COFFEY, Charles Stevens, 1938-
A NEW METHOD OF TREATING EQUILIBRIUM AND STEADY
STATE SYSTEMS AND THE CORRELATED FLUCTUATIONS
WITHIN THEM.

The University of Arizona, Ph.D., 1971
Physics, general

University Microfilms, A XEROX Company, Ann Arbor, Michigan
A NEW METHOD OF TREATING EQUILIBRIUM AND STEADY STATE SYSTEMS
AND THE CORRELATED FLUCTUATIONS WITHIN THEM

by
Charles Stevens Coffey

A Dissertation Submitted to the Faculty of the
DEPARTMENT OF PHYSICS
In Partial Fulfillment of the Requirements
For the Degree of
DOCTOR OF PHILOSOPHY
In the Graduate College
THE UNIVERSITY OF ARIZONA

1971
I hereby recommend that this dissertation prepared under my direction by Charles Stevens Coffey entitled A NEW METHOD OF TREATING EQUILIBRIUM AND STEADY STATE SYSTEMS AND THE CORRELATED FLUCTUATIONS WITHIN THEM be accepted as fulfilling the dissertation requirement of the degree of Doctor of Philosophy.

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SIGNED: Charles J. Coffey
To Rick, Matthew, Andrea and Janet,
whose cheerfulness and humor
helped keep this endeavor in perspective
and made it all worthwhile.
ACKNOWLEDGMENT

My sincere thanks to Professor Roald K. Wangsness for his aid and advice in writing this dissertation. He always listened patiently and with an open mind, made valuable suggestions, and tried to keep the physics honest by insisting on comparing the theory with experimental reality. Without his encouragement and advice this dissertation could not have been written.
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ABSTRACT

An off diagonal statistical operator is defined, which, when combined with the Liouville equation, leads to a set of equations describing physical processes within large systems. These equations are further developed to describe correlated fluctuations in systems in equilibrium or in a steady state. A number of diverse examples are treated, and it is shown that one can reproduce and amplify known results; in addition, several new phenomena are predicted. Included in these examples are the Brown Twiss result on correlated photon emission and predictions on the phenomenon of photon bunching. Photon fluctuations in a laser are treated in some detail and results are obtained which agree better with experiment than previous theory. A description of the behavior of a laser as it moves from below threshold into a lasing condition is given and is shown to agree with the available experimental data. When applied to solids, the method leads to predictions of phonon number fluctuations and to the fluctuation dissipation theorem in both the high and low temperature limits. The correlated fluctuations of a simple magnet are treated in general, and it is shown that at the Curie temperature the fluctuations vanish. This conclusion is substantiated by the available experimental data, in contrast to the classical thermodynamic prediction of large or infinite fluctuations at this point. The Barkhausen effect of magnetic noise is considered in
terms of the simple model and results are derived which agree surprisingly well with experiment.

The general method developed herein can also be used to treat a number of other problems of fluctuation phenomena and should be applicable to a general description of the steady state and nonequilibrium conditions.
CHAPTER 1

INTRODUCTION

This dissertation investigates some aspects of statistical physics. We begin with a reformulation of the concept of the off diagonal statistical operator. Combining this with the Liouville equation, we derive a linear differential equation which describes the possible changes of the operator. From this equation, we obtain a general expression for the number of correlated fluctuations for systems that are either in equilibrium or in the steady state. A number of diverse applications of this general result are discussed, including the Brown Twiss result, fluctuations in a laser, the fluctuation dissipation theorem and several others. In this chapter, we briefly review the role of the density matrix in quantum statistical mechanics.\textsuperscript{1,2}

In many complex systems, it is often only possible to give the probability $q_m$ that the system is in the particular state represented by the Ket vector $|m\rangle$. The average value of any observable quantity $F$ is then given by

$$\langle F \rangle = \sum_m a_m \langle m | F | m \rangle.$$ \hspace{1cm} (1)

Here we have assumed that the state vectors are normalized to unity $\langle m | n \rangle = \delta_{mn}$, and that the probability $q_m$ has the property $q_m \geq 0$.
and the normalization

$$\sum_{m} a_m = 1.$$  \hspace{1cm} (2)

It is convenient to describe such a system by means of the density operator or statistical operator \( \rho \), where

$$\rho = \sum_{m} \vert m \rangle a_m \langle m \vert.$$  \hspace{1cm} (3)

The average value of the observable \( F \) is the trace of \( \rho F \):

$$\langle F \rangle = \text{Tr} \rho F.$$  \hspace{1cm} (4)

Often a density matrix with off diagonal elements is formed by expanding the state vectors

$$\vert m \rangle = \sum_{p} c_{p_m} \vert p \rangle,$$  \hspace{1cm} (5)

where the quantities \( c_{p_m} \) are the expansion coefficients. Substituting this into the density operator gives the expression

$$\rho = \sum_{m, n, q} \vert q \rangle c_{q_m} a_m c_{p_m}^{*} \langle p \vert.$$  \hspace{1cm} (6)

The quantity \( c_{q_m} a_m c_{p_m}^{*} \), or some such variation, is sometimes referred to as the off diagonal density matrix.
CHAPTER 2

THE GENERALIZED STATISTICAL OPERATOR

We find that the conventional definition of the off diagonal density matrix is too restrictive. As far as its effect on the system goes, it essentially retains its diagonal character. Therefore, we shall depart from convention and define a time dependent off diagonal statistical operator that is explicitly capable of moving the system from one state to another. This is something that the conventional off diagonal density matrix is incapable of doing because of its role in the diagonal density matrix.

Using (3) and (6) as a guide, we propose to define a generalized off diagonal statistical operator in the following manner

\[ \rho = \sum_{m,n} \hat{\rho}_{m,n(t)} = \sum_{m,n} |m_{\eta}\rangle P \ a_{m_{\eta}(t)} \langle n_{\eta}|. \]  

(7)

The Bra-Kets are the Dirac notation and \( t \) is the time variable. The term \( a_{m_{\eta}(t)} \) is the time dependent system probability of finding a particle, within the system, initially in a state \( m \) (or \( n \)) and finally in a state \( n \) (or \( m \)). The determination of which is the initial state and which is the final state is made by the ordering operator \( P \), whose properties are defined in the following manner. The off diagonal
operator parts will operate on the Hamiltonian in either of two ways. From the left,

\[
\hat{P}_{m|n(t)} H = \left| m_{(t)} \right> \mathcal{P} a_{n|n(t)} \left< n_{(t)} | H = \left| m_{(t)} \right> a_{m|n(t)} \left< n_{(t)} | H . \right.
\]

(8)

In such an expression, we define the effect of the ordering operator by adopting the convention that the first state vector in the statistical operator to operate on the adjacent operator is the initial state. Clearly, the remaining state is the final state. Thus, in (8), \( m \) is the initial state and \( n \) the final state. Similarly, operating on the Hamiltonian from the right gives

\[
\hat{H} \hat{P}_{m|n(t)} = H \left| m_{(t)} \right> \mathcal{P} a_{n|n(t)} \left< n_{(t)} | = H \left| m_{(t)} \right> a_{m|n(t)} \left< n_{(t)} | . \right.
\]

(9)

Now \( m \) is the initial state and \( n \) the final state.

Notice that in (9), we have now used \( \mathcal{P} \) to also order the indices on the system probabilities. In other words, we now use a common convention that the first index designates the final state and the second the initial state. The appearance of the double indices on \( a_{m|n(t)} \) or \( a_{n|m(t)} \) serves to take into account not only the probability of finding the system in some initial state, but also to weight the probability that the system can exist in the final state. The nature of the final state weighting is determined by the statistics of the particles involved. Thus, consideration of the number of particles in the final state would be necessary for Fermions but unnecessary for Bosons or in the limit of classical statistics. Since most of what follows
encompasses only the latter two cases, the subscripts on the system
probabilities denoting the final state will be dropped.

Defining the statistical operator elements as we have done, in
no way alters the form or content of the diagonal elements from their
conventional definition. This is seen from

\[ \hat{\mathcal{D}}_{m_m(t)} = |m(t)\rangle \langle m(t)| = |m(t)\rangle a_{m(t)} \langle m(t)| . \]  

(10)
The ordering operator has no effect on the diagonal elements. How­
ever, the off diagonal elements are very different from the convention­al definition, which is based essentially on an expansion of the
diagonal elements.

As we have defined it, the off diagonal statistical operator is
only a mathematical construct. It needs to operate on some other quan­
tity just to determine which states are to be the initial states and
which are to be the final states. Later we will show in more detail
how it is related to the more familiar diagonal form. The off diagonal
statistical operator will be shown to provide a description of the
transitions that account for the fluctuations in averaged value quan­
tities. These averaged value quantities are inherently concerned with
sums over final states as they exist the instant the measurement obser­
vation is made. The motivation for defining the off diagonal statisti­
cal operator as we have is the opinion that the transitions that
produce the fluctuations in the averaged value quantities are best de­
scribed by generalizing the statistical operator used to calculate
these averaged quantities.
The introduction of the ordering operator $\mathcal{P}$ into the defini-
tion of the generalized statistical operator stems from the need to
take into account initial and final states of the transitions as they
occur in the system. This is particularly true in the case of Fermion
systems where the augmented definition of the system probability
is needed to weight both the initial and final states. Because it is
concerned with transitions within the system, it will be shown that the
off diagonal statistical operator elements arise quite naturally in
conjunction with the interactions responsible for the transitions.

Although this dissertation will concern itself solely with
fluctuations in the equilibrium or steady states, where the concepts of
a diagonal statistical operator and averaged value quantities are
valid, there appears to be no obvious reason why the generalized off
diagonal statistical operator cannot be extended to the more challeng-
ing cases of nonequilibrium phenomena.
CHAPTER 3

DERIVATION OF RATE EQUATIONS

Having arrived at a generalized form of the statistical operator and rules governing its use, it is now necessary for us to apply it in a practical way. The quantities normally of most interest and most accessible to physical observation are the diagonal matrix elements $\rho_{nn}$, which represent probabilities and for which the following well-known relations hold\(^1,3\)

\[
\text{Tr} \rho = \sum_n \langle n| \hat{\rho}_{nn(t)} |n\rangle = \sum_n \rho_{nn} = \sum_n a_n = 1
\]

and

\[
\langle F \rangle = \text{Tr} F \rho = \sum_n F_{nn} a_n ,
\]

provided that $F$ has only diagonal matrix elements.

In order to relate the off diagonal operator elements $\hat{\rho}_{mn(t)}$ to the diagonal form, we multiply $\hat{\rho}_{mn(t)}$ by the projection operator of the final states and sum over all the initial states. It remains only to select the final state, which is done through the ordering operator $\mathcal{P}$. For example, arbitrarily choosing $n$ as the final state, we have

\[
\sum_m |n\rangle \langle n| \hat{\rho}_{mn(t)} = \sum_m |n\rangle \langle n| \mathcal{P} a_{nn(t)} |n\rangle = |n\rangle a_{nn(t)} \langle n| = \hat{\rho}_{nn(t)}
\]

so that, of course

\[
a_{nn(t)} = \rho_{nn} = \langle n| \hat{\rho}_{nn(t)} |n\rangle = \rho_{nn} .
\]
The ensemble average of $F$, as given by (12), can then be expressed in terms of the off diagonal statistical operator:

$$\langle F \rangle = \text{Tr} \left( \sum_{n} F |n \rangle \langle n| \right) = \text{Tr} \left( \sum_{m,n} F |n \rangle \langle n| \rho_{nm} \right)$$

$$= \text{Tr} \left( \sum_{m,n} F |n \rangle \langle n| \rho_{nm} \right) \alpha_{nm} \langle n| = \sum_{n} \alpha_{nm} \langle n| \right) = \sum_{m} \alpha_{nm} \langle m| \right) = \sum_{n} \alpha_{nm} \langle n| \right) \rho_{nm} \right) = \sum_{m} \alpha_{nm} \langle m| \right) \right) \right)$$

$$= \text{Tr} \left( \sum_{m,n} F |n \rangle \langle n| \rho_{nm} \right) \alpha_{nm} \langle n| = \sum_{n} \alpha_{nm} \langle n| \right) = \sum_{m} \alpha_{nm} \langle m| \right) = \sum_{m} \alpha_{nm} \langle m| \right) \rho_{nm} \right) = \sum_{m} \alpha_{nm} \langle m| \right) \rho_{nm} \right)$$

Here we have arbitrarily called $n$ the final state. However, we could just as well have called $m$ the final state and written

$$\langle F \rangle = \text{Tr} \left( \sum_{m,n} F |m \rangle \langle m| \right) = \text{Tr} \left( \sum_{m} F |m \rangle \langle m| \right) \alpha_{mm} \langle m| = \sum_{m} \alpha_{mm} \langle m| \right)$$

$$\left( \right)$$

We now want to calculate the time derivative of the expectation value of the specific observable quantity, the number density $N$.

$$\frac{d}{dt} \langle N \rangle = \text{Tr} \frac{d}{dt} \left( N \rho \right) = \text{Tr} \frac{dN}{dt} \rho + \text{Tr} \left. N \frac{d}{dt} \rho \right).$$

Here we have used the operator notation because we are going to interpret the trace operation which gives the observable average as being a sum over final states of the system as they exist the moment the observation is made. The situation that lends itself most readily to solution is the equilibrium condition which requires that

$$\frac{d}{dt} \alpha_{nm} = 0 = \frac{d}{dt} \alpha_{nm} \quad \text{and} \quad \frac{d}{dt} \langle m| = 0 \right).$$

Using the Liouville equation, we can now evaluate $\frac{d}{dt} \rho$ by first evaluating $\frac{d}{dt} \rho_{mn}(t)$ and then transforming the off diagonal statistical operator elements to diagonal form by projecting out the final states and summing over the initial states. Applying the
Liouville equation to \( \frac{d\hat{\rho}_{mn}(t)}{dt} \) gives
\[
\frac{d\hat{\rho}_{mn}(t)}{dt} = \frac{\partial \hat{\rho}_{mn}(t)}{\partial t} + \frac{i}{\hbar} \left[ \hat{H}, \hat{\rho}_{mn}(t) \right].
\] (19)

In what follows, we propose to include in the commutator only those interaction Hamiltonians imposed on the system by the external world, and to avoid confusion we will write this as \( \hat{H} = \hat{H}^e \). Next, consider the term \( \frac{\partial \hat{\rho}_{mn}}{\partial t} \). Let this reflect the changes produced in the system by processes internal to the system, such as relaxation and spontaneous fluctuations. We postulate that \( \frac{\partial \hat{\rho}_{mn}}{\partial t} \) has the same form as the commutator of external interactions with \( \hat{\rho}_{mn} \). We write this as
\[
\frac{\partial \hat{\rho}_{mn}}{\partial t} = \frac{i}{\hbar} \left[ \hat{H}^i, \hat{\rho}_{mn} \right],
\] (20)
where \( \hat{H}^i \) is the internal interaction Hamiltonian. We now want to evaluate expression (17) to find \( \frac{d\langle N \rangle}{dt} \). We will assume that the only changes in number density with time are due to spontaneous fluctuations, so that \( \frac{d\langle N \rangle}{dt} \propto (\Delta N) \omega \) where \( \omega \) is the transition probability per particle per unit time and \( \Delta N \) is the number of fluctuating particles, and typically \( \Delta N = N^{1/2} \). But \( \omega \) involves the matrix elements squared and we will show next that \( \frac{d\rho}{dt} \) involves only matrix elements to the first power. This enables us to neglect the term \( Tr \frac{d\rho}{dt} \rho \) on the basis that
\[
\frac{d\langle N \rangle}{dt} \rho \ll N \frac{d\rho}{dt}.
\] (21)

Treating the trace operation as a sum on final states, we have
\[
\frac{d\langle N \rangle}{dt} = Tr N \frac{d\rho}{dt} = \sum_{final} \left( N \frac{d\rho}{dt} \right)_{final}.
\] (22)
Recalling the formalism for converting the off diagonal statistical operator elements to diagonal form by projecting out the final states and summing over the initial states, and keeping in mind the role of the ordering operator in selecting initial and final states, we have

\[
\text{Tr} \frac{\partial \rho}{\partial t} = \frac{i}{\hbar} \text{Tr} \left[ \sum_{m,n} \left( N_{nm} \delta_{nm} a_m a_n^{\dagger} \langle m | H | n \rangle - \sum_{n} a_n^{\dagger} a_n \langle m | H | n \rangle \right) \right].
\]

But for the first term inside the parentheses \( m \) is determined to be the initial state, while for the second term \( n \) is the initial state. This leads to the expression

\[
\text{Tr} \frac{\partial \rho}{\partial t} = \frac{i}{\hbar} \text{Tr} \left[ \sum_{m,n} \left( N_{nm} \delta_{nm} a_m a_n^{\dagger} - \sum_{n} a_n^{\dagger} a_n \delta_{nm} \langle m | H | n \rangle \right) \right]
\]

\[
= \frac{i}{\hbar} \sum_{m,n} \left( N_{nm} a_m H_{nm} - N_{nm} a_n H_{nm} \right). \tag{24}
\]

One result immediately available from equation (24) is that the diagonal Hamiltonian matrix elements will not directly produce changes in the number density. Thus, if we let \( H = H_0 \) and use the energy representation,

\[
H_{nm} = E_m^0 \delta_{nm}, \tag{25}
\]

then

\[
\frac{\partial \langle N \rangle}{\partial t} = 0. \tag{26}
\]

Thus, the off diagonal elements of \( H \) are those which lead to changes. On the other hand, the transitions within the system occur randomly in time, and at any instant the two terms of (24) should not be expected
to be precisely correlated. This constitutes a fluctuation about the
equilibrium or steady state values, and will be dealt with in later
chapters.

The appearance of fluctuations suggests we re-examine the ini-
tial assumptions that $\frac{dq_{nm}}{dt} = 0$ and $\frac{d}{dt} |n\rangle = 0$. We propose to
treat the number density as typically a large number. In this light,
we can hold that the random fluctuation in the system probability
and the effect of fluctuation in the wave function are of second order
as compared to the fluctuations in the number density. We will neglect
these second order effects.

Equation (24) has several distinguishing features. First, it
involves transition amplitudes, not matrix elements squared. This is
due to the Liouville equation, which expresses the time derivative in
terms of transition amplitudes. The second feature is the mixing of
initial and final state values in each term, and reflects the way in
which the off diagonal statistical operator was defined. Further, at
equilibrium where there are no externally imposed interactions, $H^E = 0$, the equation is invariant under time reversal. For, if we replace $t$ by
$-t$, we must also make the replacement $\hat{\rho}_{mn} \rightarrow \hat{\rho}_{nm}$, which will leave
the basic equation (24) unaltered, implying that a time reversal under
equilibrium conditions will produce no physically observable results.
The remainder of this dissertation deals with the application of this
equation to a number of physical problems.
CHAPTER 4

THE RESULTS OF BOLTZMANN AND PLANCK

In this chapter, the Boltzmann result for the ratio \( \frac{q_m}{q_n} \) and Planck's law for photon number density are derived. We begin by considering the fundamental equation (26) for \( \frac{d\langle N \rangle}{dt} \), and, since the problem at hand is one of equilibrium, we set the external interaction Hamiltonian equal to zero, \( H^e = 0 \). While the value of \( \frac{d\langle N \rangle}{dt} \) will fluctuate with time, its average value taken over a suitable period of time must be zero at equilibrium. This can be written as

\[
\frac{d\langle N \rangle}{dt} = 0 = \frac{i}{\hbar} \sum_{m,n} \left( a_m N_{nn} H_n^e - a_n N_{mn} H_m^e \right),
\]  

(27)

where the averaging over an extended period of time is denoted by the overhead bar, in contrast to the instantaneous system average denoted by the brackets \( \langle \rangle \). Further, under equilibrium conditions all transitions out of a given state to another state must be matched by an equal number of reverse transitions. This implies that each term in the above equation (27) must separately sum to zero, so we have

\[
0 = a_m N_{nn} H_n^e - a_n N_{mn} H_m^e.
\]  

(28)
For observations over an extended period of time, we can interpret this to require that

\[ a_m N_{nm} \frac{\mathcal{H}_{nm}^x}{N_{nm}} = a_n N_{mn} \frac{\mathcal{H}_{nm}^x}{N_{nm}} \]

(29)

where \( \mathcal{H}_{nm}^x \) signifies all the internal interaction terms allowed the system. This leads to the relation

\[ \frac{a_n}{a_m} = \frac{N_{nm}}{N_{mn}} \]

(30)

which is just the Boltzmann condition relating, at equilibrium, the ratio of the probabilities of finding a particle in states \( n \) and \( m \) to the ratio of the particle number densities in these respective states.

Contained in this derivation is the subtle point that for the Boltzmann result to hold, the two states must be connected by some form of interaction. The definition of the system probabilities, \( a_m \), can readily be extended to include systems with two or more types of distinct particles. We could, then, easily imagine a system composed of two different types of noninteracting particles. In this case, the Boltzmann result will not generally hold if it involves the ratio of a system occupancy probability characteristic of one set of particles to that of the other set.

The derivation of Planck's law parallels what has just been done. Again we assume that, at equilibrium and over some interval of time, every transition from state \( m \) to \( n \) must be matched by the
reverse transition \( n \) to \( m \). Then, as before,

\[
a_m N_{nm} H^x = a_n N_{nm} H^x .
\]

(31)

Now, since we are dealing with photons, let \( N_{nm} \) and \( N_{nm} \) be the photon numbers and \( H^x \) the photon electron interaction. Then, by the well-known selection rules governing first order photon electron interaction,

\[
N_{nn} = N_{nm} \pm 1 .
\]

(32)

Taking the case for which \( N_{nn} = N_{nm} + 1 \) and proceeding as before, we have

\[
\frac{a_m}{a_n} = \frac{N_{nn}^*}{N_{nm}} = \frac{N_{nm} + 1}{N_{nm}} .
\]

(33)

But at equilibrium

\[
A_m = \frac{e^{-\beta E_n}}{\sum_m e^{-\beta E_m}} ,
\]

(34)

where \( \beta = \frac{1}{kT} \) and \( E_m \) is the energy of the \( m \) \( \pi \) state, so that

\[
N_{nm} = \frac{l}{e^{-\beta (E_n - E_m)} - 1} .
\]

(35)

However, for \( N_{nn} = N_{nm} + 1 \) there has to be the emission of a photon, and the particle (atom) makes a transition from the higher energy state \( m \) to the lower energy state \( n \). Setting \( \hbar \omega_{nm} = E_n - E_m \), we obtain Planck's law

\[
N_{nm} = \frac{l}{e^{\beta \hbar \omega_{nm}} - 1} .
\]

(36)
We have so far developed two familiar and fundamental laws of equilibrium statistical physics. Rather than proceed further in this fashion, we will now turn to integrating, with respect to time, the basic rate equation (26). From this result we will derive a generalized expression for fluctuations correlated both with respect to time and space, and apply it to a number of examples.
In this chapter, we will develop a general expression for the average number of correlated pairs of transitions occurring in a system of particles either in equilibrium or in a steady state. By particles we mean either individual atoms or molecules, and for simplicity we will consider systems composed of only one type of particle. The results can be easily extended to systems of several different particle types. Our starting point is equation (24), which describes the time derivative of the average number density

\[ \frac{d\langle N \rangle}{dt} = \frac{i}{\hbar} \sum_{m,n} \left( a_n N_{mm} \langle \psi_m | H | \psi_n \rangle - a_m N_{nn} \langle \psi_m | H | \psi_n \rangle \right). \]  

In order to save space, all of the interaction terms to which the system is subject have been included in the symbol $H$.

Integration of this equation with respect to time gives an expression for $\langle N_{(t)} \rangle$ that explicitly contains terms responsible for fluctuations

\[ \langle N_{(t)} \rangle = \langle N_0 \rangle + \frac{i}{\hbar} \sum_{m,n} \left( \int_0^t a_n N_{mm} \langle \psi_m | H | \psi_n \rangle dt' \right) - \int_0^t a_m N_{nn} \langle \psi_m | H | \psi_n \rangle dt'. \]

The first term on the right hand side is the average number density

\[ \langle N_0 \rangle = \sum_m a_m N_{mm}, \]
and arises as the constant of integration. The second term contains
the fluctuations about the average number density. We interpret
\( \langle N(t) \rangle \) as meaning the instantaneous ensemble average of the quantity
\( N(t) \). As such, it is subject to fluctuations due to the occurrence of
random transitions within the system. Since the system probability and
number density in any state are averaged quantities, we can treat them
as constants or slowly varying quantities and take them out of the in­
tegrals. This amounts to neglecting the higher order contributions to
their own fluctuations, a procedure which is acceptable for systems
with a large number of particles but may be suspect for systems with
only a small number of particles. The instantaneous average number
density can now be written as

\[
\langle N(t) \rangle = \langle N_0 \rangle + \frac{\epsilon}{k} \sum_{m,n}(a_n N_{nm} \int_0^t \langle \psi_m \mid H \mid \psi_n \rangle dt' - a_m N_{mn} \int_0^t \langle \psi_m \mid H \mid \psi_n \rangle dt').
\]

At equilibrium, the fluctuations average to zero. This can be
seen by recalling that under equilibrium conditions \( a_m N_{nm} = a_n N_{nm} \).
The time integrals of the transition amplitudes are equivalent, so that
while the transitions can occur at different times, it is only neces­
sary to average over a sufficiently long time interval to find that the
difference between the number of transitions from state \( m \) to \( n \) and
that from state \( n \) to \( m \) approaches zero. The time averaging of
\( \langle N(t) \rangle \) over the time interval \( T \) can be written as

\[
\frac{1}{T} \int_0^T \langle N(t) \rangle dt = \langle N_0 \rangle + \frac{\epsilon}{k} \sum_{m,n}(a_n N_{nm} \int_0^t \langle \psi_m \mid H \mid \psi_n \rangle dt' - a_m N_{mn} \int_0^t \langle \psi_m \mid H \mid \psi_n \rangle dt').
\]
After a long enough time $\tau$, the difference between the two terms in the brackets will become zero, yielding the necessary result

$$\frac{1}{\tau} \int_0^\tau \langle N(t) \rangle \, dt = \langle N_0 \rangle.$$  \hspace{1cm} (41)

For the steady state, the situation is more complicated. The time average of the instantaneous number density must still be a constant. However, the number of transitions from any state $\eta$ to any other state $\eta'$ no longer has to equal the number of inverse transitions from the state $\eta'$ to the state $\eta$. Furthermore, the steady state is usually maintained by some external source which must have fluctuations of its own. These also will be added to the system fluctuations.

In order to obtain a measure of the number of correlated pair transitions per unit time, we will define the observable quantity

$$\left( \overline{\Delta N} \right)^2 = \frac{\langle N(t+\tau) N(t) \rangle - \langle N \rangle \langle N \rangle}{\tau} = \frac{\langle N(t+\tau) N(t) \rangle - \langle N \rangle^2}{\tau}.$$  \hspace{1cm} (42)

$\tau$ is the correlation time for which the fluctuations are observed. In many cases of physical interest, $\tau$ can be set equal to zero. The double bar denotes an average in time and a correlation in coordinate space in a manner which will be discussed in detail. The correlation in coordinate space is included because the interactions within the system are supposed to involve individual particles, among which there has to be some degree of common excitation if correlations are to occur. We will assume that the system's density and organization is such
that only correlations between pairs of particles are important, al-
though we will later consider several cases where this is not so.

Substituting the appropriate expressions for $\langle N_{i0} \rangle$ from (39),
and explicitly showing the volume and spatial correlation integrals, we
obtain the following:

$$\frac{\langle N \rangle}{t} = \frac{i}{\hbar^2 V^2 \tau} \int d^3 \mathbf{q} \int d^3 \mathbf{r} \sum_{s, \bar{s}} \left[ \left( a_{s(r)} N_{s(r)} \int_{\tau}^{\tau+\tau} \langle \psi_s | H | \psi_r \rangle \right)^2 d\tau - a_{s(r)} N_{s(r)} \int_{\tau}^{\tau+\tau} \langle \psi_s | H | \psi_r \rangle dt \right]$$

$$\chi \left[ a_{ms} N_{ms} \int_{\tau}^{\tau+\tau} \langle \psi_m | H | \psi_s \rangle dt - a_{ms} N_{ms} \int_{\tau}^{\tau+\tau} \langle \psi_m | H | \psi_s \rangle dt \right]$$

where now the overhead bar just symbolizes the time average. Note that
the number densities and system probabilities are evaluated at the mo-
ment the transition takes place. $V$ is the system volume and $d^3 \mathbf{q}$ is
the differential element of that volume. Therefore, the integration
over $d^3 \mathbf{q} / V$ is an average over $V$. The integration over $d^3 \mathbf{r} / V$ repre-
sents the average effect of the spatial correlations arising from the
spatial dependence of the system wave functions $\psi$. The specific man-
ner in which this can be done is discussed in detail later.

We will now assume that each type of transition has its own
characteristic rate and that this is different from all the other tran-
sition rates in the system. This will lead to a desirable simplifi-
cation. Let the time averaging be done over a long enough time
interval so that the only contributions come from transitions occurring
at exactly the same rates. This eliminates the random correlations between different rates, and in doing so acts like a pair of delta functions relating \( \gamma \) and \( S \) with \( n \) and \( m \). The summation in equation (43) is now greatly simplified, and it can be rewritten as

\[
\frac{\langle \Delta N \rangle^2}{\tau} = \frac{i}{\hbar^2 V^2} \int d^3 \rho \int d^3 \xi \left[ \sum_{m,n} (a_{m(0)} a_{n(0)} N_{mm} N_{nn}) \int_0^t \int_0^t \langle \psi_m | H | \psi_n \rangle dt \langle \psi_m | H | \psi_n \rangle dt' \right]
\]

\[
- a_{m(0)} a_{n(0)} N_{mm} N_{nn} \int_0^t \int_0^t \langle \psi_m | H | \psi_n \rangle dt \langle \psi_m | H | \psi_n \rangle dt'
\]

\[
- a_{m(0)} a_{n(0)} N_{mm} N_{nn} \int_0^t \int_0^t \langle \psi_m | H | \psi_n \rangle dt \langle \psi_m | H | \psi_n \rangle dt'
\]

\[
+ a_{m(0)} a_{n(0)} N_{mm} N_{nn} \int_0^t \int_0^t \langle \psi_m | H | \psi_n \rangle dt \langle \psi_m | H | \psi_n \rangle dt'
\]

The time integration which is signified by the overhead bar must still be done.

Let us now consider in detail the first term in equation (44). Results from the other terms will follow as an easy adaptation from this. First, the single particle interaction potential will be taken to have the time dependent form

\[
H_{(\tau)} = H^0 e^{\frac{\tau i \Delta E \tau}{\hbar}}
\]
where $H^0$ is time independent and the choice of sign depends upon whether energy is emitted or absorbed in the interaction. Next, consider the wave function $\psi$. The system is composed of a number of single particles which are distributed in some manner in the coordinate space of the system. Since we are dealing with ensemble averages for which the probabilities of the internal states of a single particle are given by $Q_m$, we can write the system wave function as a product of a single particle part which describes the internal states times a spatial part which describes the distribution of the particles in space.

$$|\psi_{\text{total}}\rangle = |\psi_{\text{single particle}}\rangle |\psi_{\text{space}}\rangle.$$ (46)

And

$$|\psi_{\text{single particle}}\rangle = |m\rangle e^{-iE_m \tau / \hbar}.$$ (47)

The ket $|m\rangle$ is assumed independent of time. The spatial wave function has the following property

$$\langle \psi_{\text{space}}, \psi_{\text{space}} \rangle = \frac{i}{\sqrt{V}} \int d^3 q \sum_{k=1}^{N_0} \delta(q - q_{\text{spatial}})$$

$$= N_0$$

where $N_0$ is the total number of particles in the system. The transition matrix elements become

$$\langle \psi_{\text{total}}, H|\psi_{\text{total}} \rangle = \frac{i}{\sqrt{V}} \int d^3 q \ H^0_{mn} e^{i(E_m - E_n \Delta E) \tau / \hbar} \sum_{k=1}^{N_0} \delta(q - q_{\text{spatial}})$$

$$= N_0 H^0_{mn} e^{i(E_m - E_n \Delta E) \tau / \hbar}.$$ (49)
We can now write the first term in the correlated pair fluctuation expression as

\[ \frac{1}{\sqrt{\pi \hbar}^2} \int d^2q \sum_{m,n} \frac{a_{m_{\infty}} a_{m_{\infty}}^* N_{n_{\infty}} N_{n_{\infty}}^* |H_{m_{\infty}}|^2}{x} \times \]

\[ \times \int d\xi \int d\eta e^{i(\xi - \eta + \Delta \xi)} \int e^{-i(\xi - \eta + \Delta \xi)} \sum_{k,l} \delta(\xi + q - \eta - q_k) \delta(q - q_l), \]

Since we have assumed that the interaction potential is not a function of a particle's location in space, it can be taken out of the spatial integrals. We can now define the conditional pair distribution function as

\[ G(\xi, \tau, \tau) = \frac{1}{\sqrt{\pi}} \int d^2q \sum_{k,l} \delta(\xi + q - \eta - q_k) \delta(q - q_l), \]  \hspace{1cm} (51)

where we have explicitly shown the time dependency. \( G(\xi, \tau, \tau) \) is the conditional probability of having encountered a particle at time \( \tau \) and location \( \eta \), and of then finding a second particle at a time \( \tau + \tau' \) later and at a distance \( \xi \) away from the first. \( \tau' \) equals correlation time and \( \xi \) the correlation distance. This definition of \( G(\xi, \tau, \tau) \) is similar to one given by L. Van Hove \(^4\) (See also Münster \(^5\)), and is related to the pair correlation function of the virial expansion. \(^6\) Under the conditions of equilibrium or steady state, we are going to assume that \( G(\xi, \tau, \tau) \) is independent of time, allowing us to drop the argument \( \tau \). This allows the particle positions to vary with time, but holds the probability of a double encounter independent of time.
For a more complete discussion of $G_{(c,\tau)}$, we refer the reader to either of the above-mentioned references or, for a slightly different application, to some of the papers of Kirkwood. Because it is independent of time, $G_{(c,\tau)}$ can come out of the time average, leaving the first term in the form

$$\frac{1}{\tau} \int dt \sum_{m,n} a_{m,c} a_{m,c}^\dagger N_{m,c} N_{m,c}^\dagger |H_{m,n}|^2 G_{(c,\tau)} \times$$

$$\times \frac{1}{t} \int_0^t dt' \int_0^{t+\tau} dt'' e^{i(E_m-E_n+i\Delta E)\frac{t'}{\hbar}} e^{-i(E_m-E_n+i\Delta E)\frac{t''}{\hbar}} .$$  (52)

The time average can be handled in the following manner. We first evaluate the time integrals due to the interaction

$$\frac{1}{t} \int_0^t dt' \int_0^{t+\tau} dt'' e^{i\beta t'} e^{-i\beta t''} = \frac{\epsilon \hbar^2}{t \beta^2} e^{i\beta t} \sin^2 \frac{\beta t}{2\hbar} ,$$  (53)

where

$$\beta = (E_m - E_n + \Delta E) .$$

This result can be put into more manageable form by noting that generally the time $t$ is sufficiently long so that $\frac{\sin^2 \frac{\beta t}{2\hbar}}{t \beta^2}$ is a sharply peaked function. Making use of the relation

$$\lim_{t \to \infty} \frac{1}{t} \frac{\sin^2 \frac{\beta t}{2\hbar}}{t \beta^2} = S(\beta)$$  (54)
we have
\[
\lim_{t \to \infty} 2\pi \hbar \left( \frac{1}{\hbar} \frac{\sin^2 \frac{\theta t}{\hbar}}{\frac{\hbar}{\Theta}} \right) e^{-i \frac{\Theta^2}{\hbar}} = 2\pi \hbar \delta(\Theta) e^{-i \frac{\Theta^2}{\hbar}}
\]
\[
= 2\pi \hbar \delta(\Theta).
\]

Taking the time average is now a simple process and the first term of equation (44) can be written in a final form as

\[
\int d^2 \mathbf{r} \sum_{m, n} a_{m(\omega)} a_{n(\gamma)} N_{m_{(\omega)}} N_{n_{(\gamma)}} \frac{2\pi}{\hbar} |H_{mn}|^2 \delta(\epsilon_m - \epsilon_n + \Delta \epsilon) G(\xi, \gamma)
\]

since
\[
|H_{mn}|^2 = |H_{mn}|^2.
\]

With each term appropriately modified, the expression for the average correlated number fluctuation per unit time can be written as

\[
\frac{(\Delta N)^2}{t} = \int d^2 \mathbf{r} \sum_{m, n} \left[ a_{m(\omega)} a_{n(\gamma)} N_{m_{(\omega)}} N_{n_{(\gamma)}} \frac{2\pi}{\hbar} |H_{mn}|^2 \delta(\epsilon_m - \epsilon_n + \Delta \epsilon) G(\xi, \gamma)
\]

\[
- a_{n_{(\omega)}} a_{m(\gamma)} N_{m_{(\omega)}} N_{n_{(\gamma)}} \frac{2\pi}{\hbar} |H_{mn}|^2 \delta(\epsilon_m - \epsilon_n + \Delta \epsilon) G(\xi, \gamma)
\]

\[
- a_{m(\omega)} a_{n_{(\gamma)}} N_{m_{(\omega)}} N_{n_{(\gamma)}} \frac{2\pi}{\hbar} |H_{mn}|^2 \delta(\epsilon_m - \epsilon_n + \Delta \epsilon) G(\xi, \gamma)
\]

\[
+ a_{n(\omega)} a_{m(\gamma)} N_{m_{(\omega)}} N_{n_{(\gamma)}} \frac{2\pi}{\hbar} |H_{mn}|^2 \delta(\epsilon_m - \epsilon_n + \Delta \epsilon) G(\xi, \gamma) \right). \]
What we have developed is a theory of correlated pair fluctuations which depends mainly on the averaged values of equilibrium or steady state quantities. The fluctuations arise from correlated quantum transitions between like states in pairs of particles separated in space by a distance $\gamma_c$. The probability of a transition occurring is just the standard quantum mechanical expression. The probability of two correlated transitions is taken into account by the conditional pair correlation function $G_{\tau_1, \tau_2}$ which measures the chance of two particles seeing essentially the same interaction potential.

The complete expression for the number of correlated pairs of transitions is a sum of four terms, each of which contains the above-mentioned probabilities and each distinguished from the others by the four ways of finding the energy states of the two correlated particles. Of these terms, the first and fourth describe correlated transitions that can be detected external to the system, as they represent a correlated energy emission or lack of emission from the system.

The effects of the second and third terms are more subtle. They represent a continuous interchange of energy between particles within the system and not observable outside it. On the average, they conserve energy but need not do so for short time intervals. At any instant of time, some amount of energy attributable to these fluctuations must be in "flight" between particles and, in a way, lost to the particles which comprise the system.

We shall apply the general expression for the number of correlated pair transitions to several diverse examples, and show that it is
capable of deriving previously-known results and of predicting new results as well.

**The Conditional Pair Probability Function**

Before going on, it is worthwhile to digress and investigate, in a limited way, the function $G(r, t)$, the conditional probability of finding a pair of particles a distance $r$ and a time $t$ apart, and also the correlation integral defined as:

$$\int G(r, t) \, d^3r = \frac{1}{V^2} \int d^3\xi \sum_{\ell,j} \delta(\xi - \frac{q}{\delta} - \frac{q_{\ell}(t)}{\delta_{\ell(0)})} \delta(q - q_{\ell(0)}) \quad (59)$$

Van Hove has devoted a considerable amount of effort to the study of the conditional pair probability function. It is not our intention to repeat in any detail what has been done, but rather to try to give a physical picture of this expression and, in one limiting case, establish an acceptable approximation. Van Hove points out that the above expression cannot rightly be integrated with respect to $d^3q$ because the coordinate operators belong to different times and do not commute.

We will try to avoid this difficulty by a more intuitive approach. The correlation integral is applied to all of the particles in the system. However, it can equally well be written in terms of the correlation integral for one typical particle multiplied by the number
of particles in the system, so that

\[
\int G(\zeta, \tau) \, d^2 \zeta = \frac{N_0}{\Delta V} \int \overline{G(\zeta, \tau)} \, d^2 \zeta. \tag{60}
\]

Here \( \overline{G(\zeta, \tau)} \) is the conditional probability of encountering an initial particle, and of then finding a second particle at a distance \( \zeta \) and a time \( \tau \) removed from the first. \( N_0 \) is the number of particles in the system and \( \Delta V \) is the volume of integration in which the correlation is to occur. In the limit as \( \Delta V \) grows to include all regions of the system, the number of correlations with the original particle is \( N_0 - 1 \), which is just the number of particles in the system that can form pairs with the initial particle; therefore

\[
L = \lim_{\Delta V \to V} \frac{N_0}{\Delta V} \int \overline{G(\zeta, \tau)} \, d^2 \zeta = N_0 (N_0 - 1). \tag{61}
\]

This is the expected result, but it is of little use in systems in which the interactions that the individual particles are subjected to do not extend equally to all particles in the system.

The essence of the spatial structure of any substance which we shall consider will appear in the conditional probability and the correlation integral. For example, a well-ordered crystalline array would give quite different values for the correlation integral if the direction of correlation were taken only along the crystal planes or if it were taken along some other direction that seldom encountered a second particle. The same holds true for the even more highly ordered systems encountered in biology: the correlated integral will be highly
direction-dependent. Certain phase transitions will also affect the correlation integral and the conditional pair probability. However, all these systems are unduly complicated for our purposes.

We will restrict ourselves to the consideration of systems whose particle locations in coordinate space are purely random or sufficiently dense so that the correlation integral is independent of direction. But direction still has an important role to play, as will be found later when we consider correlations involving photons or phonons. In most of the cases which we will consider, the correlation volume element \( d^3 r \) can be written as

\[
d^3 r = \Delta A \, d^3 r,
\]

where \( \Delta A \) is typically the cross section for the correlating interaction. The correlation integral then becomes

\[
\int \overline{G_{(r, \tau)}} \, d^3 r = \int_0^l \overline{G_{(r, \tau)}} \, \Delta A \, d^3 r,
\]

where \( l \) is the mean path length.

In order to put this integral into a form that we can integrate, we will allow the correlation volume to extend over the mean path length for the correlating interaction. The mean probability of finding a particle along this path becomes equal to unity. But this is very much the same as the conditional pair probability: having found one particle, how far do you have to go to find a second particle? We will now assume that for this case the correlation integral can be replaced by its average value, and in particular, the average value of
the conditional pair probability is set equal to unity. This approxi-
mation gives the result

$$\int \overline{G(r_\xi, r)} d^3 \xi = \Delta A \lambda$$

(64)

In a later chapter, we shall give a justification for terminating the
integral at a mean path length, and show that this assumption leads to
reasonable results.
CHAPTER 6

CORRELATED PHOTON FLUCTUATIONS

In the next few chapters, we will apply the generalized pair correlation expression to several examples. We will begin by considering a gas at equilibrium or in steady state with photon radiation. We will determine the correlated fluctuations in photon number due to first and second order effects in the photon electron interaction. The first order terms involve single annihilation and creation operators, and rather than writing out the expression for the correlated number density fluctuations per unit time, it is more instructive to present it in a symbolic form, as shown in Figure 1 on the following page. The middle photon appearing in the picture represents, symbolically, the presence of stimulating photons. Each term represents, symbolically, the corresponding term in the general expression for the number of correlated pairs of photons occurring under the photon electron interaction. The area enclosed by the dashed lines represents the region over which the stimulating photon is felt. It is in this region that $G(v_j, r)$ represents the probability of finding a second particle to excite. The separation time $τ$ between the two interactions shown in the graphs is just the correlation time. The second and third terms do not contribute to the number of correlations that would be seen by an observer external to the system, since these photons do not leave the system. The external observer could only detect the presence of correlations in
\[ \frac{(\Delta N)^2}{t} = \sum \left\{ \text{correlated pair creation} \right\} \]

\[ \text{stimulator} \]

\[ \text{correlated creation and annihilation} \]

\[ \text{correlated annihilation and creation} \]

\[ \text{correlated annihilation} \]

Fig. 1. Pictorial representation of correlated fluctuations due to photon annihilation and creation processes.
the first and fourth terms. If we imagine that the external observer views the system through a filter or some other device that allows him to look at emission lines due to individual transitions, we can drop the summation signs in the fluctuation expression. In addition, we will allow the observer the ability to look for correlations at any correlation time \( \tau \) and with an instrument that has a resolution time \( \Delta \). The number of correlated pairs of externally observable photons for any transition is given by (58) to be

\[
\langle \Delta N_{nm} \rangle^2 = \int_{\Delta} d\varepsilon' \left( a_{m,\varepsilon'} a_{m,\varepsilon} N_{nn_{\varepsilon'}} N_{nn_{\varepsilon}} \frac{2\pi}{\hbar} |H_{nm}|^4 \delta(\varepsilon_{m}-\varepsilon \pm \Delta \varepsilon) G(\varepsilon,\tau) \right) + a_{n,\varepsilon} a_{n,\varepsilon} N_{mm_{\varepsilon}} N_{mm_{\varepsilon}} \frac{2\pi}{\hbar} |H_{nm}|^4 \delta(\varepsilon_{m}-\varepsilon \pm \Delta \varepsilon) G(\varepsilon,\tau) \tag{65}
\]

From Figure 1, we can interpret the first term of equation (65) above as describing correlated emission and the fourth term as describing correlated absorption occurring in transitions between the \( n \) and \( \eta \) energy levels. We will next show that the first term leads to the experimental results of Brown and Twiss. ^8

**The Brown Twiss Experiment**

Consider a gas of radiating atoms. An observer sits outside the radiating system and measures the arrival times of the emitted photons. If the radiating system were purely random, there should be only a small chance of correlations occurring in arrival time. Brown and Twiss observed significant correlations in arrival time.
We propose the following interpretation of this result. At any instant of time, the atoms in the system are surrounded by a random photon bath. If an atom in such a surrounding can be stimulated to emit a photon, then, by the nature of the photon electron interaction, the emitted photon will have the same wave vector and polarization as the stimulating photon and, for a short time, the photon distribution in that region of space will no longer be random. If another emitter can be found and stimulated within this time, there exists an increased likelihood that its photon will be correlated with the first. Generally, the random nature of the photon distribution is quickly restored by photon absorption. Still, a significant number of correlated photons are able to escape the system to produce correlated photon pairs at a remote detector.

Since we are interested in correlations between two photons, we need consider only the first term in equation (65), which we will write as

$$\frac{(\Delta N_{m,n})^2}{t} = a_{m_{(o)}, n_{(o)}} N_{n_{(o)}} N_{n_{(o)}} \frac{2\pi}{\hbar} |H_{nm}|^2 \delta(\epsilon_m - \epsilon_n - \hbar \omega_{nm}) \int G_{(n, n)} d\gamma$$

(66)

where

$$\hbar \omega_{nm} = \epsilon_m - \epsilon_n = \Delta \epsilon$$

or pictorially

![Diagram]

$N_{n_{(o)}} = N_{n_n}$ is the average photon number density after the first
interaction. After the second interaction, the average photon number density is \( N_{n_{(r)}} = N_{n_r} + 1 \). The interaction matrix element squared is taken to be that of the photon electron interaction with a slight modification

\[
\sum_{\text{polarizat}n_{n_{m}}} \frac{e^\hbar}{2\hbar \sqrt{N_{n_{m}} \hbar \omega_{n_{m}}}} \times \sqrt{N_{n_{(r)}} (N_{n_{(r)}+1}) \hbar \omega_{n_{(r)}}} \times \langle \langle n | e^{i \mathbf{k}_{n_{(r)}} \cdot \mathbf{r}} e^{i \mathbf{k}_{n_{(r)}+1} \cdot \mathbf{r}} | m \rangle | D_{(\epsilon_{n_{(r)}} - \hbar \omega_{n_{(r)}})} \rangle
\]

\[= \left( \frac{\tau_{n_{(r)} m}}{2} \right)^{-1} \] (67)

The modification is the presence of the term \( \sqrt{N_{n_{(r)}} (N_{n_{(r)}+1}) \hbar \omega_{n_{(r)}}} \) instead of the term \( \sqrt{N_{n_{m}} \hbar \omega_{n_{m}}} = N_{n_{m}} \hbar \omega_{n_{m}} \) that is normally present in the square of the matrix element describing emission of a single photon by a single particle. It arises from the need to take into account the effect of the additional photon in stimulating the emission of the second photon. For the sake of convenience, the correlation probability has been written so as to define an inverse mean lifetime for forming correlated pairs, \( \left( \frac{\tau_{n_{(r)} m}}{2} \right)^{-1} \). The factor of two has been included to account for both directions of photon polarization. It would not appear for polarized photons. We note that the correlated photons have identical wave vectors \( \mathbf{k} \) and polarization \( \alpha \). The average number of correlated photon pairs, with frequency \( \omega_{n_{m}} \) and correlation time \( \tau \), emitted per unit time per unit volume of the source becomes

\[
\frac{(\Delta N_{n_{m}})^2}{t} = a_{n_{m}} a_{n_{(r)}} N_{n_{(r)}} (N_{n_{(r)}+1}) \frac{2}{\hbar \epsilon} \int G_{(\epsilon, \tau)} d^3 \mathbf{r} \] (68)
Experimentally, what is observed are the number of correlated photon pairs with a correlation time $\tau$ arriving at a detector usually remotely located from the source. In their experiment, Brown and Twiss divided the incoming photon beam with a half-silvered mirror, and then measured the correlations in photon arrival time between the divided beams. In order to compare the above result with experiment, we must express it in terms of the quantities observed at the detector.

The number of photons $N$ arriving at the instrument in a time $T$, in terms of the number of particles present in the source $N_o$, the probability $a_m$ that these particles will be in a state $m$ to emit a photon, the probability for emission, and the attenuation $\alpha$ between the source and detector, is given as

$$N = \alpha T a_m N_o \frac{2\pi}{\hbar} |H_{mn}|^2 \delta(\varepsilon_m - \varepsilon_n + i\omega_{mn}) \quad (69)$$

But for emission

$$\frac{2\pi}{\hbar} |H_{nm}|^2 \delta(\varepsilon_m - \varepsilon_n + i\omega_{nm}) = \frac{2 N_{mn}}{\tau_{nm}^*} \quad (70)$$

where $\tau_{nm}^*$ is the mean lifetime of the $m$ to $n$ transition when no stimulating photons are present. The average number of photons arriving at the detector in a time interval $T$ becomes

$$N = \alpha \left[ \frac{2 N_{mn} a_m N_o}{\tau_{nm}^*} \frac{T}{\tau_{nm}^*} \right] \quad (71)$$

Each of the split beams experiences an additional attenuation due to
the beam splitter. Let the attenuation of the beam splitter be \( \beta_1 \) and \( \beta_2 \) for beams 1 and 2, respectively. Then the total attenuation experienced by photon beam 1 is

\[ \alpha_1 = \beta_1 \alpha \]  

and the total attenuation of beam 2 is

\[ \alpha_2 = \beta_2 \alpha \]  

Because of the nature of the beam splitter,

\[ \beta_1 + \beta_2 = 1. \]  

Denoting the average number of photons that pass down beams 1 and 2 by \( N_1 \) and \( N_2 \), from continuity requirements it follows that

\[ N = N_1 + N_2. \]  

The number of correlations in the arrival of these photons with a correlation time \( \tau \) is

\[ \left( \Delta N_{(\tau)} \right) = \left( \Delta N_{1(\tau)} + \Delta N_{2(\tau)} \right). \]  

The cross correlation between photon beams 1 and 2 follows directly as

\[ \left( \Delta N_{1(\tau)} \Delta N_{2(\tau)} \right) = \frac{1}{2} \left[ \left( \Delta N_{1(\tau)} \right)^2 - \left( \Delta N_{1(\tau)} \right)^2 - \left( \Delta N_{2(\tau)} \right)^2 \right]. \]  

The photons arriving at the detector will exhibit the same correlation behavior as at the source, except that allowance has to be made for attenuation. The number of photon pair correlations arriving at the detector in a time interval \( T \) is given by

\[ \left( \Delta N_{(\tau)} \right) = \alpha N_{nm} \left( \alpha N_{nm} + 1 \right) a_{m\tau} b_{n\tau} \frac{2 T}{\tau_{mn}} \int_{\alpha^2} G_{mn\tau}^2 d\tau_\tau. \]
The split beams experience the additional attenuation of the beam splitter, and their pair fluctuations are

$$\overline{\left(\Delta N_{1,\nu}\right)^2} = \alpha \beta, N_{n\nu} \left(\alpha \beta, N_{n\nu} + 1\right) a_{m_{1\nu}} a_{m_{2\nu}} \frac{2T}{\tau_{nm}} \int_{\Delta\nu} G_{\nu_{1\nu}, \nu_{2\nu}} d^3 \xi, \quad (79)$$

and

$$\overline{\left(\Delta N_{2,\nu}\right)^2} = \alpha \beta, N_{n\nu} \left(\alpha \beta, N_{n\nu} + 1\right) a_{m_{1\nu}} a_{m_{2\nu}} \frac{2T}{\tau_{nm}} \int_{\Delta\nu} G_{\nu_{1\nu}, \nu_{2\nu}} d^3 \xi. \quad (80)$$

Substituting these equations into the cross correlation expression gives

$$\overline{\left(\Delta N_{1,\nu} \Delta N_{2,\nu}\right)} = a_{m_{1\nu}} a_{m_{2\nu}} \frac{T}{\tau_{nm}} \left[N_{nm}^2 \alpha^2 \left(1 - \beta_1^2 - \beta_2^2\right)\right] \int_{\Delta\nu} G_{\nu_{1\nu}, \nu_{2\nu}} d^3 \xi. \quad (81)$$

Introducing the expression for the number of photons arriving at the detector, as given in equation (72), the above result can be expressed as

$$\overline{\left(\Delta N_{1,\nu} \Delta N_{2,\nu}\right)} = \frac{1}{\nu T} \left(N_o \right)^2 \frac{\left(T_{nm}^c\right)^2}{\tau_{nm}^c} \left(1 - \beta_1^2 - \beta_2^2\right) \int_{\Delta\nu} G_{\nu_{1\nu}, \nu_{2\nu}} d^3 \xi. \quad (82)$$

Recall that $N$ is the average number of photons at the detector, and $N_o$ is the number of particles at the source. The mean lifetime with no stimulators present, $\tau_{nm}^o$, is readily seen from (67) and (70) to be related to the mean correlated lifetime

$$\tau_{nm}^c = \frac{\tau_{nm}^o}{\sqrt{N_{nm} N_{nm} + 1}}. \quad (83)$$
Finally, if we let the beam splitter form the two beams with equal intensities,

\[ N_1 = N_2 = \frac{N}{2} \]  

and

\[ \beta_1 = \beta_2 = \frac{1}{2} \]  

we obtain from (82) a result similar to that of Purcell in his elementary treatment of the Brown Twiss effect

\[
\left< \Delta N_{(\tau)} \Delta N_{(\tau)} \right> = \frac{N^2}{2} \left( \frac{\int N_{(\tau)}^2 N_{(\tau)}^2 \tau_{mn} \int G_{(\tau)} \, d^3 \zeta}{N_0^2} \right). \tag{86}
\]

For the term in the brackets, Purcell obtains a correlation time \( \tau_0 \), which, he states, is approximately the reciprocal of the band width. Our result is a bit more complicated, but it has the same dependence on \( N \) and \( \tau \).

We can interpret the correlation integral performed over all particles in the system as equivalent to the number of particles in the system \( N_0 \) times the average correlation integral for one pair of particles.

\[
\int_{\Delta \tau} G_{(\tau)} \, d^3 \zeta = \frac{1}{\sqrt{2}} \int_{\Delta \tau} \int_{\Delta \zeta} \int_{\Delta \zeta} \sum_{j,k} S_{(\zeta + \tau - \zeta_{1(\tau)})} S_{(\zeta - \zeta_{1(\tau)})}
\]

\[ = N_0 \int_{\Delta \tau} \overline{G}_{(\tau)} \, d^3 \zeta, \tag{87} \]

where \( \overline{G}_{(\tau)} \) is the conditional probability of encountering a single pair of particles a distance \( \zeta \) and a time \( \tau \) removed from each other. Because the observer is only looking for correlations with correlation
times equal to $\tau \pm \Delta$, where $\Delta$ is the resolution time of the measuring instrument and is assumed symmetrically distributed about a time $\tau$, the correlation integration has limits that extend a distance $(\tau - \frac{\Delta}{2})C$ to $(\tau + \frac{\Delta}{2})C$ away from the initial particle. $C$ is the photon velocity in the system. The correlation integral represents the number of particles in this region that can be correlated with the initial particle. Substituting this form into the cross correlation expression gives:

$$
\frac{\overline{\Delta N}_{\tau^+} \Delta N_{\tau^-}}{\frac{N_r^2}{\tau}} \left( \frac{\sqrt{N_{nn}} \epsilon_n \langle N_{nn} \rangle \epsilon_n}{N_0} \right) \int_{\Delta \nu} G_{(\epsilon, \tau)} d^3 \epsilon
$$

In essence, we differ from the Purcell result by the factor

$$
\frac{1}{N_{\nu}} \int_{\Delta \nu} G_{(\epsilon, \tau)} d^3 \epsilon
$$

But it is only natural that this term should appear here since it represents the average probability of encountering a second particle in a volume $\Delta \nu$, separated by a distance $\tau C$ away from the first particle. This is a necessary requirement if the pair of correlated transitions are to occur.

The only way that the correlation time $\tau$ enters into the right hand side of the above equation is through $\overline{G_{(\epsilon, \tau)}}$. This suggests that by measuring the cross correlation function, $\overline{(\Delta N_{\tau^+}) (\Delta N_{\tau^-})}$, as a function of $\tau$, it should be possible to obtain a measure of the average probability of encountering a pair of particles as a function of particle separation, and by so doing, obtain a measure of the particle number density. In practice, the potentially correlating photon can be
absorbed before producing a correlation. This self-absorption by the
system has to be taken into account by modifying the cross correlation
integral to include an exponential decay
\[
\frac{N_{m}^{2}}{2T} \left( \frac{\sqrt{\frac{N_{n}^{2} (N_{m}^{2})_{2,1} \tau_{nm}^{\circ}}{N_{0}}} \int_{0}^{\tau_{c}} G_{\xi} \, d\xi \right) e^{-\frac{2 \tau_{c}}{\lambda_{nm}}} \), (90)
\]
where \( \lambda_{nm} \) corresponds to the mean optical path length for the
transition.

For simple systems in which the particles are distributed ran-
domly and with uniform density throughout the system volume, the prob-
ability of finding a second particle in a volume \( \Delta V \) at a distance \( \tau_{c} \)
from the initial particle is a constant. This implies that the average
correlation integral is a constant. If, in addition, the state prob-
ability \( Q_{m} \) does not depend on the particle's location within the sys-
tem, the mean optical path length \( \lambda_{nm} \) can be directly determined by
knowledge of the cross correlation function \( (\Delta N_{\xi}, \Delta N_{\tau}) \) as a
function of correlation time. From this information, we can arrive at
the average particle number density and, with knowledge of the amount of
light output, we can determine the temperature of the radiating system.

In the case of a more realistic system, such as a distant star,
neither the particle number density nor the probability \( Q_{m} \) are uni-
form in space, presenting a more complicated problem. However, it may
be that self-absorption takes place over a relatively short distance
where these quantities are essentially uniform in space, and the anal-
ysis of the simple system can then be applied.
In order to determine the mean optical path length accurately, the resolution time \( \Delta \) of the measuring apparatus must be such that

\[ \mathcal{C} \Delta \ll \lambda_{nm}. \]

The same would hold true if the system had any structure.

**Photon Bunching**

We now want to briefly consider the concept of photon bunching, and we begin by examining the second term in equation (65) for observable correlations. This term describes the fluctuations due to correlated absorption which, to the observer, would appear as a correlated interval in time when he would observe no photons. It has already been shown that the lifetime of a correlated pair emission is

\[ \tau_{nm}^c = \frac{\tau_{nm}^{\circ}}{\sqrt{N_{nn} r_{\alpha} (N_{nn} + 1) r_{\alpha}}} \]  

(83)

while the lifetime for single photon emission is

\[ \tau_{nm} = \frac{\tau_{nm}^{\circ}}{N_{nn} r_{\alpha}} \]  

(91)

This leads to the relation

\[ \tau_{nm}^c = \frac{\sqrt{N_{nn} r_{\alpha}}}{\sqrt{(N_{nn} + 1) r_{\alpha}}} \tau_{nm} \]  

(92)

implying that photons are most likely to appear bunched together since

\[ \tau_{nm}^c < \tau_{nm} \]. It is just as easily shown that the same relation holds for correlated photon pair absorption, which correspondingly implies that "holes" in the photon arrival times are also bunched together. It
is of interest to consider the ratio of the two terms in the photon number fluctuation (65), that is, the ratio of the correlated number fluctuation of photons, denoted by \( \frac{(\Delta N_x^2)}{t} \text{photon} \), to that of the correlated number fluctuation of holes, \( \frac{(\Delta N_x^2)}{t} \text{hole} \):

\[
\frac{(\Delta N_{x,\text{photon}})^2}{(\Delta N_{x,\text{hole}})^2} = \frac{a_m \cdot a_w}{a_w \cdot a_m} \left( \frac{N_{nn}(N_{nn}+1)}{(N_{nn}+1)N_{nn}} \right). \tag{93}
\]

We have hesitated to perform the obvious cancellation of the photon number densities, because the above expression is valid at the source location only. At a remote detector, attenuation will affect the number of photon arrivals, and the numerator only must be appropriately modified. Making the assumption that \( a_{m,\text{photon}} = a_{m,\text{hole}} \), and denoting the attenuation effects by \( \alpha \), we can write that

\[
\frac{(\Delta N_{x,\text{photon}})^2}{(\Delta N_{x,\text{hole}})^2} = \frac{a_m^2}{a_n^2} \frac{\alpha (\alpha N_{nn}+1)}{N_{nn}+1} e^{-\frac{2\tau_x}{\Delta n_m}}. \tag{94}
\]

Because of attenuation effects above, the number of correlated intervals when no photons arrive will predominate over the number of correlated pairs of photons that are observed at the detector. It should be possible to experimentally observe this phenomena, although we have not been able to find such experimental data in the literature.
CHAPTER 7

CORRELATED PHOTON FLUCTUATIONS DUE TO SCATTERING

We now consider fluctuations due to photon electron scattering. The scattering process, of course, involves the second order terms in the photon electron interaction Hamiltonian. By symbolically writing the fluctuations, we are led to Figure 2 on the following page. The other diagrams are easily inferred. Each one symbolizes a process which is independent of the other fluctuation processes. All of the remarks concerning the figure of the first order processes apply here.

Consider now an experiment similar to that of Brown and Twiss. An observer is located external to a system in which there is considerable scattering, and measures the correlated number fluctuations of the photons that leave the system. Again, assume that he looks at photons of only one frequency, \( \omega_{nm} \). For photon pair correlation, only the first term in the expression for \( \frac{\langle \Delta N \rangle^2}{\tau} \) is applicable.

Writing out a typical term in the pair correlation expression, we get

\[
\frac{\langle \Delta N \rangle^2}{\tau} = \int d_\tau^3 a_{\nu(\omega)} a_{\nu(\tau)} N_{\nu\nu(\omega)} N_{\nu\nu(\tau)} \frac{2\pi}{\hbar} |H_{nm}|^2 \delta(\varepsilon_m - \varepsilon_n - \hbar \omega_{nm}) G_{(\omega, \tau)}, \tag{95}
\]

where

\[ N_{\nu\nu(\omega)} = N_{\nu\nu} \quad \text{and} \quad N_{\nu\nu(\tau)} = N_{\nu\nu} + 1. \]

Rather than writing the complete expression for \( |H_{nm}|^2 \), we will just extract the photon number dependent part in order to illustrate
\[
\frac{(\Delta N)}{t} = \sum \left\{ \begin{array}{l}
\text{correlated creation scattering} \\
\text{and annihilation scattering} \\
\text{and creation scattering} \\
\text{correlated annihilation scattering} \\
\text{and creation scattering} \\
\text{correlated annihilation scattering} \\
\text{scattering}
\end{array} \right\}
\]

Fig. 2. Pictorial representation of correlated scattering processes.
the dependence of the correlated fluctuation on the number density.

For photon electron scattering, we have that

\[ |H_{n,m}|^2 = N_{n,m} \left( N_{n,m} + 1 \right) |\overline{H}_{n,m}|^2. \] (96)

Here \( \overrightarrow{k} \) is the wave vector and \( \alpha \) is the polarization.

**The Laser Operating Below Threshold**

We are going to restrict the problem somewhat by considering a specific case, the laser, initially operating below threshold but still with a high enough photon number density to justify scattering effects predominating over the effects of photon annihilation and creation. Because of the geometry of the laser, the photon number density along the axis of the laser is very nearly equal to the average photon number density in the laser,

\[ N_{n,m} = N_{n,m}. \] (97)

In addition, we will allow only the conditional pair probability function \( g_{(r, \tau)} \) to be dependent on particle separation. We will assume that because the photon number density is so high, scattering is almost a certainty, and the correlation integral becomes solely dependent on the interparticle separation. The correlated photon fluctuations of the laser output beam can be written as

\[ \frac{(\Delta N)^2}{\tau} = \left[ N_{n,m} (N_{n,m} + 1) \right]^2 a_{m(r)} a_{m(r)} R(\omega) \int G_{(r, \tau)} d^2 \tau \] (98)

where \( R(\omega) \) is a constant and represents the cumulative effect of all
the terms contributing to pair correlations, from which we have extracted and explicitly written out the photon number density. We have arrived at the prediction that for laser systems operating below threshold but with a large photon number density, the correlated fluctuations per unit time is very closely proportional to the fourth power of the number density. This assumes that the system is in a situation where the occupation probability \( A_m \) is only weakly dependent on photon number.

There is some evidence that this may indeed be correct. A paper by C. Freed and H. Haus,\textsuperscript{11} describing a photon counting experiment on a gas laser, contains a plot, shown in Figure 3, of the ratio of excess noise to shot noise, \( S_e/S_s \), versus the photoelectron count rate occurring in a photomultiplier monitoring the laser output.

\begin{figure}[h]
\centering
\includegraphics[width=0.8\textwidth]{fig3.png}
\caption{Ratio of excess noise to shot noise versus laser power.\textsuperscript{11}}
\end{figure}
The excess noise fluctuations are due to the fluctuations in the number of photons arriving at the photocathode. The number of photoelectrons per second is directly proportional to the photon number density in the laser.

In examining the graph, look in the region labeled Below Threshold, and look, not at the solid or dashed curves, but at the actual data points. On the log log plot, a straight line with a slope of 4 makes an excellent fit to all the data points. Over the region of fit, the photoelectron count rate and, hence, photon number density in the laser, changes only by about a factor of 6. We can then assume that the occupancy probabilities change only slightly so near laser threshold and, further, that the shot noise in the photomultiplier remains nearly constant. The experimental data of Freed and Haus provides support extending over three orders of magnitude of noise fluctuations that \( \frac{(\Delta N)^2}{\tau} \propto N_n^a \). It must be mentioned however, that Freed and Haus suggest an alternative explanation, that of contributions from other laser modes, to explain the data points. This explanation accounts for the dashed curve in Figure 3.

**Correlated Fluctuations in Photons Emitted Off the Direction of the Laser Beam**

It is also possible to predict that for observations made off the direction of the laser beam, the correlated fluctuations in photon number density are very nearly proportional to the square of the laser photon number density. This is readily accomplished by noting that for the typical laser, the number of photons moving in any direction but the
laser beam is very small. Indeed, after the emission of a photon in an off beam direction, the number of photons in this direction and in the immediate area of the emitter is one. The probability of a correlated emission of a second photon is thus proportional to

\[ N_{n,n+1} \mid \overline{H}_{nm} \mid^2 = 2 \mid \overline{H}_{nm} \mid^2. \]  

The correlated fluctuations in an off beam direction are

\[ \frac{(-N)^2}{t} = a_{m_{t_0}, m_{t_\tau}} N_{nn}(N_{nn+1}) \frac{2\pi}{\hbar} N_{n,n+1} \mid \overline{H}_{nm} \mid^2 \int G(r, r') d^2 r, \]

\[ = a_{m_{t_0}, m_{t_\tau}} N_{nn}(N_{nn+1}) \frac{2\pi}{\hbar} (2) \mid \overline{H}_{nm} \mid^2 \int G(r, r') d^2 r, \]

which is just the result we set out to show.

**A Simple Description of Laser Operation Based on Fluctuations**

Next, we will illustrate a process by which the essentially random photons in the laser operating below threshold are converted into the highly coherent photon beam that characterizes the laser operating above threshold. In particular, we will show how the correlated fluctuations provide a simple account of the laser behavior as it goes from below threshold into a lasing condition.

Below threshold, the photons traveling up and down the laser tube are nearly randomly spaced in time. Some of these photons are correlated with others to form correlated photon pairs, but the correlation time is a random variable. Now consider that happens as the photon number density is gradually increased. We have already shown
that the correlated photon number fluctuations increase as the fourth power of the photon number density. However, these correlated photons are also being increasingly bunched together as the photon number density increases, because the probability of correlated scattering is proportional to

\[ N_{n\eta} \left( N_{n\eta} + 1 \right) \approx N_{n\eta}^2. \tag{101} \]

The time between two correlated photons is proportional to the inverse of the correlated scattering probability, so that the correlation time is shrinking as the inverse square of the photon number density. The correlation and bunching effects increase rapidly (\( N_{n\eta}^1 \) and \( N_{n\eta}^2 \), respectively) with increasing photon number density, until a number density is reached where a significant number of correlations between three or more photons begin to occur. This new effect further increases the local photon number density, and now regions of increased photon number density, in which the photons are all correlated, are developing in the laser. The system is now in an unstable state. Increasing the photon number density increases the photon bunching, which makes it increasingly likely that scattering will be stimulated to take place within the bunched regions. All these scattered photons will be correlated with the photons of the bunched region, thus further augmenting their number density. With increasing photon number density, the process goes quickly to completion. The laser will contain a number of regions that move along its axis and in which the photon number density is very large. All of these photons are correlated or, what amounts to the same thing, in phase. The system is now lasing.
There are two aspects of this concept of laser operation that are subject to observation. First, that the laser output should consist of a series of light pulses and second, that below threshold the correlation time should vary inversely as the photon number density.

There are some experimental observations available which support both of these hypotheses. A. L. Schawlow\textsuperscript{12} cites an experiment in which researchers at the Bell Telephone Laboratories, using a special high speed photographic technique, have photographically recorded the output of a laser as a series of intense light pulses. For the second point, the bunching of correlated photons as the photon number increases, we return to the paper of Freed and Haus and suggest an interpretation of their experimental results, which are shown in Figure 4. They have presented data on the bandwidth of the "excess noise" spectrum, or what we would call fluctuation spectrum, as a function of photon number density. But the bandwidth of the "excess noise" is directly proportional to the number of correlations with mean correlation time $\tau_{nm}^c$, where

$$\left(\tau_{nm}^c\right)' = \frac{2\pi}{\hbar} \left| H_{nm} \right|^2 \delta_{\left(\epsilon_m - \epsilon_n - \hbar \omega_{nm}\right)}$$

$$= \frac{2\pi}{\hbar} N_{nn_{c-1}} \left(N_{n+1}\right)_{\alpha\alpha} \left| H_{nm} \right|^2 \delta_{\left(\epsilon_m - \epsilon_n - \hbar \omega_{nm}\right)}.$$  

We have extracted the explicit photon number dependency from the interaction matrix element, and have denoted what remains as $\left| H_{nm} \right|^2$. What is important here is that the mean correlation time $\tau_{nm}^c$ at high photon number densities is inversely proportional to the square of the
photon number density. Therefore, the number of correlations with correlation time $\tau$, or in other words, the bandwidth of the fluctuations, is also inversely proportional to the photon number density squared. Now examine Figure 4 in the region labeled Below Threshold.

![Diagram](image)

**Fig. 4.** Bandwidth versus laser power.

If we consider only the experimental data points, we find that these are closely fit on the log log plot by a straight line with a slope that is inversely proportional to the square of the photon number density. This seems to agree with their data at least as well as the dashed line which is based on additional modes contributing.

Finally, in the region above threshold, the laser operates as a laser, its photons bunched together into regions of high number densities of in phase photons. The previous analysis which dealt with photon pair correlations probably does not hold here. It would appear more appropriate to treat the photon bunches as separate entities,
probably with a Poisson distribution, and apply an analysis similar to that of Freed and Haus to this mode of laser operation.
We will next consider correlated phonon number fluctuations as they occur in a solid. We begin by writing the correlated fluctuations in terms of correlations with respect to a single particle, keeping just those parts of the general expression that contribute to the observable effects.

\[
\frac{\langle \Delta N \rangle^2}{t} = \int d^3 \mathbf{r} \sum_{m,n} a_{m(\tau)} a_{n(\tau)} N_{m(\omega)} N_{n(\omega)} \frac{2\pi}{h} |H_{nm}|^2 \delta (\varepsilon_m - \varepsilon_n - k \cdot \mathbf{u}_{nm}) \overline{G_{(\varepsilon, \tau)}}
\]

\[
+ \int d^3 \mathbf{r} \sum_{m,n} a_{n(\tau)} a_{m(\tau)} N_{n(\omega)} N_{m(\omega)} \frac{2\pi}{h} |H_{nm}|^2 \delta (\varepsilon_m - \varepsilon_n - k \cdot \mathbf{u}_{nm}) \overline{G_{(\varepsilon, \tau)}}
\]

\( (103) \)

For fluctuations in the entire system, this expression must be multiplied by the total number of particles, \( N_p \). We will see later how this factor arises in a familiar manner.

We note that in observing phonon number fluctuations in a solid, we typically observe all the fluctuation processes simultaneously, not selectively, as was done in the case of photon fluctuations. This permits a simplification of the fluctuation expression because both
terms contribute equally to the overall fluctuations, so that

\[
\left( \frac{\Delta N}{t} \right)^2 = 2 \int d^3 \mathbf{q} \sum_{n, \omega} a_{\mathbf{m} \omega}^* a_{\mathbf{m}' \omega} N_{\mathbf{m} \omega} N_{\mathbf{m}' \omega} \frac{2\pi}{\hbar} |H_{\mathbf{m} \mathbf{m}'}|^2 \delta(\epsilon_{\mathbf{m}'} - \epsilon_\mathbf{m} - \hbar \omega_{\mathbf{m} \mathbf{m}'}) G_{\mathbf{m} \mathbf{m}'}(\tau) .
\]

(104)

**Fluctuations at High Temperature**

First, consider the high temperature region where fluctuations caused by scattering phenomena dominate over those caused by annihilation and creation processes. To facilitate matters, we will write the inverse mean lifetime of a state \( \mathbf{m} \) going to a state \( \mathbf{n} \) because of a scattering transition in which no additional stimulating phonons are present as

\[
\left( \tau_{\mathbf{n} \mathbf{m}}^\circ \right)' = \frac{2\pi}{\hbar} |H_{\mathbf{n} \mathbf{m}}|^2 \delta(\epsilon_{\mathbf{n}'} - \epsilon_\mathbf{n} - \hbar \omega_{\mathbf{n} \mathbf{m}}) .
\]

(105)

The inverse mean lifetime of state \( \mathbf{n} \) undergoing a transition to state \( \mathbf{n} \) when there are \( N_{\mathbf{n} \mathbf{n}_{\mathbf{k} \mathbf{x}}} \) stimulating phonons present is found to be

\[
\left( \tau_{\mathbf{n} \mathbf{n}}^\circ \right)' = \left( N_{\mathbf{n} \mathbf{n}_{\mathbf{k} \mathbf{x}}} \right)^2 \left( \tau_{\mathbf{n} \mathbf{m}}^\circ \right)' .
\]

(106)

In an analogous way, the inverse mean lifetime for a pair of correlated scattering transition is given by

\[
\left( \tau_{\mathbf{n} \mathbf{n}}^c \right)' = N_{\mathbf{n} \mathbf{n}_{\mathbf{k} \mathbf{x}}}^2 \left( N_{\mathbf{n} \mathbf{n}_{\mathbf{n}+1}}^c \right) \left( \tau_{\mathbf{n} \mathbf{n}}^\circ \right)' .
\]

(107)
It is appropriate to consider this latter term when dealing with correlated fluctuations, enabling us to write the correlated number density fluctuations as

\[
\frac{\langle \Delta N \rangle^t}{t} = 2 \int d^3 \zeta \sum_{\vec{m}, \vec{n}} a_{\vec{m}, \omega} a_{\vec{n}, \omega} N_{\vec{m}, \omega} N_{\vec{n}, \omega} \left( \tau_{\vec{m}, \omega}^c \right)^t C(\tau_{\vec{m}, \omega}^c, \tau_{\vec{n}, \omega}^c).
\] (108)

If we restrict ourselves to equilibrium, we can use the Planck distribution for the equilibrium phonon number density

\[
N_{\vec{m}, \omega} = \frac{1}{e^{\frac{\hbar \omega}{kT}} - 1}
\]
and

\[
N_{\vec{n}, \omega} = N_{\vec{m}, \omega} + 1 \approx \frac{1}{e^{\frac{\hbar \omega}{kT}} - 1}
\]
for high temperatures. But these are phonon number densities averaged over all directions in space. In particular, the number in any one direction with wave vector \( \vec{K} \) is only a small fraction \( S \) of this overall average

\[
N_{\vec{m}, \omega, \vec{K}} = S N_{\vec{m}, \omega}.
\] (109)

We can now write the inverse mean lifetime for correlated scattering as

\[
\left( \tau_{\vec{m}, \omega}^c \right)^{-1} = S N_{\vec{m}, \omega} \left( S N_{\vec{m}, \omega} + 1 \right) \left( \tau_{\vec{m}, \omega}^r \right)^{-1}.
\] (110)

We will assume that for the typical solid at normal room temperatures,
the phonon number density in any particular direction is less than one, \( N_{n \rightarrow \ell} \ll 1 \). Then

\[
(\tau_{nm}^c)'' \approx SN_{nm}(\tau_{nm}^o)''.
\]

(111)

Next, consider the integration over the correlation variable \( r_c \). We assume that the only term that depends on this variable is the conditional pair probability \( \overline{G_{(r_c, r)}} \), so that the fluctuation expression becomes

\[
\left( \frac{\Delta N}{N} \right)^2 = 2 \sum_{m,n} a_{m(r)} a_{m(r)} N_{n(r)} N_{n(r)} (\tau_{nm}^c)'' \int \overline{G_{(r_c, r)}} d^{3}r_c.
\]

(112)

In doing the integration over the correlation length \( r_c \), we need only consider integrating up to those distances that correspond to the mean scattering length

\[
\ell_{nm} = c \tau_{nm} = c \left( S N_{nm} \right)^2 \tau_{nm}^o
\]

(113)

where \( c \) is the phonon velocity in the system.

The concept of the mean life of a state \( m \) going to a state \( n \) under a scattering transition is, for the case at hand, dependent on phonon number density. This is in contrast to the case of the laser, where the photon number density is so extreme and the scattering probability so high that the correlation lengths depend mainly on the inter-particle separation. For the present case, the upper limit of integration represents the point beyond which the competition from other possible scattering directions has effectively eliminated correlations. Finally, to make the integration tractable, we are going to
make some reasonable determination of the pair probability function, \( \overline{G_c(r_c,r)} \). In particular, we are going to make the simple assumption (discussed on p. 28) that the probability of a phonon finding a second particle within a mean path length of the first is unity. Then, by equation (64), the correlation integral is

\[
\int \overline{G_c(r_c,r)} \, d^2 r_c = \frac{\Delta A}{V} \, c \, \tau_{nm},
\]

(114)

and the fluctuations become

\[
\frac{(\Delta N)^2}{t} = 2 \sum_{m,n} a_{m,o} a_{m,r} N_{n,n_{(o)}} N_{n,n_{(r)}} \left( \tau^c_{nm} \right)' \tau^{-2}_{nm} \frac{\Delta A}{V}.
\]

(115)

But

\[
\left( \tau^c_{nm} \right)' \tau_{nm} = \delta N_n \left( \tau^{-2}_{nm} \right)' \left( \delta N_{nn} \right)^2 \tau_{nm}
\]

\[
= \left( \delta N_{nn} \right)'
\]

(116)

The effects of the interaction have now been essentially eliminated, leaving the fluctuations in the form

\[
\frac{(\Delta N)^2}{t} = 2 \sum_{m,n} a_{m,o} a_{m,r} N_{n,n_{(o)}} c \, \delta \left( \frac{\Delta A}{V} \right).
\]

(117)

We will next use the standard approximation that the particles comprising the system behave like a set of uncoupled harmonic oscillators, enabling the probabilities \( a_m \) to be written as

\[
a_m = e^{-\frac{\hbar \omega}{kT} (m + \frac{1}{2})}.
\]

(118)
where

\[ Z = \left( \frac{2 \sinh \frac{\hbar \omega}{2kT}}{2} \right)^{-1} \]  \hspace{1cm} (119)

Also, we let \( a_{m(\nu)} = a_{m(\nu')} \), which amounts to saying that the probability of finding a particle in state \( \nu \) is independent of the states of other nearby particles. Finally, using the relation

\[ \hbar \omega_{\nu m} = |E_{\nu} - E_{m}| \]

we make the transformation

\[ \sum_{m, \nu} \rightarrow \sum_{m} \int_{0}^{\omega_{\max}} P_{(\hbar \omega)} \cdot d(\hbar \omega) \hspace{1cm} (120) \]

where \( P_{(\hbar \omega)} \) is the density of states. Substituting these into the fluctuation expression, we have

\[ \frac{(\Delta N)^2}{\bar{t}} = 2 \int P_{(\hbar \omega)} d(\hbar \omega) \sum_{m} \frac{e^{-\frac{\hbar \omega}{kT}(m+\frac{1}{2})}}{Z^2} \left( \frac{1}{e^{\frac{\hbar \omega}{kT}} - 1} + 1 \right) \bar{c} S^{-\bar{c}} \frac{\Delta A}{V}. \hspace{1cm} (121) \]

The sum on \( m \) is readily done, and the fluctuation expression is reduced to

\[ \frac{(\Delta N)^2}{\bar{t}} = 2 \int P_{(\hbar \omega)} d(\hbar \omega) \frac{\sinh \frac{\hbar \omega}{2kT}}{\frac{\hbar \omega}{2kT}} \left( \frac{1}{e^{\frac{\hbar \omega}{kT}} - 1} + 1 \right) \bar{c} S^{-\bar{c}} \frac{\Delta A}{V}. \hspace{1cm} (122) \]

Since we are dealing with high temperatures, it is appropriate to take
corresponding limits here, giving

\[
\frac{(\Delta N)^2}{t} \approx 2 \int \left( \frac{\pi \omega}{KR_T} \right)^2 \left( \frac{KT}{\hbar \omega} \right) C \cdot \frac{\Delta A}{V} \rho (\hbar \omega) d(\hbar \omega)
\]

\[
= \frac{1}{2} C \cdot S^{-1} \frac{\Delta A}{V} \left[ 3 \int_{0}^{\omega_{\text{max}}} \frac{\omega^4 d\omega}{(\pi T c)^3} \right].
\]

(123)

In evaluating \( \rho (\hbar \omega) \), we have assumed that all phonon polarizations travel at the same velocity. A more sophisticated analysis can easily be accommodated. The term in brackets is strongly reminiscent of the Debye theory of the specific heat of a solid. In fact, in that theory it represents the number of modes of vibration, \( 3 N_0 \). We can thus follow Debye and write

\[
\frac{(\Delta N)^2}{t} = \frac{3}{2} N_0 C \cdot S^{-1} \frac{\Delta A}{V}
\]

(124)

where \( N_0 \) is the number of particles in the system. But in a time \( t \), the volume occupied by those phonons moving in a direction \( \mathbf{k} \) is given by

\[
SV = \frac{C t \Delta A}{2}
\]

(125)

The factor of two appears because half the phonons move in the opposite direction. Finally, the number fluctuations become

\[
(\Delta N)^2 = 3 N_0
\]

(126)
The significant thing here is that for a solid, the phonon number fluctuations are essentially independent of temperature for high temperatures. This result is reminiscent of the usual statistical mechanical result for an ideal gas.

**Fluctuations at Low Temperature**

Now we will briefly discuss the phonon fluctuation behavior at low temperature. In this limit, phonon annihilation and creation processes predominate over phonon scattering processes, so we will allow only these first order processes to appear in the interactions. Everything proceeds as in the high temperature case, only now the inverse mean correlated lifetime becomes

\[
(T_{nm}^{c})^{-1} = \sqrt{\delta N_{nn}(\delta N_{nn}^{-1})} \left( T_{nm}^{o} \right)^{-1}
\]

\[
\approx \sqrt{\delta N_{nn}} \left( T_{nm}^{o} \right)^{-1}
\]

The inverse mean lifetime when phonon stimulators are present is

\[
(T_{nm}^{s})^{-1} = \delta N_{nn} \left( T_{nm}^{o} \right)^{-1}
\]

In both these cases, \( T_{nm}^{o} \) refers to the mean lifetime of state \( m \) going to state \( n \) via annihilation or creation processes. The
fluctuations in this limit are

\[ \frac{(\Delta N)^2}{\tau} = \sum_m \int p(k\omega) d(k\omega) a_m^2 N_m (N_m+1) \sqrt{S} N_{nn} (\tau_{nn}^0)^2 \frac{1}{S N_{nn}} \tau_{nn}^0 \frac{C \Delta A}{V} \]

\[ = 2 \int p(k\omega) d(k\omega) \frac{\sinh^2 \frac{k\omega}{2 KT}}{\sinh \frac{k\omega}{K T}} \sqrt{\frac{N_{nn}}{S}} (N_{nn}+1) \frac{C \Delta A}{V} \]. \quad (129)

Taking the limit as \( T \to 0 \), the number density becomes

\[ \lim_{T \to 0} N_{nn} = e^{-\frac{k\omega}{K T}} \]

and

\[ \lim_{T \to 0} \frac{\sinh^2 \frac{k\omega}{2 KT}}{\sinh \frac{k\omega}{K T}} = \frac{1}{2} \]

while

\[ \lim_{T \to 0} N_{nn} + 1 = 1. \]

The phonon fluctuations now become

\[ \frac{(\Delta N)^2}{\tau} = \frac{C \Delta A}{V \sqrt{S}} \int_{0}^{\omega_{\text{max}}} 4\pi e^{-\frac{k\omega}{K T}} \frac{\omega^2 \omega}{(2\pi c)^3} \]

\[ = \frac{C \Delta A}{V \sqrt{S}} \frac{4\pi}{(2\pi c)^3} \left( \frac{2 k T}{\hbar} \right)^3 \int_{0}^{\infty} e^{-f} f^2 df. \quad (130) \]

We have again emulated Debye in assuming that at low temperatures the upper limit of integration \( \omega_{\text{max}} \) is effectively so large that it can be replaced by infinity. The correlated number fluctuations follow in
a straightforward manner as

\[
\overline{\langle \Delta N \rangle^2} = \frac{2 \sqrt{3}}{\pi^2} C^3 \left( \frac{2 \kappa T}{\hbar} \right)^3.
\]

(131)

The correlated number fluctuations at low temperatures vanish
as the cube of the temperature. Also of interest is the similarity
between the temperature dependence of the correlated number fluctua-
tions and that of the specific heat.

Finally, we remind the reader that these results have been de-

erived with an approximation to the conditional pair correlation func-
tion \( \overline{G_{(\gamma,\tau)}} \). This approximation seems to work well for high tem-

eratures, but may not be appropriate for low temperatures, points of
change of phase, or anisotropic media. Further, in any of these cases,
anomalous fluctuations may occur at points where the interaction under-
goes a resonance.
CHAPTER 9
THE FLUCTUATION DISSIPATION THEOREM

In this chapter, we slightly modify our general expression for the fluctuations in number density per unit time so that it becomes an expression for energy fluctuations per unit time. The results of this procedure will lead quite naturally to a general expression of the fluctuation dissipation theorem or generalized Nyquist theorem.\textsuperscript{13}

We begin by recalling that the number density fluctuation expression actually represents the mean number of correlated pairs of transitions that take place within a system per unit time. The particles involved in these transitions have some energy, so that if, before performing the sums, we modify the fluctuation expression by inserting twice the energy of the initial state, we will have an expression for the mean correlated energy fluctuation per unit time. The reason for including the factor two is that we are considering fluctuation involving pairs of particles, each having the same energy. The mean correlated energy fluctuation per unit time per particle, obtained by modifying (104), is

\[
\overline{\langle \Delta E \rangle} = 2 \int d^3 \zeta \sum_{m,n} q_{m(l)} a_{m(r)} (2 E_m) N_{n,(l)} N_{n,(r)} \frac{\omega}{\hbar} |H_{nm}|^2 \delta_{\varepsilon_m - \varepsilon_n} G_{(\zeta, \zeta)}
\]

(132)
However, this is simply the power fluctuation

$$\Delta \overline{P} = \overline{\frac{(\Delta E)}{\tau}}.$$  (133)

We will now restrict ourselves to the consideration of voltage fluctuations in the solid state, and make use of the relation involving the power spectrum

$$\overline{\Delta P(\omega)} = \overline{(\Delta V(\omega))^2} / R(\omega)$$

or

$$(\Delta V(\omega))^2 = \Delta P(\omega) R(\omega)$$  (134)

where $R(\omega)$ represents the losses in the system.

The High Temperature Region

For the high temperature region, the fluctuations are caused mainly by scattering processes, so that the integration over the correlation length proceeds exactly as it did for the case of phonon number fluctuations. The result is

$$\overline{\Delta P(\omega)} = 4 \sum_m a^2_m E_m (N(\omega) + 1) \frac{E}{L}$$  (135)

where $L$ is the sample length. To obtain an expression for the mean square voltage fluctuations, we will multiply (135) by $\gamma(\omega)$, the dissipation per particle. Then, for the total mean square voltage fluctuations over the entire system of length $L$, it is necessary to multiply by $N_p$, the number of particles in the sample, and integrate over
the frequency $\omega$,

$$
\langle \Delta \nu \rangle^2 = 8 \int \rho_{(h\omega)} d(h\omega) \sum_m a_m^2 E_m (N_m + 1) R_{(\omega)} \epsilon,
$$

(136)

where $R_{(\omega)} = N_r(\omega)$

The sum on the index $m$ can be considered separately. Using the standard approximation that the system of particles behaves like a set of uncoupled harmonic oscillators, we have

$$
a_m = \frac{e^{-\frac{h\omega}{kT}(m+\frac{1}{2})}}{Z}
$$

(137)

and

$$
Z = \sum_m e^{-\frac{h\omega}{kT}(m+\frac{1}{2})} = \left(2 \sinh \frac{h\omega}{kT}\right)^{-1}
$$

(138)

Then

$$
\sum_m a_m^2 E_m = \frac{1}{Z^2} \sum_m e^{-\frac{2h\omega}{kT}(m+\frac{1}{2})} (m+\frac{1}{2}) \frac{h\omega}{2kT}
$$

(139)

$$
= \left(2 \sinh \frac{h\omega}{kT}\right)^2 \frac{h\omega}{2kT} \left(\frac{1}{\sinh \frac{h\omega}{kT}} + \frac{1}{\sinh \frac{h\omega}{kT}} \right).
$$

In the limit of high temperature, this becomes:

$$
\lim_{T \rightarrow 0} \frac{1}{\omega} \sum_m a_m^2 E_m \approx \frac{h\omega}{kT}
$$

(140)

Finally, we write the density of the states in the linear form

$$
\rho_{(h\omega)} d(h\omega) = 2 \frac{d\omega}{2\pi c}
$$

(141)
because we are interested only in phonon pair correlations along the
direction determined by the two probes that measure the voltage fluctu­
tuations.

Putting these together, we obtain the expression for mean
square voltage fluctuations as

\[
(\Delta V)^2 = \frac{2}{\pi} \int \frac{d\omega}{\omega} (N_{\omega} + 1) R_{\omega} d\omega. \tag{142}
\]

We have, for the moment, refrained from taking the high temper­
atture limit of the phonon number density because we wish to compare
this result with the result of Callen and Welton,\textsuperscript{14} from which it dif­
fers only slightly. They write the term in brackets as \((N_{\omega} + \frac{1}{2})\)
However, in light of the other high temperature approximations already
made, it is inappropriate not to take a similar limit on the bracket
term. Taking the limit results in the well-known Nyquist relation,

\[
(\Delta V)^2 = \frac{2 kT}{\pi} \int R_{\omega} d\omega. \tag{143}
\]

The Low Temperature Region

If we are interested in the voltage fluctuations at low temper­
atture, we must recall that annihilation and creation processes dominate
in this temperature region, and proceed in a manner similar to that
used for low temperature phonon number fluctuations. On completion of
the integration over the correlation length, the voltage fluctuations
at low temperature become

\[ \langle \Delta V \rangle^2 = \gamma \int \rho_{(\omega)} d(\omega) \left( \frac{1}{2} \omega \sin^2 \frac{\hbar \omega}{2kT} + \frac{1}{2} \omega \right) \sqrt{N(\omega) \langle N(\omega) \rangle} \, R(\omega). \]  

(144)

We take the low temperature approximation of these terms in the following manner

\[ N(\omega) \xrightarrow{T \to 0} e^{-\frac{\hbar \omega}{kT}} \]

and

\[ N(\omega)^{+1} \xrightarrow{T \to 0} 1 \]

while

\[ S \omega \hbar \frac{\hbar \omega}{2kT} \xrightarrow{T \to 0} \frac{1}{2} e^{\frac{\hbar \omega}{2kT}} \]

and, of course, it is intended that the low temperature limit of \( R(\omega) \) be taken also. When \( T \to 0 \), we get the following approximate expression for the low temperature mean square voltage fluctuations

\[ \langle \Delta V \rangle^2 = \frac{\gamma}{\pi} \int \hbar \omega \left[ e^{-\frac{\hbar \omega}{kT}} + 2 e^{-\frac{2\hbar \omega}{kT}} \right] R(\omega) \, d\omega. \]  

(145)

The first term in the brackets arises from fluctuations caused by the zero point energy and the second from the higher energy levels in the harmonic oscillators. Significantly, the second term decreases faster than the first term as the temperature goes to zero. If the dissipative term \( R(\omega) \) is reasonably well-behaved, the mean square voltage fluctuations vanish as the temperature vanishes.

One final observation: It has been implicitly assumed throughout this section that the conditional probability of encountering two particles within the allotted correlation time is unity. This seems
like a reasonable assumption at normal room temperatures. But it may not be so at low temperatures or points of changes of phase or, for that matter, any system that is highly ordered in coordinate space. In such highly ordered systems, it may be possible for phonons to propagate long distances in certain directions without ever interacting with a particle. Such a system would show an anomalous fluctuation dissipation relation and also an anomalous specific heat.
In this chapter, we investigate the correlated pair fluctuations in a system in which there is a common agent coupling all of the particles. The prototype system we have in mind is one composed of a collection of magnetic dipoles and subjected to a magnetic field. In order to keep the problem simple, the dipoles will be allowed only two directions in coordinate space, up and down along the Z axis, which will be denoted by + and −, respectively. We will not consider the other types of fluctuations that occur simultaneously with the magnetic fluctuations.

Before calculating the number density fluctuations it is first necessary to have some notion of the type of interaction Hamiltonian which couples the particles and causes them to change their orientation. We propose the following form for this interaction, since it has the appropriate properties and is simple:

\[ H = \gamma \mu B_z (S_+ + S_-) \]

where \( \gamma \) determines the range of the interaction, \( B_z \) is the \( z \) component of the magnetic induction, and \( \mu \) is the magnetic dipole moment. The operators \( S_+ \) and \( S_- \) are the usual raising and lowering
operators, defined by
\[ S_+ = S_x + iS_y \quad \text{and} \quad S_- = S_x - iS_y \]
so that
\[ \langle + | S_+ | + \rangle = 1 \]
while
\[ \langle + | S_+ | + \rangle = 0 = \langle - | S_+ | - \rangle . \]
Similar rules hold for the lowering operator \( S_- \).

In keeping with earlier assumptions, we are assuming that the fluctuations are due not to fluctuations in the occupation probabilities \( q_+ \) and \( q_- \), or to fluctuations in the space correlation function \( G(x, r) \), but to fluctuations in the number of particles that interact. This has a general validity for most macroscopic systems because typically \( (\Delta N)^2 \approx N_e \) where \( N_e \gg 1 \). With such a large number of particles, the state occupancy values will fluctuate very little from their average values.

Taking into account only the two terms that can be seen by an external observer, we can write the number of correlated pair fluctuations as

\[
\frac{(\Delta N)^2}{\epsilon} = 2 \int d^3 \sum_{m_n} \sum_{n_t} a_{m_n} \epsilon_{m_n} N_{n_t} \mu^B \frac{2\pi}{\hbar} |\mu| Y_{\epsilon_{m_n}} \delta_{\epsilon_{m_n}, \epsilon_{m_n} + \epsilon} G_{\epsilon_{m_n}}.
\]

(147)

Since we are dealing with particles, we can write

\[ N_{n_t} = a_n N_e \]

(148)

and

\[ N_{n_t + 1} = N_{n_t} \]

(149)
Because \( N_0 \) is a large number (\( N_0 \approx 10^{20} \)), an excellent approximation even at fairly low temperatures is

\[
N_{n_{(T)}} = N_{n_{(0)}}
\]  

(150)

Letting \( a_{n_{(0)}} = a_{n_{(T)}} \), and assuming that the system is homogeneous and isotropic, we find that the number of correlated pair fluctuations reduces to

\[
\langle (\Delta N)^2 \rangle = 2 \sum_{m,n} a_m^* a_n N_0^2 \mu^2 B_{z,(0)} B_{z,(T)} \frac{2\pi}{\hbar} |Y_{1(r)}(s_{+} + s_{-})| \int d^3 \xi G_{(r)}
\]

(151)

Let us further restrict the problem to the situation in which there is no external magnetic induction, so that the only induction is that generated by the alignment of the particles' magnetic dipole moments

\[
B_z = \mu N_0 (a_+ - a_-)
\]

(152)

and to an excellent approximation

\[
B_{z,(0)} = B_{z,(T)}
\]

(153)

Substituting this into the above equation and doing the sum, which is readily accomplished since it has only two equal terms, we obtain for the number of correlated pair fluctuations

\[
\langle (\Delta N)^2 \rangle = \frac{4}{\hbar} a_+^2 a_-^2 (a_+ - a_-)^2 \left( N_0^{\mu^2} \right) \frac{2\pi}{\hbar} |Y_{1(r)}|^2 \int d^3 \xi G_{(r)}
\]

(154)

At the moment, we are interested in the temperature behavior of the correlated pair fluctuations, which we will assume is solely
contained in the particle occupation probabilities \( a_+ \) and \( a_- \). We will continue to write these terms explicitly. The remainder will be lumped under the symbol \( \Gamma \). We note, however, that

\[
\int G_{(r,r')} \, d^3 \zeta \approx \frac{1}{N_c}
\]  

(155)

since it is approximately the ratio of the correlation volume to the system volume. The correlated pair fluctuation expression can then be rewritten as

\[
\frac{(\Delta N)^2}{\tau} = a_+^2 a_-^2 (a_+ - a_-)^2 \Gamma.
\]  

(156)

We restrict the system once more by allowing only equilibrium conditions. Then

\[
a_+ = \frac{e^{\frac{\mu B_3}{kT}}}{Z}
\]

and

\[
a_- = e^{-\frac{\mu B_3}{kT}}
\]

while

\[
Z = e^{\frac{\mu B_3}{kT}} + e^{-\frac{\mu B_3}{kT}}.
\]

The number of correlated pair fluctuations per unit time becomes

\[
\frac{(\Delta N)^2}{\tau} = \frac{\left(e^{\frac{\mu B_3}{kT}} - e^{-\frac{\mu B_3}{kT}}\right)^2}{Z^2} \Gamma.
\]  

(157)
At low temperatures, but still subject to the conditions that
\[ a_\ast N_\ast = N_\ast \gg 1 \]
the correlated pair fluctuations decrease exponentially with decreasing temperature:
\[ \frac{(\Delta N)^2}{\tau} = e^{-\frac{\mu B_z}{kT} \Gamma} \]  \hspace{1cm} (158)

For the temperature range in which \( a_\ast N_\ast \approx 1 \), it is necessary to take into account the fact that the approximation of equation (150) no longer holds, and that
\[ N_{\ast\tau} = N_{\ast\tau_0} + 1 = a_\ast N_\ast + 1 \]
The fluctuations then become, for this case,
\[ \frac{(\Delta N)^2}{\tau} = \frac{e^{\frac{\mu B_z}{kT}}}{Z^2} \left( \frac{e^{-\frac{\mu B_z}{kT}}}{N_\ast} + \frac{1}{N_\ast} \right) \left( e^{\frac{\mu B_z}{kT}} - e^{-\frac{\mu B_z}{kT}} \right)^2 \]  \hspace{1cm} (159)

Finally, in the temperature range where \( a_\ast N_\ast \ll 1 \), the exponential decrease of the correlated pair fluctuations with decreasing temperature resumes, but now as
\[ \frac{(\Delta N)^2}{\tau} = e^{-\frac{\mu^2 B_z^2}{kT} \Gamma} \frac{\Gamma}{N_\ast} \]  \hspace{1cm} (160)

At high temperatures there is a point, the Curie temperature, where the magnetic field vanishes and with it the correlated pair fluctuations. In this simple model, it readily follows that
\[ T_c = \frac{\mu^2 N_\ast}{k} \]  \hspace{1cm} (161)
A plot of the quantity \( a_+^2 a_- (a_+ - a_-)^2 \) versus the dimensionless quantity \( \frac{T}{T_C} \) is given in Figure 5. It shows an exponential increase up to about a ratio of \( \frac{T}{T_C} \approx 0.7 \), followed by a sharp decrease that rapidly goes to zero as \( \frac{T}{T_C} \) approaches unity. It is significant that fluctuations do vanish at the Curie temperature since classical thermodynamics predicts that infinite or, at least, very large fluctuations should occur.\(^{15}\) According to J. J. Brophy,\(^\text{16}\) as of 1965 there had been no experimental observations of fluctuations occurring at the Curie temperature of a magnet, although it is not made clear whether none have been attempted or that the measurements yielded a negative result.

However, extensive noise measurements have been made on magnetic material and also on ferroelectric material. In the case of ferroelectrics, the noise goes to a minimum at the Curie temperature.\(^\text{16}\) This provides only a qualified support for our theory because of the difference between the ferroelectric interaction and the magnetic interaction.

**The Barkhausen Effect**

We will now consider correlated pair fluctuations that occur in a simple two state system which is subjected to a constant external magnetic induction. The number of correlated pair fluctuations, as obtained from (151), is

\[
\frac{(\Delta N)^2}{t} = 4 a_+^2 a_-^2 N^2 \mu^2 B^2 \frac{2\pi}{t} |\chi_{m,-}|^2 \int G(\epsilon, \gamma)d\epsilon \gamma,
\]

(162)
Fig. 5. A plot of the quantity $a_+^2 a_-^2 (a_+ - a_-)^2$ versus the ratio of the temperature $T$ to the Curie temperature $T_C$. 
since

\[ B_{z(co)} = B_{z(\tau)} \]

for a constant external induction. Let the system be held in thermodynamic equilibrium. The number of correlated pairs per unit time is then

\[ \overline{\Delta N^2} = \frac{B_z^2}{\left( e^{-\mu B_z \kappa T} + e^{-\mu B_z \kappa T} \right)^2} \Gamma' \]

where

\[ \Gamma' = 4 N_e^2 \mu^2 \frac{2\pi}{\hbar} \left| Y_{(r),+1} \right|^2 \int G_{(r',r)} d^3 \xi \]

(164)

and contains all the terms not dependent on \( B_z \), but which may be weakly dependent on \( T \) because of thermal expansion. In the region of high magnetic induction or low temperature,

\[ e^{-\mu B_z \kappa T} \ll 1 \]

so that

\[ \overline{\Delta N^2} \approx B_z^2 e^{-\mu B_z \kappa T} \Gamma' \]

(165)

From equation (163) and (165) we see that as the magnetic induction increases from zero to some large value, the number of correlated pair fluctuations per unit time first increases approximately as the square of the magnetic induction, reaches a maximum, and then falls to zero in a modified exponential manner.

We are currently unaware of any experimental data with which to directly compare this prediction. However, it is possible to obtain
some indirect experimental verification for this prediction through an examination of the Barkhausen Effect. The Barkhausen Effect describes the small, sudden changes in the magnetic moment which occur in a ferromagnetic substance when it is subjected to a changing external magnetic induction. Usually this phenomena is attributable to the sudden growth of the magnetic domains in the material. We can interpret this growth of domains in terms of the fluctuations of the microscopic magnetic moments. Due to the changing magnetic induction, those microscopic magnetic moment fluctuations that tend to align themselves in the direction of the change outnumber those that go in the opposite direction. This produces a net change in the size and orientation of the macroscopic magnetic domains and thus can account for the Barkhausen Effect.

Because the magnetic induction is changing, the system does not strictly fit the requirement of being in a steady state or equilibrium condition. But if the change occurs slowly compared with the relaxation time of the magnetic processes of the system, we can follow R. S. Tebble et al., and assume that the system is essentially always in equilibrium. In addition, we will assume that the term in the correlated pair fluctuation expression that describes the fluctuation transitions along the direction of the changing magnetic induction predominates over the term that describes transitions in the direction opposite to the changing induction. If we neglect these latter transitions entirely, we obtain for the number of correlated pair
fluctuations along the direction of the changing magnetic induction

\[
\overline{(\Delta N)^2} = \frac{B_x^2}{\left( 2 \cos \frac{\mu B_x}{kT} \right)^\gamma} \frac{\Gamma'}{\gamma} .
\]  

(166)

Since equation (166) describes the number of correlated transitions, the jumps in the magnetic moment of the material must be proportional to it because each of these transitions involves a realignment of a magnetic dipole moment; thus,

\[
\Delta M \propto B_x^2 \left( \cos \frac{\mu B_x}{kT} \right)^\gamma .
\]

(167)

Admittedly, we have made a number of simplifying assumptions to arrive at this expression for the magnetic fluctuations in a material immersed in a slowly changing external magnetic induction. However, we now have a tractable expression for the Barkhausen Effect which can be compared with experiment. The only experiment that we have found with which to make a comparison is by R. M. Bozorth and J. M. Dillinger. They measured the average value of the longitudinal magnetic moment fluctuations in an iron rod as a function of the magnetic field strength. A plot of their results is shown in Figure 6, together with a series of predicted points based on the above expression (167) as indicated by crosses (+).

The magnetic moment fluctuations in this figure are plotted on an arbitrary scale, so we have chosen to scale the theoretical curve to agree with the experimental curve at the point of maximum fluctuations, occurring at a field strength of approximately two oersteds. Taking into account the simplicity of the theoretical model and the
approximations made in arriving at the expression for the magnetic moment fluctuations, we feel that the agreement between the experimental and theoretical curves is surprisingly good.

![Graph of Field Strength vs. Average Change in Magnetic Moment](image)

Fig. 6. Relative sizes of the longitudinal and transverse Barkhausen effect in iron.

One major omission that we have made is that we have assumed that the permeability of the material is constant over the range of magnetic field strengths used in the experiment. A more exact analysis would have to correct for this. In their experiment, Bozorth and Dillinger also measured magnetic fluctuations occurring in a direction transverse to the iron sample. Our simple model does not include this effect.
It is possible to predict the temperature dependence of the location of the point of maximum fluctuations by finding the derivative

\[
\frac{d}{dB_z} \left( \frac{(\Delta N)^2}{t} \right) = \left( 1 - 2 \frac{\mu B_z}{kT} \tanh \frac{\mu B_z}{kT} \right) \frac{\Gamma'}{\left( 2 \cosh \frac{\mu B_z}{kT} \right)^2} \tag{168}
\]

and setting it equal to zero to give

\[
B_z \tanh \frac{\mu B_z}{kT} = \frac{kT}{2\mu} \tag{169}
\]

Hence, the magnetic induction that produces the maximum magnetic fluctuations is related to the temperature in a straightforward way.
CHAPTER 11

CORRELATED FLUCTUATIONS IN THE LIMIT AS $\tau \to 0$

We will next investigate the generalized correlated pair fluctuation formalism in the limiting case as the correlation time $\tau$ approaches zero. By doing this, we will obtain a new insight into a familiar result.

Up to now, we have been dealing with correlated transitions occurring in two separate particles a distance $\chi$ and a time $\tau$ apart. As $\tau \to 0$, the distance $\chi$ separating the two particles also shrinks, until the limit is reached where the two point particles come into physical contact and may, indeed, come to occupy the same volume. In this limit, we must modify our interpretation of $G(\chi, \tau)$, for no longer are we dealing with two particles but, rather, with a single particle. Mathematically, this can be seen by examining $G(\chi, \tau)$ in the limit as $\tau \to 0$:

$$\lim_{\tau \to 0} G(\chi, \tau) = \sum_{\xi,j}^N \delta(\chi + \frac{\bar{\xi} - \bar{\xi}_0}{\xi_j} - \bar{\xi}_0)$$

$$= \sum_{\xi}^N \delta(\chi + \frac{\bar{\xi} - \bar{\xi}_0}{\xi_j} - \bar{\xi}_0) + \sum_{\xi \neq j}^N \delta(\chi + \frac{\bar{\xi} - \bar{\xi}_0}{\xi_j} - \bar{\xi}_0).$$

(170)

Because both coordinate operators occur at the same time, the
correlation integral can be done as follows:

$$\lim_{\tau \to 0} \int G_{(r_2, \tau)} d^3 r = N_0 + \int d^3 r \sum_{j \neq k} \delta_r (r_2 + \vec{r}_{j\to k} - \vec{r}_{j\to 0})$$

$$= N_0 + \int \delta(r_2) d^3 r.$$  \hspace{1cm} (171)

The first term just represents self-correlations of the particles, while the second represents instantaneous correlations of the particles with all the other particles in the system.

With this interpretation of the limiting behavior of $G_{(r_2, \tau)}$ in mind, we will apply the correlated fluctuation formalism to the problem of a beam incident on a collection of target particles. Take, for example, neutrons incident on matter, and assume that the process has been going on long enough to have allowed the system to reach steady state. We need only consider one term of the expression for the correlated fluctuations occurring among the target particles

$$\overline{(\Delta N_{nm})^2} = a_{m\to n} a_{m\to n} N_{m\to n} N_{m\to n} \frac{e^{\frac{-E_{m}}{kT}}}{1} |H_{nm}|^2 \delta(E_{m} - E_{n}) \int G_{(r_2, \tau)} d^3 r.$$  \hspace{1cm} (172)

We have left out the sum on the right side and included subscripts on the left side because we are going to assume that somehow the $m$ to $n$ transition can be detected from among the other allowed transitions. In the usual manner, $|H_{nm}|^2$ represents the square of the matrix element of the interaction producing the transitions, and $a_{m}$ the probability of finding a particle in the initial state $m$. Further, let the final state $n$ be a highly excited state not usually allowed to the
system particles unless they have undergone the scattering interaction with the incident projectiles, and let the intensity of these projectiles be low. Therefore, \( N_{nm} \), which represents the number density of final state particles, is a small number. In many cases, the number density of particles in the final states can be most easily measured by observing some external manifestation that an interaction has taken place, such as a deflection as occurs in scattering.

Now we consider the limiting case and examine the effect on \( \langle \Delta N_{nm} \rangle^2 \). For the simultaneous correlation of one particle with other particles in the system, such as in the case we have been considering up to now, we have

\[
\lim_{\tau \to 0} a_{m(\tau)} a_{m(\tau)} = a_{m(\tau)}^2
\]

However, for the self-correlation term, we are considering only single particles and we need only evaluate the probability of finding it initially in state \( m \) once. Therefore, we must replace (173) in this case by

\[
\lim_{\tau \to 0} a_{m(\tau)} a_{m(\tau)} \rightarrow a_{m(\tau)}
\]

Further, for single collisions, only one target particle will be in the final state, so that \( N_{nm(\tau)} = 1 \). Finally we obtain

\[
\langle \Delta N_{nm} \rangle^2 = a_{m(\tau)} \frac{2\pi}{\hbar} |H_{nm}|^2 \delta_{\epsilon_m - \epsilon_n - \epsilon_\tau} \left( N_0 + a_{m(\tau)} \int \frac{g(\xi)}{\epsilon_\tau} d^3 \epsilon \right) . \tag{175}
\]

This result expresses the correlated fluctuation in number density, for correlation time \( \tau < \sigma \), as the sum of two terms. The first is a term
that evaluates the fluctuations in a single particle, and is just the transition probability times the total number of particles in the initial state. The second term gives the probability per unit time of seeing a simultaneous fluctuation of two particles by weighting the single particle transition probability with the conditional probability of finding two particles within the interaction range.

The result also implies that what are observed as lifetimes and cross sections are, in reality, fluctuations or stimulated fluctuations. Actually, the possibility of interpreting scattering phenomena as a manifestation of a fluctuation is not new. Van Hove has derived, from the Born Approximation, an expression for the differential cross section which is essentially identical to that obtained by appropriately modifying the fluctuation expression \( \frac{(\Delta N_{nm})^3}{t} \) for \( \gamma = 0 \). The important feature is the presence of the term \( \mathcal{G}(\zeta) \). Van Hove and Münster show how this term appears in neutron scattering experiments, the Zernike-Prins formula for the scattering of X rays on simple liquids and in a number of other applications, none of which will be repeated here. We will only add our own understanding of the role of \( \mathcal{G}(\zeta) \) as the conditional probability of simultaneously finding two separate particles located a distance \( \zeta \) apart. This is perhaps easiest to visualize in the case of photons, where the instantaneous photon number density about a particular particle may extend a distance \( R_{\zeta} \) beyond that particle and has the probability \( \int_{0}^{R_{\zeta}} \mathcal{G}(\zeta) d^3\zeta \) of including a second particle. There then exists the probability of simultaneously stimulating both particles.
CHAPTER 12

THE RELATION OF FLUCTUATIONS TO ENTROPY AND POTENTIAL ENERGY

We will now briefly discuss the relationship between fluctuations and entropy. We begin with the definition of entropy \( S = k \ln W \) \( (176) \),

where \( k \) is the Boltzmann constant, \( W \) is the volume of phase space available to the system

\[ W = \frac{\Delta P \Delta q}{\hbar}, \] \( (177) \)

and \( S \) is the number of degrees of freedom of the system.

From the previous discussions, as illustrated specifically by (120), we see that the correlated number fluctuations are, among other things, proportional to the volume in phase space,

\[ (\Delta N)^2 \propto W. \] \( (178) \)

This leads to the familiar relation

\[ (\Delta N)^2 \propto e^{\frac{S}{k}}. \] \( (179) \)

However, there is more to the correlated number fluctuations than just phase space. The fluctuations are dependent upon such things as the probability that the transition can occur, and the probability of finding other particles with which to form a fluctuation. This is important because although a system may have a large amount of phase space available to it, the fluctuations can be inhibited due to a small
probability that the fluctuation producing interactions can occur. The entropy of such a system, while potentially large, can be held to a much smaller value if it is initially prepared in a highly organized, or otherwise unlikely, state. The fluctuations that cause the system to move from an unlikely state of low entropy toward the more random state of maximum entropy either do not occur, or occur at such a low rate as to effectively preclude the state of maximum entropy from being attained.

We next discuss the two terms in the generalized correlated pair fluctuation expression that we have up to now chosen to ignore since they did not contribute an observable effect. We recall from the discussion of Chapter 5 that these terms constitute a continuous interchange of energy between the particles in the system. We will try to give more meaning to these terms, and show that for a simple case they can lead to a \( (r)^{-1} \) form of the potential energy between two particles. In this case, these terms give rise to an observable effect, but only if the physical configuration of the system is changed by varying the distances separating the particles.

We begin by considering a simple system composed of only two particles which somehow interact, the interaction being directed along the straight line connecting the two particles. The interaction is carried out by some form of quantum which emerges from one of the particles, goes out to the second particle where it is absorbed, and then is reemitted back to the first particle, whereupon the interaction is completed. This process occurs continuously and, as can be seen from
the fluctuation expression, it conserves energy. We are mainly interested in the pair correlation behavior and not in the details of the interaction, so we will limit ourselves to discussing the pair correlation function $G(r, r)$. This we will postulate to have the following form

$$G(r, r) = \frac{\sigma^2}{\gamma \pi r^2}$$

(180)

based on the simple view that if particle number 1 emits a virtual quantum, the probability of this virtual quantum being seen by particle 2, located a distance $r$ away from particle 1, is just the ratio of the cross section of particle 2 to the area of a sphere of radius $r$. The correlation volume element for this process is just

$$d^3 \mathbf{r} = \sigma_i \, dr$$

(181)

so that the correlated number fluctuations become

$$\langle \Delta N \rangle^2 = -2 \sum_{m, n} a_{m(i)} a_{n(j)} N_n N_m \frac{2\pi}{\hbar} \left| H_{nm} \right|^2 \int S \left( E_{m(i)} - E_{n(j)} \right) \frac{\sigma^2}{\gamma \pi r^2} \, dr$$

(182)

where we have lumped the two oscillating terms into one. The first part of the expression simply describes the probability of a quantum being emitted. The integral over coordinate space gives the probability of the virtual quantum emitted from one particle finding the second particle.

If we now suppose that these quanta carry some energy, then at any one time some of this energy is in "flight" between the particles, and can be interpreted as representing the interaction potential
energy. The amount of this energy that effectively resides in the space between the particles is proportional to the number of the correlated pairs of quanta fluctuating between the two particles. The integral is elementary and in this way leads to a potential energy expression that depends inversely on the particle separation, a form which is often found in nature.
CHAPTER 13

CONCLUDING REMARKS

Starting from a definition of an off diagonal statistical operator, we have been able to develop a generalized theory of correlated fluctuations for the equilibrium and steady states. In the process, a set of rate equations was obtained which are invariant under time reversal and lead immediately to the results of Boltzmann and Planck. The generalized expression for correlated fluctuations was derived from these rate equations and shown to predict results which agree very well with known experimental observations in a wide variety of applications. In addition, results have been predicted for which there are no observational data, thus suggesting possible new experiments. More importantly, few of these subjects previously had adequate theoretical backing. We think that the generalized theory developed here more adequately describes these phenomena.

A subject of interest not included in this dissertation is the application of the set of rate equations to the description of steady state phenomena other than fluctuations. These equations, as exemplified by (26), involve the interaction matrix elements in a linear fashion. This is a significant departure from the usual rate equations for probabilities which are usually applied in this situation and which involve the matrix elements squared in the form of transition probabilities. The linear nature of our rate equations is due to the Liouville
equation. The fact that our equations differ from conventional theory is perhaps encouraging because considerable confusion presently exists in the theory of the steady state. It is hoped that it will soon be possible to compare the predictions of the linear rate equations with experiment.
REFERENCES


