\partial}{\partial y}\right)\frac{1}{1+\bar{n}(1-y)}\right]_{y=0}\right\}\Bigg|_{x=1} \qquad (3.102)$$

where x_0 is the above found function of x and t. Substituting for x_0 , and carrying out the indicated operations (using $\exp\left(a\frac{\partial}{\partial y}\right)u(y)\Big|_{y=0} = u(a)$) one finds

$$\langle a^{+}(t)\,a(0)\rangle = \bar{n}\, e^{-\frac{\gamma t}{2}} \qquad (3.103)$$

Recalling that we have left off the time factor corresponding to the free time evolution of $\langle a^+(t)\rangle$, we finally obtain

$$\langle a^+(t)a(0)\rangle = \bar{n}\, e^{i\Omega t}\, e^{-\frac{\gamma t}{2}} \tag{3.104}$$

Returning to the electric field correlation function $G^{(1)}(t,0)$ (3.4), and assuming that it is a function of $|t|$ only, we obtain for the spectrum of scattered light

$$G^{(1)}(\omega) = \int_{-\infty}^{\infty} G^{(1)}(t,0)\, e^{-i\omega t}\, dt = \mathcal{E}^2 \bar{n} \int_{-\infty}^{\infty} e^{i(\omega_0+\Omega-\omega)t}\, e^{-\frac{\gamma|t|}{2}}\, dt \tag{3.105}$$

$$G^{(1)}(\omega) = \mathcal{E}^2 \frac{\bar{n}\gamma}{(\omega-\omega_0-\Omega)^2 + \left(\frac{\gamma}{2}\right)^2} \tag{3.106}$$

We now proceed to obtain the intensity-intensity correlation function. The calculation of $\langle a^+(t)a(t)\rangle$ involves knowledge of the diagonal density matrix elements $s_{n,n}^{(t)}$. Taking the (n,n) matrix elements of the equation of motion (3.33) for s(t) we obtain:

$$\begin{aligned} \frac{\partial}{\partial t} s_{nn}^{(t)} = {} & -\gamma\left[(2\bar{n}+1)n+\bar{n}\right] s_{nn}^{(t)} \\ & + \gamma(\bar{n}+1)(n+1)\, s_{n+1,n+1}^{(t)} \\ & + \gamma\,\bar{n}\, n\, s_{n-1,n-1}^{(t)} \end{aligned} \tag{3.107}$$

Calling

$$s_{nn}(t) \equiv S'_n(t) \tag{3.108}$$

and solving the equation subject to the boundary condition

$$S'_n(0) = \delta_{n,m} \tag{3.109}$$

is at once possible since the above equation for $S'_n(t)$ is a special case of the problem treated by Shimoda, Takahashi, and Townes [30]. The result for the generating function is

$$f_{(m)}(x,t) = \frac{1}{1+G(1-x)}\left[\frac{1+(G-K)(1-x)}{1+G(1-x)}\right]^m \tag{3.110}$$

where

$$K = e^{-\gamma t} \quad ; \quad G = \bar{n}\,(1-e^{-\gamma t}) \tag{3.111}$$

Once again, the generating function which gives the diagonal density matrix elements $S'_n(t)$ which have evolved from an arbitrary initial distribution $\rho_n(0)$ is given by

$$f(x,t) = \sum_{m=0}^{\infty} f_{(m)}(x,t)\,\rho_m(0) \tag{3.112}$$

Suppose that the generating function $\mathcal{F}(y)$ of the initial distribution $\rho_m(0)$ is known:

$$\mathcal{F}(y) = \sum_{n=0}^{\infty} \rho_n(0)\,y^n \tag{3.113}$$

Thus

$$\rho_n(0) = \frac{1}{n!}\left(\frac{\partial}{\partial y}\right)^n \mathcal{F}(y)\Big|_{y=0} \tag{3.114}$$

Therefore, using the solution for $f_{(m)}$ above,

$$f(x,t) = \sum_{m=0}^{\infty} \frac{1}{1+G(1-x)}\left[\frac{1+(G-K)(1-x)}{1+G(1-x)}\right]^m \frac{1}{m!}\left(\frac{\partial}{\partial y}\right)^m \mathcal{F}(y)\Big|_{y=0} \tag{3.115}$$

$$f(x,t) = \frac{1}{1+G(1-x)}\exp\left\{\frac{1+(G-K)(1-x)}{1+G(1-x)}\frac{\partial}{\partial y}\right\}\mathcal{F}(y)\Big|_{y=0} \tag{3.116}$$

$$= \Theta(x,t)\,\mathcal{F}(y) \tag{3.117}$$

Now, the regression theorem argument in this case goes as follows: we use only one index, since all matrix elements are diagonal -

$$\rho_n(t) = \sum_K f(n,K,t)\,\rho_K(0) \tag{3.118}$$

$$\langle a^\dagger(t)a(t)\rangle = \sum_n n\,\rho_n(t) = \sum_{n,K} n\,f(n,K,t)\,\rho_K(0) \tag{3.119}$$

$$= \sum_{n,K} n \sum_i f^i(n,t)\,K^i\,\rho_K(0) \tag{3.120}$$

$$= \sum_i \left(\sum_n n\,f^i(n,t)\right)\langle Q^i\rangle \tag{3.121}$$

where

$$\langle Q^i\rangle = \sum_K (K)^i\,\rho_K(0) = \langle (a^\dagger a)^i\rangle \tag{3.122}$$

Thus, the regression theorem states that the intenisty correlation function will be given by

$$\langle a^{+}(0)a^{+}(t)a(t)a(0)\rangle = \sum_{i}\Big(\sum_{n} n f^{i}(n,t)\Big)\langle a^{+}Q^{i}a\rangle \tag{3.123}$$

$$= \sum_{i}\Big(\sum_{n} n f^{i}(n,t)\Big)\sum_{K} K(K-1)^{i}\rho_{K}(0) \tag{3.124}$$

$$= \sum_{n,K} n f(n,K-1,t)\,K\,\rho_{K}(0) \tag{3.125}$$

$$= \sum_{n,K} n f(n,K,t)\,(K+1)\,\rho_{K+1}(0) \tag{3.126}$$

This correlation function corresponds to evaluating $\langle a^{+}(t)a(t)\rangle$ with an effective initial diagonal density matrix $\tilde{\rho}(0)$:

$$\tilde{\rho}_{K}(0) \equiv (K+1)\rho_{K}(0) \tag{3.127}$$

We may obtain the elements $\tilde{\rho}_{K}(0)$ from the initial generating function $\mathcal{F}(y)$ by

$$\tilde{\rho}_{K}(0) = \frac{1}{K!}\left(\frac{\partial}{\partial x}\right)^{K+1}\mathcal{F}(x)\Big|_{x=0} \tag{3.128}$$

Hence, the generating function of the distribution $\tilde{\rho}_{K}(0)$ is obtained by:

$$\tilde{\mathcal{F}}(y) = \sum_{K=0}^{\infty}\tilde{\rho}_{K}(0)y^{K} = \sum_{0}^{\infty} y^{K}\frac{1}{K!}\left(\frac{\partial}{\partial x}\right)^{K+1}\mathcal{F}(x)\Big|_{x=0} \tag{3.129}$$

$$= \frac{\partial}{\partial x}\exp\left[y\frac{\partial}{\partial x}\right]\mathcal{F}(x)\Big|_{x=0} \tag{3.130}$$

$$= \exp\left(y\frac{\partial}{\partial x}\right)\left(\frac{\partial\mathcal{F}(x)}{\partial x}\right)\Big|_{x=0} \tag{3.131}$$

Therefore, the new generating function which we must use to compute $\langle a^{+}(0)\, a^{+}(t)\, a(t)\, a(0)\rangle$ is given by

$$\tilde{f}(x,t) = \mathcal{O}(x,t)\,\tilde{\mathcal{F}}(y) = \frac{1}{1+G(1-x)}\, \exp\left\{ \frac{1+(G-K)(1-x)}{1+G(1-x)} \frac{\partial}{\partial y} \right\} \tilde{\mathcal{F}}(y) \Big|_{y=0} \tag{3.132}$$

$$= \frac{1}{1+G(1-x)}\, \exp\left\{ \frac{1+(G-K)(1-x)}{1+G(1-x)} \frac{\partial}{\partial y} \right\} \left[\left(\exp\left(y \frac{d}{dz}\right) \frac{\partial \mathcal{F}(z)}{\partial z} \right)\Big|_{z=0} \right]\Big|_{y=0} \tag{3.133}$$

Since

$$\langle a^{+}(t)\, a(t)\rangle = \sum n\, S_n(t) = \left\{ x \frac{\partial}{\partial x} f(x,t) \right\}\Big|_{x=1} \tag{3.134}$$

we have

$$\langle a^{+}(0)\, a^{+}(t)\, a(t)\, a(0)\rangle = \left\{ x \frac{\partial}{\partial x} \tilde{f}(x,t) \right\}\Big|_{x=1} \tag{3.135}$$

with $\tilde{f}(x,t)$ given above. We are interested in the specific case that the system starts off in thermal equilibrium. Thus, $\mathcal{F}(z)$ is taken as a Bose-Einstein distribution:

$$\mathcal{F}(z) = \frac{1}{1+\bar{n}(1-z)} \tag{3.136}$$

Thus

$$\frac{\partial}{\partial z} \mathcal{F}(z) = \bar{n}\left[1+\bar{n}(1-z)\right]^{-2} \tag{3.137}$$

Using

$$\exp\left(a \frac{\partial}{\partial z}\right) \mathcal{F}'(z)\Big|_{z=0} = \mathcal{F}'(a) \tag{3.138}$$

we get

$$\exp\left(y \frac{\partial}{\partial z}\right) \mathcal{F}'(z)\Big|_{z=0} = \bar{n}\left[1+\bar{n}(1-y)\right]^{-2} \tag{3.139}$$

Next,

$$\tilde{f}(x,t) = \frac{1}{1+G(1-x)} \mathcal{F}'\left(\frac{1+(G-K)(1-x)}{1+G(1-x)}\right) \tag{3.140}$$

where $\mathcal{F}'(z)$ is the function given above, and it is to be evaluated with the indicated argument. The last step involves the operation $(x \frac{\partial}{\partial x} \tilde{f} \,\big|_{x=1}$ and, using this result for $\tilde{f}$, the algebra yields

$$\langle a^{+}(0)\, a^{+}(t)\, a(t)\, a(0)\rangle = \bar{n}^2 + \bar{n}^2 e^{-\gamma t} \tag{3.141}$$

This is the exact result since the free field time evolution of $\langle a^{+}(t)\, a(t)\rangle$ is just equal to one.

Returning to the definition of $G^{(2)}(0,t;t,0)$, (3.5), we find for the intensity fluctuation spectrum:

$$G^{(2)}(\omega) = \int_{-\infty}^{\infty} G^{(2)}(0,t;t,0)\, e^{-i\omega t}\, dt \tag{3.142}$$

$$G^{(2)}(\omega) = \mathcal{E}^4 \left\{ 2\pi \bar{n}^2 \delta(\omega) + \frac{2\gamma \bar{n}^2}{\omega^2+\gamma^2} \right\}$$

It is to be noted that this result satisfied the Gaussian "factorization" property, if the light is treated as a

Gaussian random variable, or the equivalent density matrix factorization condition. Both of these indicate that the following relation should hold:

$$G^{(2)}(\omega) = 2\pi \left[G^{(1)}(t,t)\right]^2 \delta(\omega) + \frac{1}{2\pi} G^{(1)}(\omega) \otimes G^{(1)*}(\omega) \tag{3.143}$$

where $\otimes$ denotes convolution as given by (1.17). Similar results for the oscillator correlation functions have been obtained by other methods [16,31].

PART B. FLUCTUATIONS OF A FERROMAGNET

Part B. Section I. Model and Density Operator Equation of Motion

The results of the previous Part have shown that the light scattered by "normal" systems may well be expected to be Gaussian in nature in the sense that the intensity correlation function of the scattered light is determined essentially entirely by the amplitude correlation function. That is, measurement of the second order temporal correlation function does not furnish more information than that contained in the first order correlation function.

By normal systems we meant to exclude systems undergoing phase transitions. Such systems are known to exhibit peculiar light scattering properties such as critical opalescence[34]. The interpretation of scattering and other anomalous properties[35] is usually linked to the existence of long (that is, of macroscopic dimensions) range fluctuations. The consequences of such fluctuations to the interpretation of self beating experiments have already been noted. In addition, the possibility arises that the fluctuations themselves will not have the Gaussian statistics usually associated with thermodynamic fluctuations[13,18] and thus will have properties unlike the system studied in Part A, Section III.

Motivated by these considerations, we undertake a study of fluctuations in a magnetic system. Although extensive experimental work has been done on light scattering by ferro- and antiferro magnetic crystals [36] as well as on the theoretical interpretation [37,38,39] of these experiments, we present the results of this Part as a study of the dynamical fluctuation properties of a model ferromagnetic system without an explicit calculation of the scattered light statistics.

We wish to study the dynamical properties of a ferromagnetic system [40] using density operator techniques closely related to those used to study the oscillator of Part A, Section III. Our general approach will therefore be to consider the ferromagnetic spin system to be in contact with a thermal reservoir. We shall obtain the equation of motion for the combined reservoir - ferromagnet system and then trace over reservoir states to obtain an equation of motion for the reduced density operator of the spin system. The latter operator contains all of the information relevant to the calculation of the dynamical and fluctuation properties of the ferromagnet.

We take as our total Hamiltonian H the sum of the ferromagnet Hamiltonian H_s, the reservoir Hamiltonian H_R, and an interaction V:

$$H = H_s + H_R + V \tag{1}$$

We choose the Heisenberg-Ising model [14] for our magnetic system:

$$H_S = -\frac{1}{2}\sum_{i,j=1}^{N}{}' J_{ij}\, S_i^z S_j^z - \mu H_0 \sum_{j=1}^{N}{}' S_j^z \qquad (2)$$

That is, we imagine N spin one half angular momenta, localized at fixed lattice sites and each having a magnetic moment μ, to interact ferromagnetically ($J_{ij} \geq 0$) with each other and also interact with an externally applied uniform magnetic field H_0 which is taken to define the Z-direction of the system. The angular momentum operators obey the usual commutation relations (for a spin one half total angular momentum):

$$[S_i^x, S_i^y] = i\,\delta_{ij}\, S_j^z \qquad (3)$$

(the complex constant i in (3) is not to be confused with the site index i on the spin operators) and the two similar relations obtained from (3) by cyclically permuting the spacial indices x,y, and z.

We shall imagine that the spin system is in thermal contact with another system (such as the phonons in the crystal, or spin impurities) which forms the reservoir. The specific nature of the reservoir will be clarified when particular examples are considered below. We will take the reservior Hamiltonian to represent a set of uncoupled modes:

$$H_R = \sum_\alpha \hbar \Omega_\alpha O_\alpha^+ O_\alpha \tag{4}$$

We take the following general form for the system-reservoir interaction V:

$$V = \hbar \sum_{j=1}^{N} \sum_\alpha g \left[S_j^- O_\alpha(r_j) + S_j^+ O_\alpha^+(r_j) \right] \tag{5}$$

where

$$S_j^\pm = S_j^x \pm S_j^y \tag{6}$$

are the usual spin raising (+) and lowering (-) operators for the j^{th} spin and the reservoir operators are evaluated at the position of the j^{th} spin. For simplicity, we consider a single effective coupling constant g. (In Appendix III we shall present an alternative derivation of our main equation, to be derived below; we shall see there that our choice for V, equation (5), is not quite the most general possible system - reservoir interaction, but it is entirely adequate for the main purposes of our discussion).

Having specified our total system, we proceed to consider an equation of motion for its density operator $\rho(t)$ in the interaction picture:

$$\dot{\rho}(t) = \frac{1}{i\hbar} \left[V(t), \rho(t) \right] \tag{7}$$

where

$$V(t) = e^{\frac{i}{\hbar}(H_S + H_R)t}\, V\, e^{-\frac{i}{\hbar}(H_S + H_R)t} \tag{8}$$

In order to evaluate (8), we make a molecular field approximation to the Ising Hamiltonian (2) in the form

$$\begin{aligned} H_S &\simeq -\mu \sum_{j=1}^{N} \mathcal{H}_{eff}\, S_j^z - \mu H_0 \sum_{j=1}^{N} S_j^z \\ &= -\sum_{j=1}^{N} \hbar\omega_{21}\, S_j^z \end{aligned} \tag{9}$$

where we denote the energy splitting of the two states of each spin one half system by $\hbar\omega_{21} \geqslant 0$. We shall call the state of higher energy (but lower value of z component of spin, $S^z = -\frac{1}{2}$) "2" and the state of lower energy (but higher z component of spin, $S^z = +\frac{1}{2}$) "1" so that the energies ϵ_2 , ϵ_1 of these states are related by

$$\epsilon_2 - \epsilon_1 = \hbar\omega_{21} \geqslant 0 \tag{10}$$

Using (4) and (9) we obtain from (8) [41]:

$$V(t) = \hbar \sum_{j=1}^{N}{}' \sum_{\alpha}{}' g\left[S_j^- e^{i\omega_{21}t} O_\alpha(r_j) e^{-i\Omega_\alpha t} + S_j^+ e^{-i\omega_{21}t} O_\alpha(r_j) e^{i\Omega_\alpha t}\right] \tag{11}$$

Formally integrating (7) we obtain

$$\rho(t) = \rho(-\infty) + \frac{1}{i\hbar}\int_{-\infty}^{t} dt' \left[V(t'), \rho(t')\right] \tag{12}$$

which we insert in the right side of (7) to get

$$\dot{\rho}(t) = \frac{1}{i\hbar}\left[V(t), \rho(-\infty)\right] - \frac{1}{\hbar^2}\int_{-\infty}^{t} dt'\left[V(t),\left[V(t'), \rho(t')\right]\right] \qquad (14)$$

We try a solution of the form

$$\rho(t) = \sigma(t) f_0(H_R) + \Delta \qquad (15)$$

where $\sigma(t)$ is the reduced density operator of the system defined by tracing over the reservoir

$$\sigma(t) = Tr_R\, \rho(t)$$

and $f_0(H_R)$ is the thermal equilibrium density operator of the reservoir ($\beta = (K_B T)^{-1}$)

$$f_0(H_R) = e^{-\beta H_R}\left[Tr_R\, e^{-\beta H_R}\right]^{-1} \qquad (16)$$

Assuming that the reservoir-system interaction may be adequately treated to second order, we ignore Δ and thus obtain, upon tracing over the reservoir in equation (14)

$$\dot{\sigma}(t) = -\frac{1}{\hbar^2}\int_{-\infty}^{t} dt'\, Tr_R\left[V(t),\left[V(t'), \sigma(t) f_0(H_R)\right]\right] \qquad (17)$$

where the first term in (14) does not contribute due to the form of $V(t)$ and the diagonal reservoir density operator. The Markoff approximation $\sigma(t') \rightarrow \sigma(t)$ will be justified below. Using (11) for $V(t)$ in each of the four terms obtained by expanding the double commutator in (17), one has a number of resulting terms to calculate. We shall consider one of these in detail to illustrate the calculations.

One of the terms of (17) is

$$T = -g^2 \int_{-\infty}^{t} dt' \, Tr_R \Big[\sum_{j=1}^{N} \sum_{\alpha} S_j^- e^{i\omega_{21} t} O_\alpha(r_j) e^{-i\Omega_\alpha t} \sum_{k=1}^{N} \sum_{\beta} S_j^+ \tag{18}$$

$$e^{-i\omega_{21} t'} O_\beta^+(r_k) e^{i\Omega_\beta(t')} \sigma(t) f_0(H_R) \Big]$$

Since $f_0(H_R)$ is diagonal, only the $\alpha = \beta$ term can contribute to (18). We make the following assumption about reservoir correlation functions:

$$\sum_\alpha Tr_R \left[O_\alpha(r_j) O_\alpha^+(r_k) \right] = \sum_\alpha \langle O_\alpha(r_j) O_\alpha^+(r_k) \rangle \propto \delta(r_j - r_k) \tag{19}$$

so that only terms for which $j = k$ survive. This assumption means that each spin interacts with the chaotic heat reservoir independently of the others. In the calculation of other terms of (17), we assume that

$$\sum_\alpha \langle O_\alpha^+(r_i) O_\alpha(r_k) \rangle \propto \delta(r_i - r_k) \tag{20}$$

and we have implicitly assumed that all correlations of the following forms vanish :

$$\langle O_\alpha^+(r_i) O_\beta^+(r_k) \rangle = \langle O_\alpha(r_i) O_\beta(r_k) \rangle = 0 \tag{21}$$

Thus, we obtain from (18):

$$T = -g^2 \int_{-\infty}^{t} dt' \sum_{j=1}^{N} \sum_{\alpha} S_j^- S_j^+ \langle O_\alpha O_\alpha^+ \rangle e^{i(\omega_{21} - \Omega_\alpha)(t-t')} \sigma(t) \tag{22}$$

We assume that the sum over reservoir states α may be converted into an integral over energies Ω_α by means of a density of states factor $g(\Omega_\alpha)$ so that

$$\sum_\alpha e^{i(\omega_{21} - \Omega_\alpha)(t-t')} \langle O_\alpha O_\alpha^+ \rangle = \int_0^\infty d\Omega_\alpha \, g(\Omega_\alpha) \langle O O^+(\Omega_\alpha) \rangle e^{i(\omega_{21} - \Omega_\alpha)(t-t')}$$

$$= \int_{\omega_{21}}^{-\infty} (-du) \, g(\omega_{21} - u) \langle O O^+(\omega_{21} - u) \rangle e^{iu(t-t')} \tag{23}$$

$$\cong g(\omega_{21}) \langle O O^+(\omega_{21}) \rangle \int_{-\infty}^{\infty} du \, e^{iu(t-t')}$$

$$= 2\pi \, g(\omega_{21}) \langle O O^+(\omega_{21}) \rangle \, \delta(t-t')$$

Thus we see the justification of the Markoff approximation in (17) and obtain

$$T = -\frac{1}{2} \Gamma_2 \sum_{j=1}^{N} S_j^- S_j^+ \sigma(t) \tag{24}$$

where, returning to the more general expressions, the damping constant Γ_2 is given by

$$\frac{1}{2} \Gamma_2(\omega_{21}) = g^2 \, Re \int_{-\infty}^{t} dt' \left\langle \sum_\alpha O_\alpha(t) \sum_\beta O_\beta^+(t') \right\rangle e^{i\omega_{21}(t-t')} \tag{25}$$

A similar calculation shows the damping constant of the other level of the two level systems to be

$$\frac{1}{2}\Gamma_1(\omega_{21}) = g^2 \, \mathrm{Re} \int_{-\infty}^{t} dt' \left\langle \sum_{\alpha} O_{\alpha}^{+}(t) \sum_{\beta} O_{\beta}(t') \right\rangle e^{-i\omega_{21}(t-t')} \qquad (26)$$

By taking only the real parts of the expressions involved we neglect the frequency shifts associated with these damping constants. Note that they depend upon the level separation ω_{21}. In Appendix IV we show from the general forms (25) and (26) that the following relation holds, as expected from detailed balance considerations:

$$\Gamma_1(\omega_{21}) = e^{-\beta\hbar\omega_{21}} \, \Gamma_2(\omega_{21}) \qquad (27)$$

By computing the other terms of (17) in a similar manner, we obtain the following equation of motion for the reduced density operator of the ferromagnetic system:

$$\begin{aligned} \dot{\sigma}(t) = \sum_{j=1}^{N} \Big\{ & \left[\frac{\epsilon_1}{i\hbar} - \frac{\Gamma_1}{2}\right]\left[\frac{1}{2} I_j + S_j^z\right]\sigma(t) + \left[\frac{\epsilon_2}{i\hbar} - \frac{\Gamma_2}{2}\right]\left[\frac{1}{2} I_j \right. \\ & \left. - S_j^z\right]\sigma(t) - \left[\frac{\epsilon_1}{i\hbar} + \frac{\Gamma_1}{2}\right]\sigma(t)\left[\frac{1}{2} I_j + S_j^z\right] - \left[\frac{\epsilon_2}{i\hbar} + \frac{\Gamma_2}{2}\right]\sigma(t)\left[\frac{1}{2} I_j \right. \\ & \left. - S_j^z\right] + \Gamma_1 S_j^- \sigma(t) S_j^+ \\ & + \Gamma_2 S_j^+ \sigma(t) S_j^- \Big\} \end{aligned} \qquad (28)$$

We have returned to the Schrödinger picture and I_j is the unit operator in the space of states of the j^{th} spin.

Part B. Section II. Magnetization Distribution Function

In order to work out the calculation of this section, it is convenient to rewrite (28) in terms of atomic operators rather than spin operators. Thus, denoting by $|1\rangle_j$ the lower energy state of the j^{th} spin and by $|2\rangle_j$ the higher energy state, we obtain the following relations:

$$S_j^- = (|2\rangle\langle 1|)_j \tag{29}$$

$$S_j^+ = (|1\rangle\langle 2|)_j \tag{30}$$

$$S_j^z = \tfrac{1}{2}\left[(|1\rangle\langle 1|)_j - (|2\rangle\langle 2|)_j\right] \tag{31}$$

$$I_j = (|1\rangle\langle 1|)_j + (|2\rangle\langle 2|)_j . \tag{32}$$

The inverse relations for the last pair of relations are

$$(|1\rangle\langle 1|)_j = \tfrac{1}{2} I_j + S_j^z \tag{33}$$

$$(|2\rangle\langle 2|)_j = \tfrac{1}{2} I_j - S_j^z \tag{34}$$

In terms of these operators, (28) becomes:

$$\dot{\sigma}(t) = \sum_{j=1}^{N} \left\{ \left[\frac{\epsilon_1}{i\hbar} - \frac{\Gamma_1}{2}\right] (|1\rangle\langle 1|)_j\, \sigma(t) + \left[\frac{\epsilon_2}{i\hbar} - \frac{\Gamma_2}{2}\right] (|2\rangle\langle 2|)_j\, \sigma(t) \right.$$

$$- \left[\frac{\epsilon_1}{i\hbar} + \frac{\Gamma_1}{2} \right] \sigma(t) \left(|1\rangle\langle 1| \right)_j - \left[\frac{\epsilon_2}{i\hbar} + \frac{\Gamma_2}{2} \right] \sigma(t) \left(|2\rangle\langle 2| \right)_j$$

$$+ \Gamma_1 \left(|2\rangle\langle 1| \right)_j \sigma(t) \left(|1\rangle\langle 2| \right)_j$$

$$+ \Gamma_2 \left(|1\rangle\langle 2| \right)_j \sigma(t) \left(|2\rangle\langle 1| \right)_j \Big\} \qquad (35)$$

We now define operators corresponding to the transverse components of the total (dimensionless) magnetization by

$$S^- \equiv \sum_{j=1}^{N} \left(|2\rangle\langle 1| \right)_j$$

$$S^+ \equiv \sum_{j=1}^{N} \left(|1\rangle\langle 2| \right)_j \qquad (36)$$

Since the total number of spins is fixed at N, it is convenient to introduce a third operator variable corresponding to the total upper state population:

$$N_2 \equiv \sum_{j=1}^{N} \left(|2\rangle\langle 2| \right)_j \qquad (37)$$

We may similarly define the total lower state number operator by

$$N_1 \equiv \sum_{j=1}^{N} \left(|1\rangle\langle 1| \right)_j \qquad (38)$$

and note that the following relation always holds between these two quantities:

$$N_1 + N_2 = N \tag{39}$$

We next define a c-number quasiprobability distribution function P[15,16,33] of c-number variables s, η_2 and s^* by

$$\mathcal{P}(s, \eta_2, s^*, t) \equiv Tr\left[\sigma(t)\,\delta(s - S^-)\,\delta(\eta_2 - N_2)\,\delta(s^* - S^+)\right] \tag{40}$$

where the delta functions are defined formally by integral representations

$$\delta(\eta_2 - N_2) \equiv \frac{1}{2\pi}\int_{-\infty}^{\infty} dx\, e^{i(\eta_2 - N_2)x} \tag{41}$$

and similarly for the other two. The exponential operator in (41) is defined formally by its series expansion

$$e^{ix(\eta_2 - N_2)} = \sum_{K=0}^{\infty} \frac{(ix)^K}{K!}(\eta_2 - N_2)^K \tag{42}$$

We note again that the quantities s, η_2 and s^* are not operators, but rather are c-numbers so that the distribution function $\mathcal{P}(s, \eta_2, s^*, t)$ is also a c-number function. The significant point is that in (40) we have chosen to order the (non-commuting) operators S^-, N_2 and S^+ in a particular way: that is, in every term of (40) obtained by using the expansions of the delta functions all of the S^- operators are to the left of all of the N_2 operators which in turn are to the left of the S^+ operators. We have chosen this operator order arbitrarily; choosing a different order neither changes the final form of the equation we solve nor simplifies the calculation noticeably.

The quasiprobability distribution function introduced here is closely related to the Glauber P (α) distribution [10] which was introduced in order to study statistical properties of the electromagnetic field. The mathematical properties of P (α) have been extensively studied [42]. The system being studied here requires three operators, so the distribution function (40) is a generalization of the P (α) function. The utility of introducing such a distribution comes from the fact that as we are dealing with a system involving the set (S^-, N_2 , S^+) of non-commuting operators, we may use their commutation relations to put every product of operators which may occur into a particular chosen order (namely every S^- to the left of every N_2 which in turn is to the left of every S^+). Having thus chosen a conventional ordering, we may associate with an arbitrary operator O (S^-, N_2 , S^+) (we envisage O to be sums of products of the three basic operators) a c-number function $O^{(c)}(s, n_2, s^*)$ of the c-number variables s , n_2 , s^* .

$$O(S^-, N_2, S^+) = C O^{(c)}(s, n_2, s^*) \qquad (43)$$

or

$$O(S^-, \eta_2, S^+) = \int ds\, d\eta_2\, ds^*\, O^{(C)}(s, n_2, s^*)\,\delta(s-S^-)\,\delta(\eta_2-N_2)\,\delta(s^*-S^+) \tag{44}$$

where the delta functions are defined by (41) and (42). The operator C instructs one to replace in $O^{(C)}(s, \eta_2, s^*)$ each of the variables s, η_2, and s^* by the corresponding operators S^-, N_2, S^+ --keeping the operators in the chosen order by doing the integrations in (44) in sequence. One can similarly define a c-number distribution function associated with the density operator $\sigma(t)$ by (40) so that in order to calculate the average value of the operator $O(S^-, N_2, S^+)$ we need

$$\begin{aligned}\langle O(S^-, N_2, S^+)\rangle &= Tr\left[\sigma(t)\, O(S^-, N_2, S^+)\right]\\ &= Tr\, \sigma(t) \int ds\, d\eta_2\, ds^*\, O^{(C)}(s, n_2, s^*)\,\delta(s-S^-)\,\delta(n_2-N_2)\,\delta(s^*-S^+)\\ &= \int ds\, d\eta_2\, ds^*\, O^{(C)}(s, n_2, s^*)\, Tr\left[\sigma(t)\,\delta(s-S^-)\,\delta(\eta_2-N_2)\,\delta(s^*-S^+)\right]\\ &= \int ds\, d\eta_2\, ds^*\, O^{(C)}(s, n_2, s^*)\, \mathcal{P}(s, n_2, s^*, t)\end{aligned} \tag{45}$$

The interpretation of $P(s, \eta_2, s^*, t)$ as a classical probability distribution may not always be justified. Studies[42] of the Glauber $\mathcal{P}(\alpha)$ distribution have shown that it may possess singularities as well as take on negative values for some range of its arguments. However, we shall use (40) in expressions of the type (45) and shall see that in certain cases that it does indeed have the properties of a classical distribution function.

Returning to (40) and differentiating both sides with respect to time, we see that the equation of motion of $P(s, \eta_2, s^*, t)$ is

$$\dot{P}(s,\eta_2,s^*,t) = \mathrm{Tr}\left[\dot{\sigma}(t)\,\delta(s-S^-)\,\delta(\eta_2-N_2)\,\delta(s^*-S^+)\right] \tag{46}$$

Therefore, using the equation of motion (35) we have

$$\begin{aligned}\dot{P}(s,\eta_2,s^*,t) = \mathrm{Tr}\Big\{\sigma(t)\sum_{j=1}^{N}\Big[&\left(\frac{\epsilon_1}{i\hbar}-\frac{\Gamma_1}{2}\right)\delta^c\,(|1\rangle\langle 1|)_j + \left(\frac{\epsilon_2}{i\hbar}-\frac{\Gamma_2}{2}\right)\delta^c\,(|2\rangle\langle 2|)_j \\ &-\left(\frac{\epsilon_1}{i\hbar}+\frac{\Gamma_1}{2}\right)(|1\rangle\langle 1|)_j\,\delta^c - \left(\frac{\epsilon_2}{i\hbar}+\frac{\Gamma_2}{2}\right)(|2\rangle\langle 2|)_j\,\delta^c \\ &+\Gamma_2\,(|2\rangle\langle 1|)_j\,\delta^c\,(|1\rangle\langle 2|)_j \\ &+\Gamma_1\,(|1\rangle\langle 2|)_j\,\delta^c\,(|2\rangle\langle 1|)_j\Big]\Big\}\end{aligned} \tag{47}$$

where we have abbreviated the product of the three delta functions occuring in the definition (40) by δ^c :

$$\delta^c = \delta(s-S^-)\,\delta(\eta_2-N_2)\,\delta(s^*-S^+) \tag{48}$$

Note the slightly different order of the operators in (49) compared to (35) due to our extraction of the density operator to the extreme left.

We shall evaluate the various terms in (47) by rearranging them into the chosen operator order and using various devices to compute the resulting expressions[17]. We begin by noting that the atomic states are orthogonal for different atoms (i,j)

$$ {}_i\langle 1|1\rangle_j = {}_i\langle 2|2\rangle_j = \delta_{ij} \; ; \quad {}_i\langle 1|2\rangle_j = 0 \qquad (49)$$

so that the commutators of our basic operators may be written, using the definitions (36), (37), and (38), as:

$$\begin{aligned} [S^+, N_2] &= \Big[\sum_{i=1}^{N} (|1\rangle\langle 2|)_i \, , \; \sum_{j=1}^{N} (|2\rangle\langle 2|)_j \Big] \\ &= \sum_{i=1}^{N} \sum_{j=1}^{N} \Big[(|1\rangle\langle 2|)_i (|2\rangle\langle 2|)_j - (|2\rangle\langle 2|)_j (|1\rangle\langle 2|)_i \Big] \\ &= \sum_{i=1}^{N} \sum_{j=1}^{N} \delta_{ij} \, (|1\rangle\langle 2|)_j \qquad (50) \\ &= S^+ \end{aligned}$$

Similarly we find that

$$[S^-, N_2] = -S^- \qquad (51)$$

$$[S^+, S^-] = N_1 - N_2 \qquad (52)$$

$$[S^+, N_1] = -S^+ \qquad (53)$$

$$[S^-, N_1] = S^- \qquad (54)$$

Consider the first term of (47):

$$T_1 = \left(\frac{\epsilon_1}{i\hbar} - \frac{\Gamma_1}{2}\right) Tr\left[\sigma(t)\,\delta(\alpha - S^-)\,\delta(n_2 - N_2)\,\delta(\alpha^* - S^+)\,N_1\right] \tag{55}$$

Noting that N_1 is directly proportional to N_2 via (39), we wish to commute it through the first delta function so as to bring it next to the middle delta function. We note that we may rewrite the chosen order delta function as

$$\delta^c = e^{-S^-\frac{\partial}{\partial \alpha}}\, e^{-N_2\frac{\partial}{\partial n_2}}\, e^{-S^+\frac{\partial}{\partial \alpha^*}}\,\delta(\alpha)\,\delta(n_2)\,\delta(\alpha^*) \tag{56a}$$

by interpreting the exponential factors as displacement operators which act on c-number (delta) functions. We next evaluate

$$t = e^{-S^+\partial_{\alpha^*}}\, N_1\, e^{S^+\partial_{\alpha^*}} \tag{56b}$$

(we shall use the shorthand notation that $\frac{\partial}{\partial \alpha} \equiv \partial_\alpha$, etc.) by considering the following object

$$f(\xi) = e^{-\xi S^+}\, N_1\, e^{\xi S^+} \tag{57}$$

where ξ is a c-number variable. Evidently $f'(\xi)$ satisfies

$$\frac{df(\xi)}{d\xi} = e^{-\xi S^+}\left[-S^+ N_1 + N_1 S^+\right] e^{\xi S^+} = e^{-\xi S^+}\, S^+\, e^{\xi S^+} = S^+ \tag{58}$$

using the commutator (53). We may integrate (58) to get

$$f(\xi) = f(0) + S^+\xi \tag{59}$$

where, from (57),

$$f(0) = N_1 \tag{60}$$.

Thus, using (60) in (59) and replacing ξ by ∂_{s^*} we obtain for t

$$t = N_1 + \partial_{s^*} S^+ \tag{61}$$

Thus we may write

$$e^{-S^+\partial_{s^*}} N_1 = \left[e^{-S^+\partial_{s^*}} N_1 e^{S^+\partial_{s^*}}\right] e^{-S^+\partial_{s^*}} = (N_1 + \partial_{s^*} S^+)\, e^{-S^+\partial_{s^*}} \tag{62}$$

so that T_1 becomes

$$T_1 = \left(\frac{\epsilon_1}{i\hbar} - \frac{\Gamma_1}{2}\right) Tr\left[\sigma(t)\, e^{-S^-\partial_s} e^{-N_2\partial_{n_2}} (N_1 + \partial_{s^*} S^+)\, e^{-S^+\partial_{s^*}}\right] \delta(s)\delta(n_2)\delta(s^*) \tag{63}$$

The operators in (63) are in the correct order so that no other ordering must be done. We evaluate it as follows:

$$T_1 = \left(\frac{\epsilon_1}{i\hbar} - \frac{\Gamma_1}{2}\right) Tr\Big[\sigma(t) \int ds'\, dn_2'\, ds^{*\prime}\, e^{-s'\partial_s} e^{-n_2'\partial_{n_2}} (N - n_2' \tag{64}$$
$$+ \partial_{s^*} s'^*)\, e^{-s'^*\partial_{s^*}} \delta(s' - S^-)\delta(n_2' - N_2)\delta(s'^* - S^+)\delta(s)\delta(n_2)\delta(s^*)\Big]$$

where we have introduced new variables s', n_2' , s'^* .

We note that the only operators in (64), besides $\sigma(t)$, are the three delta function terms so that, recognizing that the exponential derivative factors operate on the last three c-number delta functions, we get

$$T_1 = \left(\frac{\epsilon_1}{i\hbar} - \frac{\Gamma_1}{2}\right)\int ds'\,d\eta_2'\,ds'^* \left(N - \eta_2' + \partial_{s^*} s'^*\right)\delta(s-s')\,\delta(\eta_2-\eta_2')\,\delta(s^*-s'^*)$$

$$\mathrm{Tr}\left[\sigma(t)\,\delta(s'-S^-)\,\delta(\eta_2'-N_2)\,\delta(s'^*-S^+)\right]$$

$$= \left(\frac{\epsilon_1}{i\hbar} - \frac{\Gamma_1}{2}\right)\int ds'\,d\eta_2'\,ds'^* \left(N - \eta_2' + \partial_{s^*} s^{*\prime}\right)\delta(s-s')\,\delta(\eta_2-\eta_2')$$

$$\delta(s^*-s'^*)\;\underline{\rho}\,(s', \eta_2', s'^*, t) \tag{65}$$

$$= \left(\frac{\epsilon_1}{i\hbar} - \frac{\Gamma_1}{2}\right)\left(\eta_1 + \partial_{s^*} s^*\right)\underline{\rho}\,(s, \eta_2, s^*, t)$$

where we have called $\eta_1 = N - \eta_2$.

Using the same sort of manipulations, we may calculate the contributions of the following three terms:

$$T_2 = \left(\frac{\epsilon_2}{i\hbar} - \frac{\Gamma_2}{2}\right)\mathrm{Tr}\left[\sigma(t)\,\delta^c N_2\right] = \left(\frac{\epsilon_2}{i\hbar} - \frac{\Gamma_2}{2}\right)\left(\eta_2 - \partial_{s^*} s^*\right)\underline{\rho}\,(s, \eta_2, s^*, t) \tag{66}$$

$$T_3 = -\left(\frac{\epsilon_1}{i\hbar} + \frac{\Gamma_1}{2}\right)\mathrm{Tr}\left[\sigma(t) N_1 \delta^c\right] = -\left(\frac{\epsilon_1}{i\hbar} + \frac{\Gamma_1}{2}\right)\left(\eta_1 + \partial_s s\right)\underline{\rho}\,(s, \eta_2, s^*, t) \tag{67}$$

$$T_4 = -\left(\frac{\epsilon_2}{i\hbar} + \frac{\Gamma_2}{2}\right) Tr\left[\sigma(t)\, N_2\, \delta^c\right] \tag{68}$$

$$= -\left(\frac{\epsilon_2}{i\hbar} + \frac{\Gamma_2}{2}\right)\left(n_2 - \partial_\alpha \alpha\right) \rho(\alpha, n_2, \alpha^*, t)$$

We next consider the fifth term in (47):

$$T_5 = \Gamma_2\, Tr\left[\sigma(t) \sum_{j=1}^{N} (|2\rangle\langle 1|)_j\, \delta^c\, (|1\rangle\langle 2|)_j\right] \tag{69}$$

We use the displacement operation (56a)to write it as

$$T_5 = \Gamma_2\, Tr\left[\sigma(t) \sum_{j=1}^{N} (|2\rangle\langle 1|)_j\, e^{-S^-\partial_\alpha}\, e^{-N_2 \partial_{n_2}}\, e^{-S^+\partial_{\alpha^*}} (|1\rangle\langle 2|)_j\right] \delta(\alpha)\delta(n_2)\delta(\alpha^*) \tag{70}$$

Using the definitions (36) and (37), we write the exponential displacement factors out explicitly: the quantities within the brackets of (70) become

$$[\cdots] = \sigma(t) \sum_{j=1}^{N} (|2\rangle\langle 1|)_j\, e^{-\partial_\alpha \sum_p (|2\rangle\langle 1|)_p}\, e^{-\partial_{n_2} \sum_q (|2\rangle\langle 2|)_q}\, e^{-\partial_{\alpha^*} \sum_r (|1\rangle\langle 2|)_r} (|1\rangle\langle 2|)_j$$

$$= \sigma(t) \sum_{j=1}^{N} (|2\rangle\langle 1|)_j \prod_p e^{-\partial_\alpha (|2\rangle\langle 1|)_p} \prod_q e^{-\partial_{n_2} (|2\rangle\langle 2|)_q} \prod_r e^{-\partial_{\alpha^*} (|1\rangle\langle 2|)_r} (|1\rangle\langle 2|)_j \tag{71}$$

$$= \sigma(t) \prod_{p \neq j} T_p \sum_{j=1}^{N} \left[|2\rangle\langle 1|\, e^{-\partial_\alpha |2\rangle\langle 1|}\, e^{-\partial_{n_2} |2\rangle\langle 2|}\, e^{-\partial_{\alpha^*} |1\rangle\langle 2|}\, |1\rangle\langle 2|\right]_j$$

where

$$T_p = \left[e^{-\partial_\alpha |2\rangle\langle 1|}\, e^{-\partial_{n_2} |2\rangle\langle 2|}\, e^{-\partial_{\alpha^*} |1\rangle\langle 2|}\right]_p \tag{72}$$

We have used the fact that the atomic operators for different atoms commute so that all terms for $p \neq j$ can be pulled through the sum over j. The notation $[\cdots]_p$ denotes the operator expression within the brackets refers to the p^{th} atom. We next consider

$$t = e^{-|2\rangle\langle 2|\partial_{n_2}} |1\rangle\langle 2| e^{|2\rangle\langle 2|\partial_{n_2}} \tag{73}$$

in which the operators refer to a single atom. By expanding the exponential operators explicitly we see that higher powers of $|2\rangle\langle 2|$ are equal to $|2\rangle\langle 2|$, for example

$$(|2\rangle\langle 2|)^2 = |2\rangle\langle 2|2\rangle\langle 2| = |2\rangle\langle 2|$$

so that we have

$$t = \left[1 + |2\rangle\langle 2|(e^{-\partial_{n_2}} - 1)\right] |1\rangle\langle 2| \left[1 + |2\rangle\langle 2|(e^{\partial_{n_2}} - 1)\right] = e^{\partial_{n_2}} |1\rangle\langle 2| \tag{74}$$

Therefore

$$\begin{aligned}
\left[|2\rangle\langle 1| e^{-\partial_\alpha |2\rangle\langle 1|} e^{-\partial_{n_2}|2\rangle\langle 2|} e^{-\partial_{\alpha^*}|1\rangle\langle 2|} |1\rangle\langle 2|\right]_j &= \left[e^{-\partial_\alpha |2\rangle\langle 1|} |2\rangle\langle 1| e^{-\partial_{n_2}|2\rangle\langle 2|} |1\rangle\langle 2| e^{-\partial_{\alpha^*}|1\rangle\langle 2|}\right]_j \\
&= \left[e^{-\partial_\alpha |2\rangle\langle 1|} |2\rangle\langle 1| e^{-\partial_{n_2}|2\rangle\langle 2|} |1\rangle\langle 2| e^{\partial_{n_2}|2\rangle\langle 2|} e^{-\partial_{n_2}|2\rangle\langle 2|} e^{-\partial_{\alpha^*}|1\rangle\langle 2|}\right]_j \\
&= e^{\partial_{n_2}} \left[e^{-\partial_\alpha |2\rangle\langle 1|} |2\rangle\langle 2| e^{-\partial_{n_2}|2\rangle\langle 2|} e^{-\partial_{\alpha^*}|1\rangle\langle 2|}\right]_j \\
&= \left[e^{-\partial_\alpha |2\rangle\langle 1|} |2\rangle\langle 1|1\rangle\langle 2| e^{\partial_{n_2}} e^{-\partial_{n_2}|2\rangle\langle 2|} e^{-\partial_{\alpha^*}|1\rangle\langle 2|}\right]_j
\end{aligned} \tag{75}$$

We may use (75) in (71) and recognize that since the product over ρ does not include $\rho = j$, we may recombine the exponential operators and get

$$T_5 = e^{\partial n_2} \Gamma_2 \, T_r \left[\sigma(t) \, e^{-S^- \partial_s} N_2 \, e^{-N_2 \partial n_2} e^{-S^+ \partial_{s^*}} \right] \delta(s) \, \delta(n_2) \, \delta(s^*) \tag{76}$$

The operators in (76) are in the chosen order so that, using a manipulation as in (64) and (65), we obtain the final result:

$$T_5 = e^{\partial n_2} n_2 \, \Gamma_2 \, P(s, n_2, s^*, t) \tag{77}$$

The calculation of the last term in (47) is quite complicated due to the reordering of the operators:

$$T_6 = \Gamma_1 \, T_r \, \sigma(t) \sum_{j=1}^{N} (|1\rangle\langle 2|)_j \, \delta^{c}(|2\rangle\langle 1|)_j \tag{78}$$

Using the displacement operation (56) and the factorization as done in (71), this can be written as

$$T_6 = \Gamma_1 \, T_r \, \sigma(t) \prod_{\rho \neq j} T_\rho \sum_{j=1}^{N} \Big[|1\rangle\langle 2| \, e^{-\partial_s |2\rangle\langle 1|} \, e^{-\partial n_2 |2\rangle\langle 2|} \tag{79}$$

$$e^{-\partial_{s^*}|1\rangle\langle 2|}\,|2\rangle\langle 1|\Big]_j \,\delta(s)\,\delta(n_2)\,\delta(s^*)$$

By expansion of the exponential and orthogonality of the states

$$e^{\pm\partial_s|2\rangle\langle 1|} = \underline{1} \pm \partial_s|2\rangle\langle 1| \tag{80}$$

so that we may calculate

$$e^{\partial_s|2\rangle\langle 1|}\,|1\rangle\langle 2|\,e^{-\partial_s|2\rangle\langle 1|} = |1\rangle\langle 2| - |1\rangle\langle 1|\partial_s + |2\rangle\langle 2|\partial_s - \partial_s^2|2\rangle\langle 1| \tag{81}$$

Doing a similar treatment of the term on the right end of (79) we obtain the following expression for T_6 ;

$$\begin{aligned} T_6 = \Gamma_1 \, \mathrm{Tr}\Big\{ \sigma(t) \prod_{p\neq j} T_p \sum_{j=1}^{N} \Big[e^{-\partial_s|2\rangle\langle 1|} \Big(&|1\rangle\langle 2| - |1\rangle\langle 1|\partial_s \\ &+ |2\rangle\langle 2|\partial_s - \partial_s^2|2\rangle\langle 1|\Big) e^{-\partial_{n_2}|2\rangle\langle 2|} \Big(|2\rangle\langle 1| + |2\rangle\langle 2|\partial_{s^*} \\ &- |1\rangle\langle 1|\partial_{s^*} \\ &- \partial_{s^*}^2|1\rangle\langle 2|\Big) e^{-\partial_{s^*}|1\rangle\langle 2|} \Big]_j \Big\} \end{aligned} \tag{82}$$

It is clear that a total of sixteen terms result from multiplying out the factors in the brackets of (82). We have indicated the methods used to reorder the operator products and calculate the trace over $\sigma(t)$. For completeness we list the results for those terms individually in Appendix V. Here we quote the final result for this entire term:

$$T_6 = \Big\{ e^{-\partial n_2} n_1 + e^{-\partial n_2} \left(\partial_s s + \partial_{s^*} s^* \right) + \frac{\partial^2}{\partial s \partial s^*} N + \frac{\partial^3}{\partial s \partial s^{*2}} s^* + \frac{\partial^3}{\partial s^* \partial s^2} s + \frac{\partial^4}{\partial s^2 \partial s^{*2}} e^{\partial n_2} n_2 \Big\} \Gamma_1 \, \underline{\rho}(s, n_2, s^*, t) \tag{83}$$

Before combining the six terms computed, we rewrite slightly the following expressions which appear in T_5 and T_6:

$$e^{\partial n_2} n_2 \Gamma_2 = \left(e^{\partial n_2} - 1 \right) n_2 \Gamma_2 + n_2 \Gamma_2$$

$$e^{-\partial n_2} n_1 \Gamma_1 = \left(e^{-\partial n_2} - 1 \right) n_1 \Gamma_1 + n_1 \Gamma_1$$

$$e^{-\partial n_2} \Gamma_1 \partial_{s^*} s^* = \left(e^{-\partial n_2} - 1 \right) \Gamma_1 \partial_{s^*} s^* + \Gamma_1 \partial_{s^*} s^*$$

$$e^{-\partial n_2} \Gamma_1 \partial_s s = \left(e^{-\partial n_2} - 1 \right) \Gamma_1 \partial_s s + \Gamma_1 \partial_s s$$

Using these expressions in T_5 and T_6, and defining

$$\Gamma_{12} = \frac{1}{2} \left(\Gamma_1 + \Gamma_2 \right) \tag{84}$$

we combine the six terms to get the following equation of motion for the magnetization distribution function:

$$\begin{aligned}\dot{\underline{\rho}}(\alpha, n_2, \alpha^*, t) = \Big\{ & \frac{\partial}{\partial\alpha^*}(\Gamma_{12}+i\omega_{21})\alpha^* + \frac{\partial}{\partial\alpha}(\Gamma_{12}-i\omega_{21})\alpha \\ & + (e^{-\partial n_2}-1)\Gamma_1 n_1 + (e^{\partial n_2}-1)\Gamma_2 n_2 + \frac{\partial^2}{\partial\alpha\,\partial\alpha^*}\Gamma_1 N \\ & + \frac{\partial}{\partial\alpha^*}(e^{-\partial n_2}-1)\Gamma_1\alpha^* + \frac{\partial}{\partial\alpha}(e^{-\partial n_2}-1)\Gamma_1\alpha \\ & + \frac{\partial^3}{\partial\alpha\,\partial\alpha^{*2}}\Gamma_1\alpha^* + \frac{\partial^3}{\partial\alpha^2\partial\alpha^*}\Gamma_1\alpha \\ & + \frac{\partial^4}{\partial\alpha^2\partial\alpha^{*2}} e^{\partial n_2}\Gamma_1 n_2 \Big\} \underline{\rho}(\alpha, n_2, \alpha^*, t)\end{aligned} \tag{85}$$

This equation evidently couples the transverse components of the magnetization α, α^* to the upper state population (which is clearly proportional to the longitudinal magnetization). Its complication precludes any simple treatment, so instead of working with the full distribution function, we shall integrate over the transverse components in (85) to obtain an equation for the distribution function of the upper state population alone. Assuming that Γ_1 , Γ_2 , and ω_{21} are independent of α and α^* , we obtain

$$\begin{aligned}\dot{\underline{\rho}}(n_2, t) &= \int d\alpha\, d\alpha^*\, \dot{\underline{\rho}}(\alpha, n_2, \alpha^*, t) \\ &= \Big\{(e^{-\partial n_2}-1)\Gamma_1 n_1 + (e^{\partial n_2}-1)\Gamma_2 n_2\Big\}\underline{\rho}(n_2, t)\end{aligned} \tag{86}$$

Since the other terms involve derivatives with respect to the transverse variables, they may be immediately integrated.

We have assumed that the distribution function vanishes when evaluated at the limits of integration for the transverse components (which lie at infinity).

It may be noted that (86) might have been obtained directly from (35) if we had defined

$$\underline{\rho}\,(n_2, t) = Tr\left[\sigma(t)\;\delta(n_2 - N_2)\right] \tag{87}$$

and proceeded to compute its equation of motion using the techniques described above. In this way we of course lose all information about the transverse components, and furthermore it is not a priori obvious that (35), which does involve three basic operators, is of a form such that using a one variable distribution function as (87) will result in an equation in terms of only that single variable.

In view of our original definition of the atomic variables in (31), we may define the (dimensionless) longitudinal component of the magnetization by

$$M_z = \sum_{j=1}^{N} S_j^{z} = \tfrac{1}{2}\left(N_1 - N_2\right) \tag{87}$$

We shall denote by m the c-number variable corresponding to the operator M_z. Thus

$$m = \tfrac{1}{2}(n_1 - n_2)\;;\quad \partial_m = -\partial n_2 \tag{88}$$

and (86) yields

$$\dot{\rho}(m,t) = \left\{ \frac{N}{2}\left[(e^{-\partial_m}-1)\Gamma_2 + (e^{\partial_m}-1)\Gamma_1\right] + \left[(e^{\partial_m}-1)\Gamma_1 - (e^{-\partial_m}-1)\Gamma_2\right] m \right\} \rho(m,t) \tag{89}$$

This then is the final form of the equation of motion for the magnetization distribution function.

Part B. Section III. Critical Fluctuations

A useful approximation to the full equation of motion (89) for the longitudinal component of the magnetization may be made as follows: there is ample evidence[43,44,45] that large fluctuations exist in the magnetization for temperatures near the Curie temperature T_c above which the system is paramagnetic. This means that near T_c the function $\rho(m)$ of (89) should be a relatively slowly varying function of m so as to increase the fluctuations, which the mean square fluctuation

$$\sigma_m^2 = \langle m^2\rangle - \langle m\rangle^2 \tag{90}$$

is one measure. Therefore, in this region we expect that the higher order derivatives, obtained by expanding the exponential operators in (89), will rapidly become smaller than the lowest order terms. It is thus reasonable to approximate the complete equation by retaining only the lowest order terms.

Expanding the exponential factors up to second derivatives only, we obtain from (89)

$$\begin{aligned}\dot{\rho}(m,t) = \Big\{ &\frac{\partial}{\partial m}\Big[\frac{N}{2}\big(\Gamma_1(m) - \Gamma_2(m)\big) + \big(\Gamma_1(m) + \Gamma_2(m)\big)m\Big] \\ &+ \frac{1}{2}\frac{\partial^2}{\partial m^2}\Big[\frac{N}{2}\big(\Gamma_1(m) + \Gamma_2(m)\big) + \big(\Gamma_1(m) - \Gamma_2(m)\big)m\Big]\Big\}\rho(m,t)\end{aligned} \tag{91}$$

Equations of the form

$$\frac{\partial}{\partial t} F(x,t) = \left\{ -\frac{\partial}{\partial x} A + \frac{\partial^2}{\partial x^2} B \right\} F(x,t) \tag{92}$$

are referred to as Fokker-Planck equations[46] with drift coefficient A and diffusion coefficient B. It is seen that our equation is of this general type, but with non-constant drift and diffusion coefficients. From the definition

$$\langle \dot{m}(t) \rangle = \int dm \; m \; \dot{\underline{\rho}}(m,t) \tag{93}$$

we obtain from equation (91) the equation of motion for the average magnetization:

$$\langle \dot{m}(t) \rangle = \left\langle \frac{N}{2} \left[\Gamma_2(m) - \Gamma_1(m) \right] \right\rangle - \left\langle \left[\Gamma_1(m) + \Gamma_2(m) \right] m \right\rangle \tag{94}$$

Here we have integrated by parts and assumed that the distribution function and its derivative vanish at the extreme limits of integration.

To proceed further we must know what the functions Γ_1 and Γ_2 are. Although, as stated in (27) and demonstrated in Appendix IV, the ratio of these quantities is known from the general forms (25) and (26), their explicit forms depend upon the details of the reservoir chosen to represent the thermal bath to which the ferromagnetic spin system is coupled. We therefore introduce two specific reservoir models and study the dynamical properties of the spin system as it interacts with each one in turn.

(a) We first consider the reservoir to be composed of an infinite array of harmonic oscillators of a broad distribution of frequencies whose density operator is the canonical ensemble thermal equilibrium density operator. This reservoir coincides with that chosen in Part A, Section III above. Thus, we identify the reservoir operators O_α , O^+_α of (4) with the boson absorbtion and creation operators b_j, b^+_j which obey the commutation relations

$$[b_j , b^+_{j'}] = \delta_{jj'} \; ; \; [b_j , b_{j'}] = [b^+_j , b^+_{j'}] = 0 \tag{95}$$

Thus the reservoir Hamiltonian is

$$H_R = \sum_j{}' \hbar\omega_j \, b^+_j b_j \tag{96}$$

Taking the thermal equilibrium reservoir density operator (16) and the general formula (25) for Γ_2 , we have

$$\begin{aligned}
\tfrac{1}{2}\Gamma_2(\omega_{21}) &= g^2 R_e \int_{-\infty}^{t} dt' \left\langle \sum_\alpha O_\alpha(t) \sum_\beta O^+_\beta(t') \right\rangle e^{i\omega_{21}(t-t')} \\
&= g^2 R_e \int_{-\infty}^{t} dt' \, Tr_R \Big[f_0(H_R) \sum_j b_j e^{-i\omega_j t} \sum_k b^+_k e^{i\omega_k t'} \Big] e^{i\omega_{21}(t-t')} \\
&= g^2 R_e \int_{-\infty}^{t} dt' \sum_j{}' \langle b_j b^+_j \rangle \, e^{i(\omega_{21}-\omega_j)(t-t')} \\
&= g^2 R_e \int_{-\infty}^{t} dt' \sum_j \left[\bar{n}(\omega_j) + 1 \right] e^{i(\omega_{21}-\omega_j)(t-t')}
\end{aligned} \tag{97}$$

where

$$\bar{n}(\omega_j) = \left[e^{\beta\hbar\omega_j} - 1\right]^{-1} \tag{98}$$

is the average thermal occupation number of phonons in the j^{th} mode. We assume that the number of degrees of freedom (modes) of the reservoir is large enough to enable us to convert the sum in (97) into an integral by means of a density of states function $\Omega(\omega_j)$:

$$\begin{aligned} \tfrac{1}{2}\Gamma_2(\omega_{21}) &= g^2 \,\mathrm{Re} \int_{-\infty}^{t} dt' \int_{0}^{\infty} d\omega_j \,\Omega(\omega_j)\left[\bar{n}(\omega_j)+1\right] e^{i(\omega_{21}-\omega_j)(t-t')} \\ &= g^2 \,\mathrm{Re} \int_{-\infty}^{t} \int_{\omega_{21}}^{-\infty} (-du)\,\Omega(\omega_{21}-u)\left[\bar{n}(\omega_{21}-u)+1\right] e^{iu(t-t')} \\ &\simeq g^2 \,\mathrm{Re} \int_{-\infty}^{t} dt' \int_{-\infty}^{\infty} du\,\Omega(\omega_{21}-u)\left[\bar{n}(\omega_{21}-u)+1\right] e^{iu(t-t')} \end{aligned} \tag{99}$$

Arguing that the density of states function and the thermal occupation number are slowly varying functions of their arguments compared to the exponential function in (99), we approximately calculate the integral by extracting those two functions from under the integral sign - evaluated at the point of least rapid variation of the exponential, which occurs for $u = 0$.

$$\Gamma_2(\omega_{21}) = 2g^2 \, Re \int_{-\infty}^{t} dt' \, \Omega(\omega_{21}) \left[\bar{n}(\omega_{21}) + 1\right] \int_{-\infty}^{\infty} du \, e^{iu(t-t')}$$

$$= 2\pi g^2 \, \Omega(\omega_{21}) \left[\bar{n}(\omega_{21}) + 1\right]$$

$$= \gamma^{(a)} \left[\bar{n}(\omega_{21}) + 1\right] \tag{100}$$

where we denote by $\gamma^{(a)}$ the spontaneous phonon emission rate.

We obtain the Γ_1 in a similar calculation from its defining relation (26):

$$\Gamma_1(\omega_{21}) = 2g^2 \, Re \int_{-\infty}^{t} dt' \left\langle \sum_{\alpha} O^{+}_{\alpha}(t) \sum_{\beta} O_{\beta}(t') \right\rangle e^{-i\omega_{21}(t-t')}$$

$$= 2g^2 \, Re \int_{-\infty}^{t} dt' \, Tr\left[f_0(H_R) \sum_j b_j^{+} e^{i\omega_j t} \sum_{k} b_k e^{-i\omega_k t'}\right] e^{-i\omega_{21}(t-t')}$$

$$= 2g^2 \, Re \int_{-\infty}^{t} dt' \, \sum_j \langle b_j^{+} b_j \rangle \, e^{i(\omega_j - \omega_{21})(t-t')}$$

$$= 2g^2 \, Re \int_{-\infty}^{t} dt' \int_{0}^{\infty} d\omega_j \, \Omega(\omega_j) \, \bar{n}(\omega_j) \, e^{i(\omega_j - \omega_{21})(t-t')} \tag{101}$$

$$= 2g^2 \, Re \int_{-\infty}^{t} dt' \, \Omega(\omega_{21}) \, \bar{n}(\omega_{21}) \int_{-\infty}^{\infty} du \, e^{iu(t-t')}$$

$$= 2\pi g^2 \, \Omega(\omega_{21}) \, \bar{n}(\omega_{21})$$

$$= \gamma^{(a)} \, \bar{n}(\omega_{21})$$

We may now return to the equation of motion for the magnetization distribution function. However, first we note that the molecular field approximation yields the following connection between the two level energy separation $\hbar\omega_{21}$ and the magnetization:

$$\hbar\omega_{21} = 2\mu\left(H_0 + \lambda M\right) \tag{102}$$

where H_0 is the uniform external field, λ is the Weiss internal field constant, and M is the (correctly dimensioned) magnetization given by

$$M = \frac{2\mu}{V}\, m \tag{103}$$

where μ is the magnetic moment of each spin and V is the volume of the system. Using these relations in (91) we obtain

$$\begin{aligned}\dot{\underline{\rho}}^{(a)}(M,t) = \Big\{ &\frac{\partial}{\partial M}\gamma^{(a)}\left[-\frac{N\mu}{V} + M \coth\left[\beta\mu(H_0+\lambda M)\right]\right] \\ &+ \frac{\mu}{V}\frac{\partial^2}{\partial M^2}\gamma^{(a)}\left[\frac{N\mu}{V}\coth\left[\beta\mu(H_0+\lambda M)\right] - M\right]\Big\}\,\underline{\rho}(M,t)\end{aligned} \tag{104}$$

We have used (98) to obtain

$$\begin{aligned}\Gamma_1(\omega_{21}) - \Gamma_2(\omega_{21}) &= -1 \\ \Gamma_1(\omega_{21}) + \Gamma_2(\omega_{21}) &= \coth\left[\tfrac{1}{2}\beta\hbar\omega_{21}\right]\end{aligned} \tag{105}$$

From (104) we see that the mean magnetization satisfies the equation of motion

$$\frac{1}{\gamma^{(a)}}\frac{d}{dt}\langle M(t)\rangle = \frac{N\mu}{V} - \langle M(t)\rangle \coth\left[\beta\mu\left(H_0 + \lambda\langle M(t)\rangle\right)\right] \tag{106}$$

where we have neglected fluctuation effects in this equation by writing the average of a product of M(t) factors as the product of the average value $\langle M(t)\rangle$. We note at once that the steady state (time independent) solution of (131) yields the molecular field equation of state:

$$\begin{aligned} 0 &= \frac{N\mu}{V} - \langle M\rangle \coth\left[\beta\mu\left(H_0 + \lambda\langle M\rangle\right)\right] \\ \langle M\rangle &= \frac{N\mu}{V} \tanh\left[\beta\mu\left(H_0 + \lambda\langle M\rangle\right)\right] \end{aligned} \tag{107}$$

Equation (106) is difficult to solve exactly, but if we assume that the magnetization's time rate of change is small, as it may be for small perturbations away from the equilibrium value given by (107), and that the equilibrium value itself is small compared to the saturation value (as it would be at temperatures near T_c), we may obtain an approximate solution. Equation (106) may be rewritten as

$$\tanh\left[\beta\mu\left(H_0 + \lambda\langle M(t)\rangle\right)\right] = \frac{\langle M(t)\rangle}{\dfrac{N\mu}{V} - \dfrac{1}{\gamma^{(a)}}\dfrac{d}{dt}\langle M(t)\rangle} \tag{108}$$

Defining dimensionless variables by

$$x(t) = \frac{\langle M(t) \rangle}{(N\mu / v)} \; ; \quad h = \frac{H_0}{\lambda (N\mu / v)} \; ; \quad \tau = \frac{T_c}{T} \tag{109}$$

where the transition temperature T_c is defined by

$$K_B T_c = \frac{\lambda N \mu^2}{v} \tag{110}$$

and using the following expansion of the inverse hyperbolic tangent function for small values of its argument

$$\tanh^{-1} u \simeq u + \frac{u^3}{3} + O(u^5) \tag{111}$$

we obtain from (108)

$$\tau (h + x) \simeq x + \frac{1}{3} x^3 + x \Delta \tag{112}$$

where

$$\Delta = \frac{1}{\gamma^{(a)}} \frac{d}{dt} x(t) \tag{113}$$

and we have kept only the lowest order term in the product of the two small quantities x and Δ. Setting the external field to zero and solving for Δ, we find that the resulting equation may be readily integrated to give

$$\frac{dx}{(\tau - 1) - \frac{1}{3} x^2} = \gamma^{(a)} dt = \sqrt{\frac{3}{\tau - 1}} \; \frac{d}{dx} \; \mathrm{Tanh}^{-1} \left[\frac{x}{\sqrt{3(\tau - 1)}} \right]$$

$$\tanh^{-1}\frac{x}{\sqrt{3(\tau-1)}} - \tanh^{-1}\frac{x_0}{\sqrt{3(\tau-1)}} = \sqrt{\frac{3}{\tau-1}}\,\gamma^{(a)}t$$

$$x(t) = \sqrt{3(\tau-1)}\left[\frac{\dfrac{x_0}{\sqrt{3(\tau-1)}} + \tanh\left[\sqrt{\dfrac{3}{\tau-1}}\,\gamma^{(a)}t\right]}{1 + \dfrac{x_0}{\sqrt{3(\tau-1)}}\tanh\left[\sqrt{\dfrac{3}{\tau-1}}\,\gamma^{(a)}t\right]}\right] \tag{114}$$

Here x_0 is determined via (109) from the initial value of the magnetization $\langle M(0)\rangle$, and we have assumed that $\gamma^{(a)}$ is a constant. Note that (114) reduces to the correct form (as obtained by expanding the molecular field equation of state (107) near T_c) for $t \to +\infty$.

Returning to (104), which we write in terms of the dimensionless variables (109) as

$$\dot{P}^{(a)}(x,t) = \left\{\frac{\partial}{\partial x}\gamma^{(a)}\Big(x\coth[\tau(h+x)] - 1\Big) + \frac{1}{N}\frac{\partial^2}{\partial x^2}\gamma^{(a)}\Big(\coth[\tau(h+x)] - x\Big)\right\}P^{(a)}(x,t) \tag{115}$$

we may easily obtain its steady state (time independent) solution. Calling the drift and diffusion coefficients of (115) A and B respectively, as in (92), we see that the steady-state solution $P^{(a)}(x)$ satisfies

$$-\frac{\partial}{\partial x}\left[A P^{(a)}(x)\right]+\frac{\partial^2}{\partial x^2}\left[B P^{(a)}(x)\right]=0=\frac{\partial}{\partial x}\left[-A P^{(a)}(x)+\frac{\partial}{\partial x}\,B P^{(a)}(x)\right] \tag{116}$$

We note that the Fokker-Planck equation in general has the form of a conservation of probability law:

$$\frac{\partial P}{\partial t}+\frac{\partial J}{\partial x}=0 \tag{117}$$

Here J is a probability current. At steady state, provided we are restricted to a finite range of the variable (in our case $|x| \leq 1$), we must choose J=0 in order not to violate this general conservation requirement[46]. Thus, the integration constant from (116) must vanish and we have

$$\left[-\frac{A}{B}+\frac{\partial}{\partial x}\right]U(x)=0 \tag{118}$$

where

$$U(x)=B\,P^{(a)}(x) \tag{119}$$

Changing variables to

$$W(x)=\exp\left[K\int_0^x \frac{A(x')}{B(x')}\,dx'\right]U(x) \tag{120}$$

we find upon substitution into (118) that it is convenient to set $K=-1$ and so obtain the steady-state solution:

$$\rho^{(a)}(x) = \frac{C}{B(x)} \exp\left[\int_0^x dx' \, \frac{A(x')}{B(x')}\right] \tag{121}$$

$$= \frac{NC\left[\gamma^{(a)}\right]^{-1}}{\coth\left[\tau(h+x)\right]-x} \exp\left\{-N\int_0^x dx'\left[\frac{x'\coth\left[\tau(h+x')\right]-1}{\coth\left[\tau(h+x')\right]-x'}\right]\right\} \tag{122}$$

where C is a normalization constant (depending upon N, h, and τ). We note that we may write (122) in the form

$$\rho^{(a)}(x) = C\exp -N\left\{\int_0^x dx' \, \frac{x'\coth\left[\tau(h+x')\right]-1}{\coth\left[\tau(h+x')\right]-x'} - \frac{1}{N}\ln\left[\gamma^{(a)}\left(\coth\left[\tau(h+x)\right]-x\right)\right]\right\} \tag{123}$$

Since N is an extremely large number ($N \sim 10^{23}$), it is a reasonable approximation to neglect the second term in (148) in comparison with the first, so we take as the steady-state solution to (115)

$$\rho^{(a)}(x) = C\exp -N\int_0^x dx' \, \frac{x'\coth\left[\tau(h+x')\right]-1}{\coth\left[\tau(h+x')\right]-x'} \tag{124}$$

We note that the first derivative of this function vanishes at points $x = x_0$ where x_0 satisfies the molecular field equation of state (107). Because of the great numerical value of N, the peak of the maximum is extremely sharp for low temperatures. However, since we neglected higher derivatives in obtaining our Fokker-Planck equation, this solution is not expected to have any validity in that region where it is so sharply peaked. However, for temperatures near T_c ($\tau \sim 1$), we may expand the solution (124) for small x (since the peak of the distribution occurs at the molecular field value of x, which will be small near T_c) to obtain (for zero external field)

$$\underline{\rho}^{(a)}(x) \simeq C \exp - N \int_0^x dx' \left([(1-\tau)] x' + [\tau(1-\tau) + \tfrac{1}{3}\tau^3] x'^3 \right) \tag{125}$$

$$= C \exp \left\{ -\frac{1}{K_B T} \left[c(T - T_c) \frac{m^2}{2} + d' T \frac{m^4}{4} \right] \right\}$$

where

$$c = \frac{K_B}{N\mu^2} \; ; \quad d' = [\tau^3 + 3\tau(1-\tau)] d \; ; \quad d = \frac{K_B}{3N^3\mu^4} \tag{126}$$

The thermodynamic theory of fluctuations [18,47] predicts that the probability density for the fluctuations of a thermodynamic variable η is given by

$$\underline{\rho}(\eta) = C e^{-\frac{\phi(\eta)}{K_B T}} \tag{127}$$

where $\phi(\eta)$ is the appropriate thermodynamic potential for the situation considered. Besides having this general form, (125) also has the form for the free energy which the Landau theory of a second order phase transition [18] predicts:

namely, Landau theory predicts that near such a phase transition $\phi(\eta)$ is of the form

$$\phi(\eta) = \phi_0 + \alpha(T-T_c)\eta^2 + \Delta\eta^4 \tag{128}$$

where η is the order parameter of the transition. That is, η is a thermodynamic variable which is assumed to vanish above the transition temperature T_c (corresponding to the disordered state) and be non-zero below T_c (in the ordered state). In the case of a ferromagnet the order parameter is the magnetic moment m . The precise values of the constants α and Δ (α is independent of temperature while Δ may be a function of temperature) are not predicted by the Landau theory.

However, in fact the constants c and d are precisely those which come from expanding the free energy which leads one to the molecular field equation of state. The appropriate free energy is that obtained from the Bragg-Williams[19] approximation, which corresponds to making a mean field approximation. Thus, the solution of the Fokker-Planck equation (104), or (115), for the distribution function of the magnetization yields the correct free energy of the system as its steady-state value in the region where this approximation to the exact equation (89) is expected to be valid.

(b) We turn next to a different model for the thermal reservoir with which the ferromagnetic system is in contact. For the reservoir we take an assembly of two level systems possessing a broad distribution of energy level splittings $\hbar\omega_K$. The reservoir Hamiltonian H_R now becomes

$$H_R = \sum_K{}' \hbar\omega_K S_K^z \tag{129}$$

where the spin one half angular momentum operators of the reservoir obey the usual commutation relations

$$[S_K^+, S_{K'}^z] = -\delta_{KK'} S_K^+ \tag{130}$$

$$[S_K^-, S_{K'}^z] = \delta_{KK'} S_K^- \tag{131}$$

$$S_K^+ = S_K^x + iS_K^y ; \quad S_K^- = S_K^x - iS_K^y \tag{132}$$

Once again it is convenient to relate these to atomic operators via

$$S_K^+ = (|2\rangle\langle 1|)_K ; \quad S_K^- = (|1\rangle\langle 2|)_K \tag{133}$$

where the energies of the two states obey

$$\epsilon_2(K) - \epsilon_1(K) = \hbar\omega_K \geq 0 \tag{134}$$

We thus identify the reservoir operators O_α, O_α^+ of (4) with $\mathcal{S}_K^-$, $\mathcal{S}_K^+$ and take the reservoir density operator $f_o(H_R)$ to be

$$f_o(H_R) = e^{-\beta H_R}\left[\mathrm{Tr}_R\, e^{-\beta H_R}\right]$$

$$= \prod_K \frac{1}{Z_K}\, \exp\left\{-\beta\left[\epsilon_1^{(K)}(|1\rangle\langle 1|)_K + \epsilon_2(K)(|2\rangle\langle 2|)_K\right]\right\} \tag{135}$$

where

$$Z_K = e^{-\beta\epsilon_1(K)} + e^{-\beta\epsilon_2(K)} \tag{136}$$

Thus, from (25) we may calculate Γ_2 :

$$\Gamma_2(\omega_{21}) = 2g^2\, \mathrm{Re} \int_{-\infty}^{t} dt' \left\langle \sum_K \mathcal{S}_K^-\, e^{-i\omega_K t} \sum_{K'} \mathcal{S}_{K'}^+\, e^{i\omega_{K'} t} \right\rangle e^{i\omega_{21}(t-t')}$$

$$= 2g^2\, \mathrm{Re} \int_{-\infty}^{t} dt' \sum_K \left\langle \mathcal{S}_K^-\, \mathcal{S}_K^+ \right\rangle e^{i(\omega_{21}-\omega_K)(t-t')} \tag{137}$$

$$= 2g^2\, \mathrm{Re} \int_{-\infty}^{t} dt' \sum_K \left\langle (|1\rangle\langle 1|)_K \right\rangle e^{i(\omega_{21}-\omega_K)(t-t')}$$

$$= 2g^2\, \mathrm{Re} \int_{-\infty}^{t} dt' \sum_K \bar{n}_1(\omega_K)\, e^{i(\omega_{21}-\omega_K)(t-t')}$$

$$= 2g^2\, \mathrm{Re} \int_{-\infty}^{t} dt' \int_0^{\infty} d\omega_K\, \Omega(\omega_K)\, \bar{n}_1(\omega_K)\, e^{i(\omega_{21}-\omega_K)(t-t')}$$

where

$$\bar{n}_1(\omega_K) = \frac{e^{-\beta\epsilon_2(K)}}{e^{-\beta\epsilon_1(K)} + e^{-\beta\epsilon_2(K)}} = \frac{1}{1+e^{\beta\hbar\omega_K}} \tag{138}$$

Once again assuming that the reservoir density of states $\Omega(\omega_K)$ and $\bar{n}_1(\omega_K)$ are slowly varying in comparison with the exponential function in (137), we obtain

$$\begin{aligned} \Gamma_2(\omega_{21}) &= 2\pi g^2 \Omega(\omega_{21})\, \bar{n}_1(\omega_{21}) \\ &= \gamma^{(b)}\, \bar{n}_1(\omega_{21}) \end{aligned} \tag{139}$$

where $\gamma^{(b)}$ is the spontaneous decay rate.

In a similar fashion we may obtain Γ_1 from (26):

$$\begin{aligned} \Gamma_1(\omega_{21}) &= 2g^2\,\mathrm{Re}\int_{-\infty}^{t} dt' \sum_K \langle S_K^+ S_K^- \rangle\, e^{i(\omega_K-\omega_{21})(t-t')} \\ &= 2g^2\,\mathrm{Re}\int_{-\infty}^{t} dt' \Big\langle \sum_K S_K^+ e^{i\omega_K t} \sum_{K'} S_{K'}^- e^{-i\omega_{K'} t'} \Big\rangle e^{-i\omega_{21}(t-t')} \\ &= 2g^2\,\mathrm{Re}\int_{-\infty}^{t} dt' \sum_K \langle (|2\rangle\langle 2|)_K \rangle\, e^{i(\omega_K-\omega_{21})t-t')} \\ &= 2g^2\,\mathrm{Re}\int_{-\infty}^{t} dt' \int_0^{\infty} d\omega_K\, \Omega(\omega_K)\, \bar{n}_2(\omega_K)\, e^{i(\omega_K-\omega_{21})(t-t')} \end{aligned} \tag{140}$$

where

$$\bar{n}_1(\omega_K) = \frac{e^{-\beta\epsilon_2(K)}}{e^{-\beta\epsilon_1(K)} + e^{-\beta\epsilon_2(K)}} = \frac{1}{1 + e^{-\beta\hbar\omega_K}} \tag{141}$$

Thus we obtain from (140)

$$\begin{aligned}\Gamma_1(\omega_{21}) &= 2\pi g^2\, \Omega(\omega_{21})\, \bar{n}_2(\omega_{21}) \\ &= \gamma^{(b)}\, \bar{n}_2(\omega_{21})\end{aligned} \tag{142}$$

Returning to the general equation of motion for the magnetization distribution function (91), we obtain for this spin reservoir, using (139) and (142) and the molecular field approximation (102) for $\hbar\omega_{21}$:

$$\begin{aligned}\dot{\rho}^{(b)}(M,t) = \Big\{ &\frac{\partial}{\partial M}\gamma^{(b)}\Big[M - \frac{N\mu}{V}\tanh\big[\beta\mu(H_0 + \lambda M)\big]\Big] \\ &+ \frac{\mu}{V}\frac{\partial^2}{\partial M^2}\gamma^{(b)}\Big[\frac{N\mu}{V} - M\tanh\big[\beta\mu(H_0 + \lambda M)\big]\Big]\Big\}\rho^{(b)}(M,t)\end{aligned} \tag{143}$$

where we have used (103) to define the magnetization M and the results for this reservoir

$$\Gamma_1(\omega_{21}) - \Gamma_2(\omega_{21}) = -\tanh\left[\tfrac{1}{2}\beta\hbar\omega_{21}\right] \tag{144}$$

$$\Gamma_1(\omega_{21}) + \Gamma_2(\omega_{21}) = 1 \tag{145}$$

We see at once from (143) that the equation of motion for the average magnetization is given for this case by

$$\frac{1}{\gamma^{(b)}} \frac{\partial}{\partial t} \langle M(t)\rangle = \frac{N\mu}{V} \tanh\left[\beta\mu\left(H_0 + \lambda\langle M(t)\rangle\right)\right] - \langle M(t)\rangle \tag{146}$$

where we have neglected fluctuation effects in this mean equation of motion by writing the average of a product of factors of M(t) as the product of the average value $\langle M(t)\rangle$. We see immediately that the steady state solution of (146) is the molecular field equation of state (107). Therefore, although the equation of motion for the average magnetization for this thermal reservoir is different from the corresponding equation of motion for the phonon reservoir, equation (106), both equations have the same steady-state solution.

In terms of the dimensionless variables x, h and τ defined by (109), the Fokker-Planck equation (143) may be written

$$\dot{\rho}^{(b)}(x,t) = \Big\{ \gamma^{(b)}\left[x - \tanh\left[\tau(h+x)\right]\right] + \frac{1}{N}\frac{\partial^2}{\partial x^2}\gamma^{(b)}\left[1 - x\tanh\left[\tau(h+x)\right]\right]\Big\}\rho^{(b)}(x,t) \tag{147}$$

The steady-state solution, from (121), is therefore

$$\rho^{(b)}(x) = \frac{N\,C\left[\gamma^{(b)}\right]^{-1}}{1 - x\tanh[\tau(h+x)]}\,\exp - N\int_0^x dx'\left[\frac{x' - \tanh[\tau(h+x')]}{1 - x'\tanh[\tau(h+x')]}\right] \tag{148}$$

where C is a normalization constant. We write (148) in the form

$$\rho^{(b)}(x) = C\exp - N\left\{\int_0^x dx'\,\frac{x' - \tanh[\tau(h+x')]}{1 - x'\tanh[\tau(h+x')]} - \frac{1}{N}\ln\left[\gamma^{(b)}\left(1 - x\tanh[\tau(h+x)]\right)\right]\right\} \tag{149}$$

Since N is such an enormous number ($\sim 10^{23}$), we may neglect the second term in comparison to the first one in (149). Thus, we take as the steady-state solution to the equation of motion of the magnetization distribution function, equation (143) or (147),

$$\rho^{(b)}(x) = C\exp - N\int_0^x dx'\left[\frac{x' - \tanh[\tau(h+x')]}{1 - x'\tanh[\tau(h+x')]}\right] \tag{150}$$

We note that, although the equations of motion for the magnetization distribution function are different for the two types of reservoirs, the steady-state solutions are in fact the same, as comparison of (150) with (124) immediately shows. Thus, although the time-dependent properties of the ferromagnetic spin system depend upon the detailed nature of the thermal reservoir with which it is in contact, the steady-state properties are the same. The mean magnetization at steady-state for both types of reservoirs satisfies the molecular field equation of state, and the steady-state magnetization distributions agree with the Landau form of the magnetic free energy for temperatures near the Curie temperature, and identically with the Bragg-Williams free energy when it is approximated near the transition temperature.

If one could show that the dielectric constant fluctuations in our ferromagnetic model were proportional to the magnetization fluctuations (as one might expect, for example, in thinking of the Raman scattering of light by spin flip processes - light being down-shifted in frequency and a single spin flipped from $|1\rangle$ to $|2\rangle$ and the converse process for the anti-Stokes component - in which the Stokes intensity would be proportional to the average number of spins in state $|1\rangle$ while the anti-Stokes intensity would be proportional to the average number of spins in $|2\rangle$, both average values expressable in terms of the magnetization), one would not expect that the scattered light would have Gaussian statistics at temperatures near T_c as the distributions for the magnetization deviate from the Gaussian form.

Part B. Section IV. Discrete Representation and Bragg-Williams Free Energy

As we have noted earlier, exponential operators such as those found in the full equation of motion for the magnetization distribution function, equation (89), are in effect displacement operators acting on the functions to their right. Thus, we may let the displacement operators in (89) act on the functions of the variable m that are placed on their right side. Noting that these include the damping functions $\Gamma_1(m)$ and $\Gamma_2(m)$, and the factor m itself as well as $P(m,t)$ we carry out the displacements to get

$$\dot{P}(m,t) = \Gamma_2(m-1)\left[\frac{N}{2}-(m-1)\right]P(m-1,t) - \Gamma_2(m)\left[\frac{N}{2}-m\right]P(m,t)$$
$$-\Gamma_1(m)\left[\frac{N}{2}+m\right]P(m,t) + \Gamma_1(m+1)\left[\frac{N}{2}+m+1\right]P(m+1,t) \qquad (151)$$

We have indicated explicitly that Γ_1 and Γ_2 depend on the variable m, since, from their definitions (25) and (26), they are shown to depend on the energy difference between the two spin states. This splitting in turn, via our molecular field approximation to the Ising Hamiltonian in (9) depends upon the effective field at the spin site and so upon the resultant field produced by all of the aligned magnetic moments as well as the external field H_0. The displacement operators in (89) thus act on Γ_1 and Γ_2 also and yield (151).

The solution of (151) corresponding to steady-state ($\rho(m)$ independent of time) may easily be found to be

$$\frac{\rho(m)}{\rho(m-1)} = \frac{\Gamma_2(m-1)}{\Gamma_1(m)}\left[\frac{\frac{N}{2}-(m-1)}{\frac{N}{2}+m}\right] \tag{152a}$$

$$\frac{\rho(m+1)}{\rho(m)} = \frac{\Gamma_2(m)}{\Gamma_1(m+1)}\left[\frac{\frac{N}{2}-m}{\frac{N}{2}+m+1}\right] \tag{152b}$$

where (152a) corresponds to the balance of the first and third terms of (151) and (152b) to the balance of the second and fourth terms. It is seen that (152a) and (152b) are identical in fact.

We imagine that m is a discrete variable which, by (88) and the fixed total number of spins, obeys the inequality

$$-\frac{N}{2} \leq m \leq +\frac{N}{2} \tag{153}$$

We first consider the case $m > 0$. Thus, (152a) gives

$$\rho(m) = e^{\beta(Jm+\mu H_0-\frac{J}{2})}\left[\frac{\frac{N}{2}-(m-1)}{\frac{N}{2}+m}\right]\rho(m-1) \tag{154}$$

where we have used the detailed balance relation (27) connecting Γ_1 and Γ_2:

$$\frac{\Gamma_2(m-1)}{\Gamma_1(m)} = e^{\beta[E(m-1)-E(m)]} \tag{155}$$

where the energy of the system E(m) is found from the molecular field approximation to the Ising Hamiltonian to be

$$E(m) = -\frac{1}{2} J m^2 - \mu H_0 m \tag{156}$$

Here we have for simplicity taken the exchange energy J_{ij} of (2) to be independent of i and j and equal to J.

Thus, using (154) repeatedly we obtain

$$\rho(m) = e^{\beta[Jm+\mu H_0 - \frac{J}{2}]}\, e^{\beta[J(m-1)+\mu H_0 - \frac{J}{2}]} \cdots e^{\beta[J+\mu H_0 - \frac{J}{2}]}$$

$$\left[\frac{\frac{N}{2}-(m-1)}{\frac{N}{2}+m}\right]\left[\frac{\frac{N}{2}-(m-2)}{\frac{N}{2}+m-1}\right]\cdots\left[\frac{\frac{N}{2}-0}{\frac{N}{2}+1}\right]\rho_0$$

where ρ_0 is the value of $\rho(m=0)$. Thus

$$\rho(m) = e^{\beta J(\sum_{n=1}^{m} n)}\, e^{\beta(\mu H_0 - \frac{J}{2})m}\, \frac{(\frac{N}{2})!\,(\frac{N}{2})(\frac{N}{2}-1)\cdots[\frac{N}{2}-(m-1)][\frac{N}{2}-m]!\,\rho_0}{[\frac{N}{2}+m]\cdots[\frac{N}{2}+1](\frac{N}{2})!\,[\frac{N}{2}-m]!}$$

Using $\sum_{n=1}^{m} n = \frac{m(m+1)}{2}$ we thus obtain

$$\rho(m) = e^{\frac{1}{2}\beta J m^2 + \beta\mu H_0 m} \frac{\left[\left(\frac{N}{2}\right)!\right]^2}{\left[\frac{N}{2}+m\right]!\left[\frac{N}{2}-m\right]!}\rho_0 \qquad (157)$$

If we imagine that m < 0, we may use (152b) to get

$$\rho(m) = e^{-\beta\left[J(m+1)+\mu H_0 - \frac{J}{2}\right]}\left[\frac{\frac{N}{2}+m+1}{\frac{N}{2}-m}\right]\rho(m+1) \qquad (158)$$

where we have used

$$\frac{\Gamma_1(m+1)}{\Gamma_2(m)} = e^{-\beta\left[E(m)-E(m+1)\right]} = e^{-\beta\left[J(m+1)+\mu H_0 - \frac{J}{2}\right]} \qquad (159)$$

Thus:

$$\rho(m) = e^{-\beta\left[J(m+1)+\mu H_0 - \frac{J}{2}\right]} e^{-\beta\left[J(m+2)+\mu H_0 - \frac{J}{2}\right]} \cdots e^{-\beta\left[J\cdot 0+\mu H_0 - \frac{J}{2}\right]}$$

$$\frac{\left[\frac{N}{2}+m+1\right]\left[\frac{N}{2}+m+2\right]\cdots\left[\frac{N}{2}+0\right]}{\left[\frac{N}{2}-m\right]\left[\frac{N}{2}-m-1\right]\cdots\left[\frac{N}{2}+1\right]}\rho_0$$

Since m $<$ 0 we have

$$\rho(m) = e^{-\beta J\left[0-1-2\cdots-(|m|+1)\right]}\, e^{-\beta\left[\mu H_0 - \frac{J}{2}\right]|m|}$$

$$\frac{\left(\frac{N}{2}\right)!\left(\frac{N}{2}\right)\cdots\left[\frac{N}{2}+1-|m|\right]\left[\frac{N}{2}-|m|\right]!}{\left[\frac{N}{2}-m\right]\cdots\left[\frac{N}{2}+1\right]\left(\frac{N}{2}\right)!\left[\frac{N}{2}-|m|\right]!}\,\rho_0$$

Using

$$\sum_{n=0}^{|m|-1}{}' n = \frac{(|m|-1)|m|}{2} = \frac{(-m-1)(-m)}{2} = \frac{m(m+1)}{2}$$

we have

$$\rho(m) = e^{\frac{1}{2}\beta J m^2 + \beta \mu H_0 m}\,\frac{\left[\left(\frac{N}{2}\right)!\right]^2}{\left[\frac{N}{2}-m\right]!\left[\frac{N}{2}+m\right]!}\,\rho_0 \qquad (160)$$

Thus, from (157) and (160) we have the complete solution. The constant ρ_0 (which is independent of m but will depend on the external field H_0 and the temperature β) can be obtained from the normalization requirement:

$$\sum_{m=-\frac{N}{2}}^{+\frac{N}{2}}{}' \rho(m) = 1 \qquad (161)$$

Using either solution (157) or (160), if we compute

$$F(m) - F_0 = -\frac{1}{\beta} \ln \rho(m) \tag{162}$$

we find that F(m) is exactly the free energy of an Ising model ferromagnet as computed from the Bragg-Williams method [19] (we include all terms independent of m in F_0).

We note that the conventional Ising Hamiltonian[19] is written as a function of variables that assume only the values ± 1:

$$H_I = -\frac{1}{2} J \sum_{\substack{i,j=1 \\ i \neq j}}^{N} \sigma_i \sigma_j - \mu H_0 \sum_{i=1}^{N} \sigma_i \tag{163}$$

Our Hamiltonian (2) is written in terms of spin one half angular momentum operators S_i^z which have eigenvalues ± 1/2. Thus, our energy differs by a constant factor from the conventional model (163): that is, for example in the state $|0\rangle$ where all of the spins are perfectly aligned ($\sigma_i = +1$, all i; $S_j^z = +\frac{1}{2}$, all j) our energy (in the molecular field approximation) is

$$\langle 0|H|0\rangle = -\frac{1}{2} J \left(\frac{N}{2}\right)\left(\frac{N}{2}\right) - \mu H_0 \left(\frac{N}{2}\right) \tag{164}$$

while the energy of (102) is

$$\langle 0|H_I|0\rangle = -\frac{1}{2} J (N)(N) - \mu H_0 (N) \tag{165}$$

Thus, to compare our results with the Bragg-Williams theory of the conventional Ising model (163), we must change our energies E(m) from (156) to

$$E'(m) = 4\left[-\frac{1}{2} J m^2\right] + 2\left[-\mu H_0 m\right] \tag{166}$$

Noting that E(m) is precisely the factor which enters into the solutions (157) and (160) for ρ (m), we substitute E'(m) in these expressions and then compute the free energy F(m) given by (162) to be

$$F(m) = -2Jm^2 - 2\mu H_0 m + N k_B T \left\{ \frac{1}{2}\left(1 + \frac{2m}{N}\right) \ln\left[\frac{1}{2}\left(1 + \frac{2m}{N}\right)\right] + \frac{1}{2}\left(1 - \frac{2m}{N}\right) \ln\left[\frac{1}{2}\left(1 - \frac{2m}{N}\right)\right]\right\} \tag{167}$$

Noting that our variable m still corresponds to a spin one half system, we may change variables to model a two level system with arbitrary value μ of magnetic moment by defining

$$M = 2\mu m = \mu(n_1 - n_2) \tag{168}$$

Defining a dimensionless variable x as the ratio of M to its saturation value $N\mu$, we obtain from (167)

$$F(x) = -\frac{1}{2} J N^2 x^2 - \mu H_0 N x + N k_B T \left\{ \frac{1}{2}(1+x) \ln\left[\frac{1}{2}(1+x)\right] + \frac{1}{2}(1-x) \ln\left[\frac{1}{2}(1-x)\right]\right\} \tag{169}$$

$$X = \frac{M}{N\mu} = \frac{2m}{N} \tag{170}$$

Minimizing the free energy F(x) with respect to x by setting

$$\frac{\partial F(x)}{\partial x} = 0 \tag{171}$$

and solving for x we obtain the conventional molecular field equation of state:

$$X = \frac{M}{N\mu} = \tanh\left[\frac{\mu H_0}{K_B T} + \frac{NJ}{K_B T}\left(\frac{M}{N\mu}\right)\right] \tag{172}$$

Had we been slightly more careful in evaluating the energy (166) by taking each spin to interact with only z of its neighbors instead of all N of them, we would have observed the factor (NJ) in (172) to be changed to (zJ).

Referring to (169), if we set $H_0 = 0$ and assume that x << 1, we may expand the logarithymic factors in order to approximate the expression. Thus, keeping terms up to the fourth degree, we get

$$F(x) \simeq \frac{1}{2}\left(NK_B T - JN^2\right)x^2 + \frac{NK_B T}{12}x^4$$

$$= \frac{NK_B}{(N\mu)^2}\left(T - \frac{NJ}{K_B}\right)\frac{M^2}{2} + \frac{NK_B}{3}\frac{1}{N^4\mu^4}\frac{M^4}{4}$$

using the definition (170) for x. If we make the identification

$$T_c = \frac{NJ}{K_B}$$

which in view of the definition (110) of T_c relates the Weiss internal field constant λ to the exchange energy J via

$$\frac{\lambda \mu^2}{v} = J$$

we see that the Bragg-Williams free energy becomes, near T_c,

$$F(M) = \left(\frac{K_B}{N\mu^2}\right)(T-T_c)\,\frac{M^2}{2} + \left(\frac{K_B}{3N^3\mu^4}\right)T\,\frac{M^4}{4}$$

$$= C(T-T_c)\,\frac{M^2}{2} + d\,T\,\frac{M^4}{4}$$

which is exactly like the expression found by expanding the solutions of the Fokker-Planck equations near T_c, equation (125) (the constant d differs negligibly from the constant d' of (126) for $\tau \sim 1$).

Although we have not obtained the complete time-dependent solution of the equation of motion of the magnetization distribution function, equation (151), we may consider the equation for the mean magnetization:

$$\langle \dot{m}(t) \rangle = \sum_{m=-\frac{N}{2}}^{+\frac{N}{2}}{}' m\,\dot{\rho}(m,t) = \sum_{i=1}^{4} T_i \tag{173}$$

Using the equation of motion (151) we may easily evaluate the various terms implied by (173). For example

$$\begin{aligned} T_1 &= \sum_{m=-\frac{N}{2}}^{+\frac{N}{2}} m \left[\frac{N}{2}-(m-1)\right] \Gamma_2(m-1)\, \rho(m-1,t) \\ &= \sum_{u=-\frac{N}{2}-1}^{\frac{N}{2}-1} (u+1)\left[\frac{N}{2}-u\right] \Gamma_2(u)\, \rho(u,t) \\ &= \sum_{u=-\frac{N}{2}}^{\frac{N}{2}} (u+1)\left[\frac{N}{2}-u\right] \Gamma_2(u)\, \rho(u,t) \\ &= \left\langle (m+1)\left[\frac{N}{2}-m\right]\Gamma_2(m)\right\rangle \end{aligned} \qquad (174)$$

Similarly evaluating the other three terms we get

$$T_2 = -\left\langle m\left(\frac{N}{2}-m\right)\Gamma_2(m)\right\rangle$$

$$T_3 = -\left\langle m\left(\frac{N}{2}+m\right)\Gamma_1(m)\right\rangle$$

$$T_4 = \left\langle (m-1)\left[\frac{N}{2}+m\right]\Gamma_1(m)\right\rangle$$

Hence, we obtain from (173)

$$\langle \dot{m}(t)\rangle = \frac{N}{2}\left\langle \Gamma_2(m)-\Gamma_1(m)\right\rangle - \left\langle m\left[\Gamma_2(m)+\Gamma_1(m)\right]\right\rangle \qquad (175)$$

Thus we see that the equation of motion (94) of the average magnetization deduced from the Fokker-Planck equation for the distribution function agrees exactly with the equation for the mean motion (175) which followed from the complete equation of motion for the distribution function.

We have therefore shown that the steady state solution of the full equation of motion for the magnetization distribution function, equation (89), yields the same free energy (for all temperatures) as the Bragg-Williams approximation. We note that nowhere in our equation of motion method did we have to use any of the statistical counting procedures usually employed in the derivation of the Bragg-Williams free energy.

CONCLUSION

The results of our studies of the statistical properties of light scattered inelastically by material systems in Part A of this work have indicated that such systems as are well represented by our models would exhibit scattered light having Gaussian statistical properties. We have shown that under the general conditions of a diagonal, factorizable density matrix, the first and second order temporal correlation functions of the electric field of a radiation field are related such that the latter is essentially determined by the former. In terms of frequencies, the intensity spectrum is the convolution of the amplitude spectrum with itself and therefore does not contain more information than is obtained by measuring the power spectrum of the light. We found that the radiation density matrix for Raman scattered light did satisfy the two general conditions, and that the Gaussian factorization relation between the amplitude and intensity spectrum of light scattered by density fluctuations also held. In these studies we focussed our attention on a single component of the inelastically scattered light and neglected components at far different frequencies which may be present in the light scattered by a real system. We also made extensive use of density operator techniques to study the temporal evolution of the modes of interest to us, treating the effects of all unobserved degrees of freedom as reservoirs with which those modes interacted.

The possible breakdown of thermodynamic fluctuation theory, which generally predicts Gaussian statistics for the fluctuations of a material system, near a phase transition critical point led us to consider a Heisenberg-Ising model of a ferromagnet in Part B. We obtained equations of motion for the density operator of the system by assuming that it was coupled to a larger system which served as a thermal reservoir. Using techniques developed originally to treat laser problems, we transcribed the operator equations into a c-number quasiprobability density function formalism which could be treated by algebraic methods. Approximating the magnetic interactions of the system by a molecular field, we found that near the Curie temperature the dynamical evolution of the magnetization could be described by an equation of the Fokker-Planck type with nonlinear drift and diffusion coefficients. The specific form of the equation depended upon details of the thermal reservoir system to which the ferromagnet was coupled, but the steady state solutions for the two types of reservoirs considered in detail were identical and yielded the Landau form for the magnetic free energy. It was then demonstrated that the exact steady-state solution of the equation of motion of the magnetization distribution function yielded the Bragg-Williams free energy at all temperatures and the molecular field equation of state [48]. Thus, we have obtained the free energy of a ferromagnetic system starting with microscopic Hamiltonian dynamics and using a density operator technique which at no point involved the statistical counting procedures usually encountered in derivations of the Bragg-Williams approximation and which also yielded equations for the temporal evolution of the system through nonequilibrium states toward thermal equilibrium.

Appendix I. Gaussian Factorization Relation

The first order temporal correlation function $G^{(1)}(t,t')$ and the second order temporal correlation function $G^{(2)}(t,t';t',t)$ are defined by (1.8) and (1.9) as

$$G^{(1)}(t,t') = Tr\left\{\rho E^{(-)}(t)E^{(+)}(t')\right\} \tag{I.1}$$

$$G^{(2)}(t,t';t',t) = Tr\left\{\rho E^{(-)}(t)E^{(-)}(t')E^{(+)}(t')E^{(+)}(t)\right\} \tag{I.2}$$

For a stationary radiation field the density operator is time independent and the field operators may be expanded in free-field normal modes (1.5):

$$E^{(+)}(\vec{r},t) = i\sum_K{}'\left(\tfrac{1}{2}\hbar\omega_K\right)^{\frac{1}{2}} a_K \; U_K(\vec{r})\, e^{-i\omega_K t} \tag{I.3a}$$

$$E^{(-)}(\vec{r},t) = \left[E^{(+)}(\vec{r},t)\right]^{\dagger} \tag{I.3b}$$

The mode functions $U_K(\vec{r})$ are assumed to form a complete orthonormal set of functions and the operators a_K, a_K^{+} obey the Bose commutation relations (1.6). Using the expansions of the field operators in the defining equation for $G^{(2)}(t,t';t',t)$ gives:

$$G^{(2)}(t,t';t',t) = Tr\left\{\sum_l a_l^{+} U_l^{*} e^{i\omega_l t}\sum_p{}' a_p^{+} U_p^{*} e^{i\omega_p t'}\sum_m a_m U_m e^{-i\omega_m t'} \sum_q{}' a_q U_q e^{-i\omega_q t}\rho\right\} \tag{I.4}$$

$$G^{(2)}(t,t';t',t) = \sum_{n_1=0}^{\infty} \sum_{n_2=0}^{\infty} \cdots \left\{ \sum_{\substack{l,p\\q,m}} \mathcal{U}_l^* \mathcal{U}_p^* \mathcal{U}_m \mathcal{U}_q e^{i(\omega_l t + \omega_p t' - \omega_m t' - \omega_q t)} \langle \{n_1 n_2 \cdots\} | a_l^+ a_p^+ a_m a_q \rho | \{n_3\} \rangle \right\}$$

Here we have labeled the occupation numbers of the field modes by n_1, n_2, etc., and to save writing we take

$$\mathcal{U}_l^* \equiv \left[i \sqrt{\tfrac{1}{2} \hbar \omega_l}\, u_l(\vec{r}) \right]^* \tag{I.5}$$

Assuming that the density operator is diagonal as in (1.11)

$$\langle \{n\} | \rho | \{n'\} \rangle = \rho_{\{n\}\{n\}} \delta_{\{n\}\{n'\}} \tag{I.6}$$

we have three cases to consider in (I.4):

(i) $l = m \neq p = q$

(ii) $l = q \neq p = m$

(iii) $l = m = p = q$

Hence,

$$\begin{aligned} G^{(2)}(t,t';t',t) = \sum_{\{n\}} \Bigg\{ & \sum_{\substack{p\\p\neq l}} \sum_{l} |\mathcal{U}_l|^2 e^{i\omega_l (t-t')} |\mathcal{U}_p|^2 e^{i\omega_p (t'-t)} \langle \{n\} | a_l^+ a_p^+ a_l a_p | \{n\} \rangle \\ & + \sum_{\substack{p\\p\neq l}} \sum_{l} |\mathcal{U}_l|^2 |\mathcal{U}_p|^2 \langle \{n\} | a_l^+ a_p^+ a_p a_l | \{n\} \rangle \\ & + \sum_{l} |\mathcal{U}_l|^4 \langle \{n\} | a_l^+ a_l^+ a_l a_l | \{n\} \rangle \Bigg\} \rho_{\{n\}\{n\}} \end{aligned} \tag{I.7}$$

We next assume, as in (1.12), that

$$\rho_{\{n\}\{n\}} = \rho_{\substack{n_1 n_2 \cdots \\ n_1 n_2 \cdots}} = \rho^{(1)}_{n_1 n_1} \otimes \rho^{(2)}_{n_2 n_2} \otimes \cdots \otimes \rho^{(K)}_{n_K n_K} \otimes \cdots \tag{I.8}$$

Since the normalization

$$\mathrm{Tr}\, \rho = 1 \tag{I.9}$$

implies that

$$\mathrm{Tr}_{(K)}\, \rho^{(K)} = 1 = \sum_{n_K = 0}^{\infty} \rho^{(K)}_{n_K n_K} \tag{I.10}$$

for arbitrary K, the second order correlation function may be written as

$$\begin{aligned} G^{(2)}(t,t';t',t) = {} & \sum_{n_\ell, n_p} \sum_{\substack{\ell, p \\ \ell \neq p}} |U_\ell|^2 |U_p|^2 e^{i[\omega_\ell (t-t') + \omega_p (t'-t)]} \\ & \quad \langle n_\ell n_p | a_\ell^+ a_p^+ a_\ell a_p | n_\ell n_p \rangle \rho^{(\ell)}_{n_\ell n_\ell} \rho^{(p)}_{n_p n_p} \\ & + \sum_{n_\ell, n_p} \sum_{\substack{\ell, p \\ \ell \neq p}} |U_\ell|^2 |U_p|^2 \langle n_\ell n_p | a_\ell^+ a_p^+ a_p a_\ell | n_\ell n_p \rangle \rho^{(\ell)}_{n_\ell n_\ell} \rho^{(p)}_{n_p n_p} \\ & + \sum_{n_\ell} \sum_{\ell} |U_\ell|^4 \langle n_\ell | a_\ell^+ a_\ell^+ a_\ell a_\ell | n_\ell \rangle \rho^{(\ell)}_{n_\ell n_\ell} \end{aligned} \tag{I.11}$$

In the first two terms, since the sums are restricted to terms for which $\ell \neq \rho$ we have that

$$\sum_{n_\ell n_\rho} \langle n_\ell n_\rho | a_\ell^+ a_\rho^+ a_\ell a_\rho | n_\ell n_\rho \rangle \rho^{(\ell)}_{n_\ell n_\ell} \rho^{(\rho)}_{n_\rho n_\rho} = \Big[\sum_{n_\ell} n_\ell \rho^{(\ell)}_{n_\ell n_\ell} \Big] \Big[\sum_{n_\rho} n_\rho \rho^{(\rho)}_{n_\rho n_\rho} \Big] = \bar{n}_\rho \bar{n}_\ell \qquad \text{(I.12)}$$

while in the third term

$$\sum_{n_\ell} \langle n_\ell | a_\ell^+ a_\ell^+ a_\ell a_\ell | n_\ell \rangle \rho^{(\ell)}_{n_\ell n_\ell} = \overline{n_\ell^2} - \bar{n}_\ell \qquad \text{(I.13)}$$

Hence:

$$\begin{aligned} G^{(2)}(t,t';t',t) &= \sum_{\substack{\rho \\ \rho \neq \ell}} \sum_{\ell} |V_\ell|^2 e^{i\omega_\ell (t-t')} \bar{n}_\ell |V_\rho|^2 e^{i\omega_\rho (t'-t)} \bar{n}_\rho \\ &\quad + \sum_{\substack{\ell,\rho \\ \ell \neq \rho}}' |V_\ell|^2 |V_\rho|^2 \bar{n}_\ell \bar{n}_\rho + \sum_\ell |V_\ell|^4 \big(\overline{n_\ell^2} - \bar{n}_\ell \big) \end{aligned} \qquad \text{(I.14)}$$

We rewrite this as

$$\begin{aligned} G^{(2)}(t,t';t',t) &= \sum_\rho \Big\{ \sum_{\substack{\ell \\ \ell \neq \rho}} |V_\ell|^2 e^{i\omega_\ell (t-t')} \bar{n}_\ell + |V_\rho|^2 e^{i\omega_\rho (t-t')} \bar{n}_\rho \\ &\qquad - |V_\rho|^2 e^{i\omega_\rho (t-t')} \bar{n}_\rho \Big\} |V_\rho|^2 e^{i\omega_\rho (t'-t)} \bar{n}_\rho \\ &\quad + \sum_\rho \Big\{ \sum_{\substack{\ell \\ \ell \neq \rho}}' |V_\ell|^2 \bar{n}_\ell + |V_\rho|^2 \bar{n}_\rho - |V_\rho|^2 \bar{n}_\rho \Big\} |V_\rho|^2 \bar{n}_\rho \\ &\quad + \sum_\ell |V_\ell|^4 \big(\overline{n_\ell^2} - \bar{n}_\ell \big) \end{aligned} \qquad \text{(I.15)}$$

Using the expansions of the field operators (I.3) and the same assumptions of the form of the radiation density matrix (I.6) and (I.8), one can easily find that the first order correlation function can be expressed as

$$G^{(1)}(t,t') = \sum_{\ell} |V_\ell|^2 e^{i\omega_\ell (t-t')} \bar{n}_\ell \tag{I.16}$$

so that the above expression for G (t,t'; t',t) can be written as

$$G^{(2)}(t,t';t',t) = \sum_{\rho} \left\{ G^{(1)}(t,t') - |V_\rho|^2 e^{i\omega_\rho (t-t')} \bar{n}_\rho \right\} |V_\rho|^2 e^{i\omega_\rho (t-t')} \bar{n}_\rho$$

$$+ \sum_{\rho}' \left\{ G^{(1)}(t,t) - |V_\rho|^2 \bar{n}_\rho \right\} |V_\rho|^2 \bar{n}_\rho + \sum_{\ell}' |V_\ell|^4 (\overline{n_\ell^2} - \bar{n}_\ell) \tag{I.17}$$

$$G^{(2)}(t,t';t',t) = \sum_{\rho} \left\{ |V_\rho|^2 e^{i\omega_\rho (t'-t)} \bar{n}_\rho \right\} G^{(1)}(t,t') - \sum_{\rho}' |V_\rho|^4 \bar{n}_\rho^{\,2}$$

$$+ \sum_{\rho}' |V_\rho|^2 \bar{n}_\rho G^{(1)}(t,t) - \sum_{\rho}' |V_\rho|^4 \bar{n}_\rho^2 + \sum_{\ell} |V_\ell|^4 (\overline{n_\ell^2} - \bar{n}_\ell) \tag{I.18}$$

$$G^{(2)}(t,t';t',t) = G^{(1)}(t,t')\, G^{(1)}(t',t) + G^{(1)}(t,t)\, G^{(1)}(t',t') \tag{I.19}$$

$$+ \sum_{\ell} |V_\ell|^4 (\overline{n_\ell^2} - \bar{n}_\ell - 2\bar{n}_\ell^{\,2})$$

Now, setting t' = t in (I.19) gives

$$G^{(2)}(t,t;t,t) = 2\left|G^{(1)}(t,t)\right|^2 + \sum_{\ell} |U_\ell|^4 \left(\overline{n_\ell^2} - \bar{n}_\ell - 2\bar{n}_\ell^2\right) \qquad \text{(I.20)}$$

so that finally the relation between the first and second order temporal correlation functions for a stationary radiation field with a diagonal, factorizable density operator becomes

$$G^{(2)}(t,t';t',t) = G^{(1)}(t,t')\,G^{(1)}(t',t) + G^{(2)}(t,t;t,t) - G^{(1)}(t,t)\,G^{(1)}(t',t') \qquad \text{(I.21)}$$

which is the same as (1.13). Using the definitions (1.15) and (1.16) for the intensity and amplitude spectrum respectively, one obtains (1.14) by Fourier transforming (1.13).

For Gaussian light, that is, light whose individual mode density matrices are of the form

$$\rho^{(j)}_{n_j n_j} = (\bar{n}_j)^{n_j}\left[1 + \bar{n}_j\right]^{-(n_j+1)} \qquad \text{(I.22)}$$

it is easily shown that

$$\overline{n_j^2} = 2\bar{n}_j^2 + \bar{n}_j \qquad \text{(I.23)}$$

so that in this case one has

$$G^{(2)}(t,t;t,t) = 2\left|G^{(1)}(t,t)\right|^2 \qquad \text{(I.24)}$$

and the relation between the intensity and amplitude spectra is

$$G^{(2)}(\omega) = 2\pi \left| G^{(1)}(t,t) \right|^{2} \delta(\omega) + \frac{1}{2\pi} G^{(1)}(\omega) \otimes G^{(1)*}(\omega) \tag{I.25}$$

which is the same relation found to hold if the electric field is considered to be a classical Gaussian random process. We see that it is not expected to hold in general. In particular, if the radiation field consists of a single mode Glauber coherent state [10]

$$\rho^{(j)}_{n_j n_j} = (\bar{n}_j)^{n_j} e^{-\bar{n}_j} \left[n_j ! \right]^{-1} \tag{I.26}$$

one can show that

$$\overline{n_j^2} = \bar{n}_j^{\,2} + \bar{n}_j \tag{I.27}$$

so that

$$\begin{aligned} G^{(2)}(t,t;t,t) &= 2\left[|U_j|^{2} \bar{n}_j \right]^{2} - |U_j|^{4} \left(\bar{n}_j^{2} + \bar{n}_j - \bar{n}_j - 2\bar{n}_j^{2} \right) \\ &= \left| G^{(1)}(t,t) \right|^{2} \end{aligned} \tag{I.28}$$

so that the frequency relation becomes simply

$$G^{(2)}(\omega) = \frac{1}{2\pi} G^{(1)}(\omega) \otimes G^{(1)*}(\omega)$$

Appendix II. Regression Theorem for Markoff Processes

Following Lax [7], we consider the relation of a two time correlation function to a single time average for a Markoffian stochastic process. A process is specified by its multitime probability density functions

$$P(a_1, a_2, \cdots a_n) \equiv P[a(t_1), t_1; a(t_2), t_2; \cdots a(t_n), t_n]$$

which give the joint probability of finding the variable a to have value $a(t_1) = a_1$ at t_1, $a(t_2) = a_2$ at t_2 etc. for the case of a single variable process. Defining the conditional probability in general by

$$P(A|B) = P(A, B)/P(B)$$

a Markoffian process is defined by $(t_n > t_{n-1} > \cdots > t_1)$

$$P(a_n | a_{n-1} \cdots a_1) = P(a_n | a_{n-1})$$

The identity

$$P(a_3, a_2, a_1)/P(a_1) = [P(a_3, a_2, a_1)/P(a_2, a_1)][P(a_2, a_1)/P(a_1)]$$

Thus becomes for Markoffian processes

$$P(a_3 a_2 | a_1) = P(a_3 | a_2) P(a_2 | a_1)$$

$$P(a, t; a', t' | a_0, t_0) = P(a, t | a', t') P(a', t' | a_0, t_0)$$

multiplying by $M(a(t)) = M(a)$ and integrating over a and a'

$$\left\langle M(a(t)) \right\rangle_{a_0} = \int \left\langle M(a(t)) \right\rangle_{a(t')=a'} da' \, P(a',t'|a_0,t_0)$$

Similarly multiplying by $M(a(t)) N(a(t')) = M(a) N(a')$ we get

$$\left\langle M(a(t)) N(a(t')) \right\rangle_{a_0} = \int \left\langle M(a(t)) \right\rangle_{a(t')=a'} da' \, N(a') P(a',t'|a_0,t_0)$$

Multiplying by the initial distribution and averaging we get

$$\left\langle M(a(t)) N(a(t')) \right\rangle = \int \left\langle M(a(t)) \right\rangle_{a'} N(a') \, P(a',t') \, da'$$

Specializing to N=1 we thus have

$$\langle M(a(t)) \rangle = \int \langle M(a(t)) \rangle_{a'} \, da' \, P(a',t')$$

Thus, the one and two time averages have the same time dependence. This is the idea of the regression theorem. In the case that the mean of a variable can be linearly expressed in terms of some set of variables at an earlier time

$$\left\langle M(a(t)) \right\rangle_{a'} = \sum_{\mu} O_{\mu}(t,t') M_{\mu}(a')$$

then it follows that

$$\langle M(a(t)) N(a(t')) \rangle = \sum_{\mu} O_{\mu}(t,t') \langle M_{\mu}(a') N(a') \rangle$$

$$\langle M_{\mu}(a') N(a') \rangle = \int M_{\mu}(a') N(a') \, P(a',t') \, da'$$

This theorem has been proved by Lax for quantum systems [8,32] by techniques not used elsewhere in this work. It has also been demonstrated using ordered c-number distribution function methods [31,33].

Appendix III. Damping Theory

Here we follow Louisell [17] in obtaining a density matrix equation for a reservoir-system interaction slightly more general than that taken in Part B, Section I. We treat the case of a single atom with states $|K\rangle$ (energy $\mathcal{E}_K$) coupled to a reservoir by a general interaction V which may be written as

$$V = \sum_{K,\ell} |K\rangle\langle K|V|\ell\rangle\langle\ell| \equiv \hbar \sum_{K,\ell} f_{K\ell}\ |K\rangle\langle\ell| \tag{III.1}$$

$$\hbar f_{K\ell} = \langle K|V|\ell\rangle \tag{III.2}$$

Assuming a thermal oscillator reservoir as in Part A Section III and arguing in ways similar to that Section we obtain the Markoffian equation of motion for the reduced system density operator $\mathcal{S}_I(t)$ in the interaction picture:

$$\frac{\partial \mathcal{S}_I(t)}{\partial t} = -\frac{1}{\hbar^2}\, Tr_R \int_{-\infty}^{t} dt' \left[V(t), \left[V(t'), S_I(t)\, f_0(H_R)\right]\right] \tag{III.3}$$

$$V(t) = e^{\frac{i}{\hbar}(H_s+H_R)t}\, V\, e^{-\frac{i}{\hbar}(H_s+H_R)t} = \hbar {\sum_{K,\ell}}' f_{K\ell}(t)|K\rangle\langle\ell|\, e^{i\omega_{K\ell}t} \tag{III.4}$$

$$f_{K\ell}(t) = e^{\frac{i}{\hbar}H_R t}\, f_{K\ell}\, e^{-\frac{i}{\hbar}H_R t} \tag{III.5}$$

$$\hbar\omega_{K\ell} = \epsilon_K - \epsilon_\ell \tag{III.6}$$

We have assumed that V contains no matrix elements diagonal in reservoir states since such terms could be absorbed into the reservoir Hamiltonian. Inserting for V(t) in (III.3) we get

$$\dot{\rho}_I(t) = -\sum_{k\ell mn}\left[\delta_{\ell m}|k\rangle\langle n|\rho_I(t) - |m\rangle\langle n|\rho_I(t)|k\rangle\langle \ell|\right]\theta^{(t)}_{k\ell mn} \tag{III.7}$$

$$-\sum_{k\ell mn}\left[\rho_I(t)|m\rangle\langle \ell|\delta_{nk} - |k\rangle\langle \ell|\rho_I(t)|m\rangle\langle n|\right]\theta^{(t)}_{mnk\ell}$$

$$\theta^{(t)}_{k\ell mn} = \int_{-\infty}^{t} dt'\,\langle f_{k\ell}(t)\,f_{mn}(t')\rangle\, e^{i[\omega_{k\ell}t+\omega_{mn}t']} \tag{III.8}$$

$$\theta^{(t)}_{mnk\ell} = \int_{-\infty}^{t} dt'\,\langle f_{mn}(t')\,f_{k\ell}(t)\rangle\, e^{i[\omega_{k\ell}t+\omega_{mn}t']} \tag{III.9}$$

Clearly the reservoir correlation functions depend only upon the time difference t-t' so that we may write

$$\theta^{(t)}_{k\ell mn} = \int_0^{\infty} du\,\langle f_{k\ell}(u)\,f_{mn}(0)\rangle\, e^{i[\omega_{k\ell}t+\omega_{mn}(t-u)]}$$

$$\equiv W^{(+)}_{k\ell mn}\, e^{i[\omega_{k\ell}+\omega_{mn}]t} \tag{III.10}$$

$$\theta^{(t)}_{mnk\ell} = \int_0^{\infty} du\,\langle f_{mn}(0)\,f_{k\ell}(u)\rangle\, e^{-i\omega_{mn}u}\, e^{i[\omega_{k\ell}+\omega_{mn}]t}$$

$$\equiv W^{(-)}_{mnk\ell}\, e^{i[\omega_{k\ell}+\omega_{mn}]t} \tag{III.11}$$

Substituting these results into (III.7) and assuming that $\omega_{mn} t >> 1$ for any pair of levels m,n so that we may keep only the energy conserving terms where $\omega_{mn}+\omega_{kl} = 0$, we obtain

$$\dot{S}'(t) = {\sum_l}' \frac{\epsilon_l}{i\hbar}\left[|l\rangle\langle l|, S'(t)\right] + {\sum_{k,m}}' W_{km} |m\rangle\langle m| S_{kk}(t) + \sum_{k,m}\Big[W^{(+)}_{kkmm} \tag{III.12}$$

$$+ W^{(-)}_{kkmm} \Big] |m\rangle\langle k| S_{mk}(t) - \sum_{k,l}\left[W^{(+)}_{kmmk} |k\rangle\langle k| S(t) + S(t) |k\rangle\langle k| W^{(-)}_{kmmk} \right]$$

where ${\sum_{m,n}}'$ means leaving out the m=n terms of the double sum. We have transformed back to the Schrödinger picture and

$$W_{km} \equiv W^{(+)}_{kmmk} + W^{(-)}_{kmmk}$$

$$= W^{(+)}_{kmmk} + \left(W^{(+)}_{kmmk}\right)^* = 2\,\mathrm{Re}\, W^{(+)}_{kmmk} \tag{III.13}$$

$$= \left(W^{(-)}_{kmmk}\right)^* + W^{(-)}_{kmmk} = 2\,\mathrm{Re}\, W^{(-)}_{kmmk}$$

We define

$$\Gamma^{(c)}_{lk} = - W^{(+)}_{llkk} - W^{(-)}_{llkk} + \sum_m \left(W^{(+)}_{kmmk} + W^{(-)}_{lmml} \right) \tag{III.14}$$

Returning to the definitions of the quanties on the right side we write

$$\Gamma^{(c)}_{lk} = - \int_0^\infty dt \left[\langle f_{ll}(t) f_{kk}(0) \rangle + \langle f_{ll}(0) f_{kk}(t) \rangle \right]$$

$$
\begin{aligned}
&+ \sum_m \Big\{ \int_0^\infty dt\, e^{-i\omega_{mn} t} \langle f_{km}(t)\, f_{mk}(0) \rangle \\
&+ \int_0^\infty dt\, e^{-i\omega_{\ell m} t} \langle f_{\ell m}(0)\, f_{m\ell}(t) \rangle \Big\}
\end{aligned}
$$

Using the diagonal representation of reservoir states $H_R |R\rangle = \hbar R | R\rangle$ we get

$$
\begin{aligned}
\Gamma^{(c)}_{\ell k} = - \int_0^\infty dt \Big\{ &\sum_{R',R''} e^{-i(R'-R'')t} f_0(R') \langle R' | f_{\ell\ell} | R''\rangle \langle R'' | f_{kk} | R'\rangle \\
&+ e^{-i(R''-R')t} f_0(R') \langle R' | f_{\ell\ell} | R''\rangle \langle R'' | f_{kk} | R'\rangle \Big\} \\
+ \int_0^\infty dt \sum_m \sum_{R',R''} \Big\{ &e^{-i\omega_{mk} t}\, e^{i(R'-R'')t} f_0(R') \langle R' | f_{km} | R''\rangle \langle R'' | f_{mk} | R'\rangle \\
&+ e^{-i\omega_{\ell m} t}\, e^{-i(R''-R')t} f_0(R') \langle R' | f_{\ell m} | R''\rangle \langle R'' | f_{m\ell} | R'\rangle \Big\}
\end{aligned}
\tag{III.15}
$$

Defining $Re\, \Gamma^{(c)}_{\ell k} = \Gamma_{\ell k}$ and thus extending the time integrals in (III.15) to minus infinity, we obtain

$$
\begin{aligned}
\Gamma_{\ell k} = \pi \sum_R f_0(R) \Big\{ &| \langle R | (f_{\ell\ell} - f_{kk})^2 | R\rangle |^2 \\
&+ \sum_{R'} \Big[\sum_{\substack{m \\ m \neq \ell}} | \langle R | f_{\ell m} | R'\rangle |^2 \delta(R' - R + \omega_{\ell m}) \\
&+ \sum_{m \neq k} | \langle R | f_{km} | R'\rangle |^2 \delta(R' - R + \omega_{km}) \Big\}
\end{aligned}
\tag{III.16}
$$

$$
= \Gamma^{ph}_{\ell k} + \frac{1}{2} \Big(\sum_m W_{m\ell} + \sum_m W_{mk} \Big) = \Gamma^{ph}_{\ell k} + \frac{1}{2} \left(\Gamma_\ell + \Gamma_k \right)
$$

Thus we obtain the usual Fermi Golden Rule expressions for the level damping constants Γ_l and Γ_K but we also obtain a new damping constant which did not appear in Part B Section I because of the form of the interaction chosen there. However, we shall find below that this new constant in no way modifies our final equation (89) for the longitudinal magnetization distribution function although it does change somewhat the full equation of motion involving the transverse components of the magnetization.

Continuing, we note that

$$\begin{aligned} R_e\left(W^{(+)}_{llKK} + W^{(-)}_{llKK}\right) &= -\Gamma_{lK} + R_e \sum_m \left(W^{(+)}_{KmmK} + W^{(-)}_{lmml}\right) \\ &= -\Gamma_{lK} + \frac{1}{2} \sum_{\substack{m \\ m \neq K, \\ m \neq l}} \left(W_{mK} + W_{ml}\right) \\ &= -\left[\Gamma^{ph}_{lK} + \frac{1}{2}(\Gamma_l + \Gamma_K)\right] + \frac{1}{2}(\Gamma_K + \Gamma_l) + \frac{1}{2}\left(W_{KK} + W_{ll}\right) \qquad \text{(III.17)} \\ &= -\Gamma^{ph}_{lK} + \frac{1}{2}\left(W_{ll} + W_{KK}\right) \end{aligned}$$

Returning to the density operator equation of motion (III.12) and generalizing to the case of N atoms, each interacting with its own thermal reservoir but not directly with any other atom, we get (using the same symbol S(t) for the density operator of the whole collection of atoms, a product of the atomic density operators)

$$\dot{S}(t) = \sum_{j=1}^{N} \Big\{ \sum_{\ell} \Big[\Big(\frac{\epsilon_\ell}{i\hbar} - \frac{\Gamma_\ell}{2}\Big)(|\ell\rangle\langle\ell|)_j S(t) - \Big(\frac{\epsilon_\ell}{i\hbar} + \frac{\Gamma_\ell}{2}\Big) S(t)\,(|\ell\rangle\langle\ell|)_j \Big]$$

$$+ \sum_{k,\ell}{}' \Big[W_{\ell k}\, (|\ell\rangle\langle k|)_j\, S(t)\, (|k\rangle\langle\ell|)_j$$

$$- \Gamma^{ph}_{\ell k}\, (|k\rangle\langle k|)_j\, S(t)\, (|\ell\rangle\langle\ell|)_j \Big] \Big\} \qquad \text{(III.18)}$$

If we specialize (III.18) to the case of N two level atoms to represent our ferromagnetic spin system and carry out the transition to the c-number magnetization distribution function as in Part B, Section II we obtain the following equation:

$$\dot{\rho}(s, n_2, s^*, t) = \Big\{ \frac{\partial}{\partial s^*} (\Gamma_{12} + i\omega_{21})\, s^* + \frac{\partial}{\partial s} (\Gamma_{12} - i\omega_{21})\, s$$

$$+ (e^{-\partial_{n_2}} - 1)\Gamma_1 n_1 + (e^{\partial_{n_2}} - 1)\Gamma_2 n_2$$

$$+ \frac{\partial^2}{\partial s \partial s^*} \Big[2\Gamma^{ph}_{12} e^{\partial_{n_2}} n_2 + N\Gamma_1 \Big] \qquad \text{(III.19)}$$

$$+ \frac{\partial}{\partial s^*} (e^{-\partial_{n_2}} - 1)\Gamma_1 s^* + \frac{\partial}{\partial s} (e^{-\partial_{n_2}} - 1)\Gamma_1 s + \frac{\partial^3}{\partial s \partial s^{*2}} \Gamma_1 s^*$$

$$+ \frac{\partial^3}{\partial s^2 \partial s^*} \Gamma_1 s + \frac{\partial^4}{\partial s^2 \partial s^{*2}} e^{\partial_{n_2}} \Gamma_1 n_2 \Big\} \rho(s, n_2, s^*, t)$$

where

$$\Gamma_{12} = \Gamma^{ph}_{12} + \frac{1}{2}(\Gamma_1 + \Gamma_2) \qquad \text{(III.20)}$$

Thus, aside from the slight change in Γ_{12} given by (III.20) and in one other term of (III.19), we obtain the same equation of motion as before, in particular after integrating over the transverse variables.

Appendix IV. Detailed Balance Relation

We shall derive the detailed balance relation (27)

$$\Gamma_1(\omega_{21}) = e^{-\beta\hbar\omega_{21}}\,\Gamma_2(\omega_{21})$$

using the general expressions (25) and (26). Thus

$$\frac{1}{2}\Gamma_1(\omega_{21}) = g^2 \, Re \int_{-\infty}^{t} dt' \left\langle \sum_\alpha O_\alpha^+(t) \sum_\beta O_\beta(t') \right\rangle e^{i\omega_{21}(t-t')}$$

$$= g^2 \, Re \int_{-\infty}^{t} dt' \frac{1}{Z_R} Tr_R \left[e^{-\beta H_R} e^{\frac{i}{\hbar}H_R t} \theta^+ e^{-\frac{i}{\hbar}H_R t} e^{\frac{i}{\hbar}H_R t'} \theta e^{-\frac{i}{\hbar}H_R t'} \right] e^{-i\omega_{21}(t-t')}$$

where

$$\theta \equiv \sum_\alpha O_\alpha \, ; \; \theta^+ \equiv \sum_\alpha O_\alpha^+$$

$$Z_R = Tr_R \left[e^{-\beta H_R} \right]$$

Thus

$$\frac{1}{2}\Gamma_1(\omega_{21}) = g^2 \, Re \int_{-\infty}^{t} dt' Z_R^{-1} Tr_R \left[e^{-\beta H_R} e^{\frac{i}{\hbar}H_R(t-t')} \theta^+ e^{-\frac{i}{\hbar}H_R(t-t')} \theta \right] e^{-i\omega_{21}(t-t')}$$

$$= g^2 \, Re \int_{0}^{\infty} (+du) \, Z_R^{-1} \sum_{m,n} \left[e^{-\beta E_n} e^{\frac{i}{\hbar}E_n u} \langle n|\theta^+|m\rangle\langle m|\theta|n\rangle e^{-\frac{i}{\hbar}(E_m+\hbar\omega_{21})u} \right]$$

$$= g^2 \, Re \int_{\infty}^{0} (-du) \, Z_R^{-1} Tr_R \left[e^{-\beta H_R} e^{\frac{i}{\hbar}H_R u} \theta^+ e^{-\frac{i}{\hbar}H_R u} \theta \right] e^{-i\omega_{21} u}$$

$$\frac{1}{2}\Gamma_1(\omega_{21}) = \frac{g^2}{2} Z_R^{-1} \sum_{n,m} e^{-\beta E_n} \int_{-\infty}^{\infty} du\, e^{\frac{i}{\hbar}(E_n - E_m - \hbar\omega_{21})u} \langle n|\theta^+|m\rangle\langle m|\theta|n\rangle$$

$$= \pi g^2 Z_R^{-1} \sum_{n,m}' e^{-\beta E_n} \delta\left(\frac{E_n}{\hbar} - \frac{E_m}{\hbar} - \omega_{21}\right) \langle n|\theta^+|m\rangle\langle m|\theta|n\rangle$$

We change the summation indices by n → m, m → n to get

$$\frac{1}{2}\Gamma_1(\omega_{21}) = \pi g^2 Z_R^{-1} \sum_{n,m}' e^{-\beta E_m} \delta\left(\frac{E_m}{\hbar} - \frac{E_n}{\hbar} - \omega_{21}\right) \langle m|\theta^+|n\rangle\langle n|\theta|m\rangle$$

Since $\delta(-x) = \delta(x)$ and substituting for Em

$$\frac{1}{2}\Gamma_1(\omega_{21}) = \pi g^2 Z_R^{-1} \sum_{n,m}' e^{-\beta(E_n + \hbar\omega_{22})} \delta\left(\frac{E_n}{\hbar} - \frac{E_m}{\hbar} + \omega_{21}\right) \langle n|\theta|m\rangle\langle m|\theta^+|n\rangle$$

$$= \pi g^2 Z_R^{-1} e^{-\beta\hbar\omega_{21}} \sum_{n,m}' e^{-\beta E_n} \delta\left(\frac{E_n}{\hbar} - \frac{E_m}{\hbar} + \omega_{21}\right) \langle n|\theta|m\rangle\langle m|\theta^+|n\rangle$$

$$= \frac{g^2}{2} e^{-\beta\hbar\omega_{21}} Z_R^{-1} \sum_{n,m} e^{-\beta E_n} \int_{-\infty}^{\infty} du\, e^{\frac{i}{\hbar}(E_n - E_m + \hbar\omega_{21})} \langle n|\theta|m\rangle\langle m|\theta^+|n\rangle$$

$$= g^2 e^{-\beta\hbar\omega_{21}} \mathrm{Re} \sum_{n,m}' Z_R^{-1} e^{-\beta E_n} \int_0^{\infty} du\, e^{\frac{i}{\hbar}(E_n - E_m + \omega_{21})u} \langle n|\theta|m\rangle\langle m|\theta^+|n\rangle$$

$$= g^2 e^{-\beta\hbar\omega_{21}} \mathrm{Re} \sum_{n,m}' Z_R^{-1} e^{-\beta E_n} \int_{-\infty}^{t} dt'\, e^{\frac{i}{\hbar}(E_n - E_m + \hbar\omega_{21})(t-t')} \langle n|\theta|m\rangle\langle m|\theta^+|n\rangle$$

$$\frac{1}{2}\Gamma_1(\omega_{21}) = g^2 e^{-\beta\hbar\omega_{21}} \, \mathrm{Re}\int_{-\infty}^{t} dt' \sum_{n,m}{}' Z_R^{-1} e^{-\beta E_n}$$

$$\langle n| e^{\frac{i}{\hbar}E_n(t-t')} \, \theta \, e^{-\frac{i}{\hbar}E_m(t-t')} |m\rangle\langle m|\theta^{+}|n\rangle \, e^{-\hbar\omega_{21}(t-t')}$$

$$= g^2 e^{-\beta\hbar\omega_{21}} \, \mathrm{Re}\int_{-\infty}^{t} dt' \, Z_R^{-1} \, \mathrm{Tr}_R\Big[e^{-\beta H_R} e^{\frac{i}{\hbar}H_R t} \, \theta$$

$$e^{-\frac{i}{\hbar}H_R t} \, e^{\frac{i}{\hbar}H_R t'} \, \theta^{+} e^{-\frac{i}{\hbar}H_R t'}\Big] \, e^{-\hbar\omega_{21}(t-t')}$$

$$= g^2 e^{-\beta\hbar\omega_{21}} \, \mathrm{Re}\int_{-\infty}^{t} dt' \, \langle \theta(t)\, \theta^{+}(t') \rangle \, e^{-\hbar\omega_{21}(t-t')}$$

$$= e^{-\beta\hbar\omega_{21}}\left[\frac{1}{2}\Gamma_2(\omega_{21})\right]$$

Appendix V: T_6

For completeness we list the sixteen terms obtained from (82) (we abbreviate the notation to save writing by including only the identifying parts of each term) and the final results of properly ordering them and computing the trace over $\sigma(t)$.

$$t_1 = |1\rangle\langle 2|\, e^{-\partial n_2 |2\rangle\langle 2|}\, |2\rangle\langle 1| \rightarrow e^{-\partial n_2}\, n_1\, \rho(s, n_2, s^*, t)$$

$$t_2 = |1\rangle\langle 2|\, e^{-\partial n_2 |2\rangle\langle 2|}\, \partial_{s^*} |2\rangle\langle 2| \rightarrow e^{-\partial n_2}\, \partial_{s^*} s^*\, \Gamma_1\, \underline{\rho}(s, n_2, s^*, t)$$

$$t_3 = |1\rangle\langle 2|\, e^{-\partial n_2 |2\rangle\langle 2|}\, (-\partial_{s^*} |1\rangle\langle 1|) \rightarrow 0$$

$$t_4 = |1\rangle\langle 2|\, e^{-\partial n_2 |2\rangle\langle 2|}\, (-\partial^2_{s^*} |1\rangle\langle 2|) \rightarrow 0$$

$$t_5 = (-|1\rangle\langle 1|\partial_s)\, e^{-\partial n_2 |2\rangle\langle 2|}\, |2\rangle\langle 1| \rightarrow 0$$

$$t_6 = (-|1\rangle\langle 1|\partial_s)\, e^{-\partial n_2 |2\rangle\langle 2|}\, \partial_{s^*} |2\rangle\langle 2| \rightarrow 0$$

$$t_7 = (-|1\rangle\langle 1|\partial_s)\, e^{-\partial n_2 |2\rangle\langle 2|}\, (-|1\rangle\langle 1|\partial_{s^*}) \rightarrow \frac{\partial^2}{\partial s \partial s^*}\, n_1 \Gamma_1\, \rho(s, n_2, s^*, t)$$

$$t_8 = (-|1\rangle\langle 1|\partial_s)\, e^{-\partial n_2 |2\rangle\langle 2|}\, (-|1\rangle\langle 2|\partial^2_{s^*}) \rightarrow \frac{\partial^3}{\partial s\, \partial s^{*2}}\, s^* \Gamma_1\, \rho(s, n_2, s^*, t)$$

$$t_9 = |2\rangle\langle 2|\partial_\alpha e^{-\partial n_2 |2\rangle\langle 2|} |2\rangle\langle 1| \rightarrow \partial_\alpha \alpha \Gamma_1 \rho(\alpha, n_2, \alpha^*, t)$$

$$t_{10} = |2\rangle\langle 2|\partial_\alpha e^{-\partial n_2 |2\rangle\langle 2|} |2\rangle\langle 2|\partial_{\alpha^*} \rightarrow \frac{\partial^2}{\partial\alpha\partial\alpha^*} n_2 \Gamma_1 \rho(\alpha, n_2, \alpha^*, t)$$

$$t_{11} = |2\rangle\langle 2|\partial_\alpha e^{-\partial n_2 |2\rangle\langle 2|} (-|1\rangle\langle 1|\partial_{\alpha^*}) \rightarrow 0$$

$$t_{12} = |2\rangle\langle 2|\partial_\alpha e^{-\partial n_2 |2\rangle\langle 2|} (-|1\rangle\langle 2|\partial^2_{\alpha^*}) \rightarrow 0$$

$$t_{13} = (-\partial^2_\alpha |2\rangle\langle 1|) e^{-\partial n_2 |2\rangle\langle 2|} |2\rangle\langle 1| \rightarrow 0$$

$$t_{14} = (-\partial^2_\alpha |2\rangle\langle 1|) e^{-\partial n_2 |2\rangle\langle 2|} |2\rangle\langle 2|\partial_{\alpha^*} \rightarrow 0$$

$$t_{15} = (-\partial^2_\alpha |2\rangle\langle 1|) e^{-\partial n_2 |2\rangle\langle 2|} (-|1\rangle\langle 1|\partial_{\alpha^*}) \rightarrow \frac{\partial^3}{\partial\alpha\partial\alpha^{*2}} \alpha \Gamma_1 \rho(\alpha, n_2, \alpha^*, t)$$

$$t_{16} = (-\partial^2_\alpha |2\rangle\langle 1|) e^{-\partial n_2 |2\rangle\langle 2|} (-|1\rangle\langle 2|\partial^2_{\alpha^*}) \rightarrow \frac{\partial^4}{\partial\alpha^2\partial\alpha^{*2}} e^{\partial n_2} n_2 \Gamma_1 \rho(\alpha, n_2, \alpha^*, t)$$

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BIOGRAPHY

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