THE EXPANSION OF
THE MASTER EQUATION

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I. INTRODUCTION

Macroscopic physics deals with macroscopic quantities $q_v$, such as the positions and velocities of bodies, electrical charges and currents, amounts or concentrations of chemical compounds, temperatures at various points in a material, or local density and velocity of a fluid. They obey macroscopic laws

$$\dot{q}_v = F_v(q_1, q_2, \ldots)$$

(1.1)
which on a macroscopic level can be derived from general principles such as conservation laws, together with some specific assumptions of phenomenological nature, for example, those of Fourier and Fick. On the one hand this macroscopic picture is incomplete, because it has to introduce phenomenological coefficients, as many as there are phenomena, but this is of no concern to us here. On the other hand the macroscopic laws (1.1) are merely an approximation, valid when so many particles are involved that fluctuations are negligible. The present work is concerned with improving on this macroscopic approximation by taking into account the fluctuations. Of course the quantities that characterize the discreteness are essential: Boltzmann's constant, the elementary charge, and the masses of individual particles.

In order to study the corrections to (1.1) caused by the discrete nature of matter one must view the macroscopic phenomena as the outcome of the collective behavior of many particles. This does not merely require an investigation of the phenomenological assumptions mentioned above, but a reappraisal of the very definitions of the $q_i$ is needed.

In principle all information is contained in the microscopic equations of motion of all particles, but it hardly needs saying that an exact solution of these equations is beyond human means, excepting a small number of simple models. Even the macroscopic laws (1.1) can only be derived from them with the help of simplifications and assumptions, which are no more reliable than the phenomenological assumptions used in the purely macroscopic approach. It is therefore sensible to embark upon a less ambitious program and to develop a theory which goes beyond the macroscopic description in that it includes fluctuations, but short-cuts the connection with the microscopic equations by an appeal to some suitably chosen semiphenomenological assumptions. This is the customary approach in noise theory; we propose to call it the mesoscopic level of description.

One popular mesoscopic approach consists in adding to (1.1) a fluctuating term

\[ \dot{q}_i = F_i(q_1, q_2, \ldots) + \lambda_i(t) \]  

(1.2)

and making suitable assumptions concerning the statistical properties of the random functions $\lambda_i(t)$ (see Section XI). It should be clear that this device changes the nature of the $q_i$; they are now also stochastic quantities. The macroscopic values that enter into (1.1) are identified with the averages of the $q_i$ in (1.2). This approach was first used by Langevin in his treatment of the Brownian movement, and his success has led many authors to apply the same device to other systems. However, we shall show in Section XI that in many cases it leads to wrong results.

A second approach starts out by introducing the probability distribution $P(q_1, q_2, \ldots; t)$ defined as follows: $P(q_1, q_2, \ldots; t) dq_1 dq_2 \ldots = the joint
probability that at time $t$ the first quantity has a value between $q_1$ and $q_1 + dq_1$, and the second one between $q_2$ and $q_2 + dq_2$, etc. Note that the $q$'s have changed their nature again and are merely coordinates in some $q$-space on which the probability density $P$ is defined. One then assumes that $P$ obeys the Fokker-Planck equation

$$\frac{\partial P}{\partial t} = -\sum_y \frac{\partial}{\partial q_y} F_y(q) P + \sum_{y\mu} \frac{\partial^2}{\partial q_y \partial q_\mu} D_{y\mu} P$$

(1.3)

The $F_y$ are the same as in (1.1), and the new coefficients $D_{y\mu}$ are found from the fluctuation-dissipation theorem. Although this looks quite different from the Langevin approach, it is actually equivalent to it, and therefore subject to the same criticism (Section XXII).

A third mesoscopic approach is the basis of this article. It also starts out from the probability density $P$, but merely assumes that it obeys an equation of type

$$\dot{P} = WP$$

(1.4)

where $W$ is a linear operator acting on the $q$-dependence. Let $W(q|q')$ be the integral kernel of $W$; then the requirement that the total probability must remain equal to unity tells

$$\int W(q|q') \, dq = 0$$

(Each $q$ stands for the whole set of $q_y$, and $dq$ is a volume element in $q$-space.) Hence one may write (1.4) in the physically more transparent form

$$\dot{P}(q, t) = \int \{ W(q|q') P(q', t) - W(q'|q) P(q, t) \} \, dq'$$

(1.5)

The kernel $W(q|q')$ for $q \neq q'$ represents the transition probability per unit time from $q'$ to $q$ and must be nonnegative. The second term represents the decrease of $P(q, t)$ due to transitions to other values $q'$. Equation (1.4) or (1.5) is called the “master equation”.*

The assumption (1.4) implies that the stochastic process described by $q$ is a Markov process. This is a strong assumption, which in most applications is only approximately true and with the conditions that a suitable coarse-grained time scale is used, and that the correct set of variables $q = \{q_1, q_2, \ldots\}$ is chosen. On the other hand, it is weaker than the assumptions needed in the two previously mentioned mesoscopic approaches. Moreover it is easier to assess on physical grounds. The transition probabilities $W(q|q')$ usually have a direct physical interpretation in terms of the microscopic quantities

* Throughout this article the term is used in its original sense: an equation of the type (1.4) for a probability distribution.
Fig. 1. The evolution of the probability density towards equilibrium.

Figuring the system, for instance, collision cross-sections or quantum mechanical matrix elements. We shall meet cases where the master equation (1.5) obviously holds, but neither the Langevin approach, nor the Fokker-Planck equation (1.3) leads to correct results.

The master equation purports to describe the entire behavior of the $q$ and hence also the macroscopic equation (1.1) should follow from it. How is it possible for an equation governing the probability density in $q$-space to give rise to a deterministic set of equations for the $q_v$? The idea is that $P(q, t)$ is a sharp peak located at a rather well-defined point in $q$-space (Fig. 1). If the width of the peak may be neglected it is possible to consider its position in $q$-space as the macroscopic value of $q_v$. While $P$ varies in time according to (1.5) the peak moves through $q$-space according to (1.1). Note that there is no contradiction between the fact that (1.5) is linear whereas (1.1) may well be nonlinear. The situation is analogous to the way in which a linear Schroedinger equation gives rise to a nonlinear classical equation of motion in the approximation in which the particle is heavy enough to neglect the spreading of the wave function.

The mathematical scheme describing this state of affairs was developed some time ago. The present review is confined to the special but frequently occurring class of master equations in which the variable $q$ takes only integral values. We therefore write $n$ rather than $q$ and the master equation is

$$\dot{P}(n, t) = \sum_{n'} \{ W(n | n')P(n', t) - W(n' | n)P(n, t) \}$$

It may happen that $n$ runs from $-\infty$ to $+\infty$, or from 0 to $\infty$, or only takes values in some finite range. The transition probabilities $W(n | n')$ need only be defined for $n \neq n'$ and are nonnegative. They are properties of the system and, of course, independent of the $P(n, t)$, which describe the special mesoscopic state considered. It is possible to include the case that $W$ depends on time (nonautonomous systems), but we shall not do so. Equation (1.6) may also be written by means of a matrix $W$

$$\dot{P}(n, t) = \sum_{n'} W_{nn'}P(n', t)$$
The master equation is "solved" if one can find the \( P(n, t) \) that obey (1.6) and take arbitrarily prescribed initials values at \( t = 0 \). Obviously it suffices to consider the initial condition
\[
P(n, 0) = \delta_{n,m}
\] (1.7)
for each \( m \). The corresponding solution is
\[
P(n, t | m, 0) = (e^{\mathbf{W}})_{nm}
\] (1.8)
In order to evaluate this formal expression one has to diagonalize \( \mathbf{W} \), but only in rare cases can that be done exactly. Hence it is necessary to have a systematic approximation scheme in the form of a power series expansion in some physical parameter. It appeared that the appropriate quantity is \( \Omega^{-1/2} \), where \( \Omega \) is a measure for the size of the system or the total number of particles involved. This scheme is demonstrated on a simple example in Section III, formulated in general in Sections IV and V, and subsequently applied to various problems.

It will appear that most of the problems treated in the literature can be readily handled with the \( \Omega \)-expansion method. Many of the existing controversies and paradoxes\(^5\) are caused by unsystematic approximations, in which terms are neglected according to the taste of the author. In addition it will be shown that the popular Langevin approach may lead to wrong results even in simple cases (Section XI), and the limitations of the Fokker–Planck equation are discussed in Section XXII. On the other hand, it must be stressed that the expansion is essentially based on the smallness of fluctuations and has only limited validity in unstable situations (Section XVIII) or phase transitions (Section XX).

### II. PRELIMINARIES

The jump moments or derivate moments\(^8\) are defined by
\[
a_p(n) = \sum_{n'} (n' - n)^p W(n'|n) \quad (p = 1, 2, \ldots)
\] (2.1)
Multiply (1.6) with \( n \) and sum
\[
\frac{d}{dt} \langle n \rangle = \sum_{nn'} \{nW(n|n')P(n') - nW(n'|n)P(n)\}
\]
\[
= \sum_{nn'} (n' - n)W(n'|n)P(n)
\]
\[
= \langle a_1(n) \rangle
\] (2.2)
If $a_1(n)$ is a linear function this is identical with

$$\frac{d}{dt} \langle n \rangle = a_1(\langle n \rangle)$$

(2.3)

which permits us to determine $\langle n \rangle$ as a function of $t$. If, however, $a_1(n)$ is not linear, (2.3) is at best an approximation, which amounts to neglecting all fluctuations. We shall see that (2.3) is, indeed, the zeroth approximation in the $\Omega$-expansion scheme, and is therefore to be identified with the macroscopic equation (1.1).* The exact identity (2.2) is not a closed equation for $\langle n \rangle$ but involves higher moments of $n$ as well. To improve on the approximation (2.3) we expand in (2.2) the function $a_1(n)$ in $n - \langle n \rangle$ and break off after the second derivative:

$$\frac{d}{dt} \langle n \rangle = a_1(\langle n \rangle) + \frac{1}{2}a_1''(\langle n \rangle)\sigma_n^2$$

(2.4)

where $\sigma_n^2 = \langle (n - \langle n \rangle)^2 \rangle$. As this equation involves $\langle n^2 \rangle$ we also multiply (1.6) with $n^2$ and sum

$$\frac{d}{dt} \langle n^2 \rangle = \sum_{nn'} (n^2 - n^2)W(n'|n)P(n)$$

$$= \langle a_2(n) \rangle + 2\langle na_1(n) \rangle$$

(2.5)

Combination with (2.2) yields the exact identity

$$\frac{d}{dt} \sigma_n^2 = \langle a_2(n) \rangle + 2\{\langle na_1(n) \rangle - \langle n \rangle \langle a_1(n) \rangle \}$$

(2.6)

Making somewhat loose approximations similar to (2.3) we write for this

$$\frac{d}{dt} \sigma_n^2 = a_2(\langle n \rangle) + 2a_1'(\langle n \rangle)\sigma_n^2$$

(2.7)

In Section V it will be shown that the pair of equations (2.4) and (2.7) together actually constitute a consistent approximation.\textsuperscript{9,10}

The upshot is that in order to improve on (2.3) two coupled equations [(2.4) and (2.7)] are needed (unless $a_1(n)$ happens to be linear). That means that it is no longer possible to determine $\langle n \rangle$ from its initial value; one also needs to know the initial value of $\sigma_n^2$. All this is subject to the condition that $\sigma_n^2$ remains finite (of order $n$), otherwise there is no justification for omitting higher moments. This condition amounts to $a_1'(\langle n \rangle) < 0$, that is, the system must be stable (compare Section V).

* This statement requires a minor modification, see Section IV.
A special but important class of discrete Markov processes are the one-step or birth-and-death processes. They are defined by $W(n|n') = 0$ unless $n = n' \pm 1$, that is,

$$W(n|n') = r(n')\delta_{n,n'-1} + g(n')\delta_{n,n'+1}$$

$r(n)$ and $g(n)$ may be any two nonnegative functions, usually analytic; their names stem from recombination and generation of charge carriers in semiconductors.$^{11}$

The master equation of a one-step process has the following form:

$$\dot{P}(n, t) = r(n + 1)P(n + 1, t) + g(n - 1)P(n - 1, t) - [r(n) + g(n)]P(n, t)$$

It is convenient to define the difference operator $E$ by$^{12}$

$$Ef(n) = f(n + 1), \quad E^{-1}f(n) = f(n - 1)$$

With its aid the master equation (2.8) may be written

$$\dot{P} = (E - 1)r(n)P + (E^{-1} - 1)g(n)P$$

The jump moments are

$$a_\rho(n) = (-1)^\rho r(n) + g(n)$$

The macroscopic rate equation (2.3) takes the form

$$\frac{d}{dt} \langle n \rangle = -r\langle n \rangle + g\langle n \rangle$$

and the coupled equations [(2.4) and (2.7)] are

$$\frac{d}{dt} \langle n \rangle = g\langle n \rangle - r\langle n \rangle + \frac{1}{2}[g'\langle n \rangle - r'\langle n \rangle]\sigma_n^2$$

$$\frac{d}{dt} \sigma_n^2 = g\langle n \rangle + r\langle n \rangle + 2[g'\langle n \rangle - r'\langle n \rangle]\sigma_n^2$$

Some general properties of one-step processes are listed in Section VI.

**III. FIRST EXAMPLE: SPREADING OF AN EPIDEMIC**

As a first example for demonstrating the expansion we choose a simple nonlinear one-step process, which describes the spreading of an epidemic in a population of $\Omega$ individuals.$^{6,13}$ If $n$ is the number of infected individuals, the probability per unit time for a new infection to occur is proportional to $n$, and to the number $\Omega - n$ of uninfected. Thus $g(n) = \beta n (\Omega - n)$ with constant
Furthermore we take \( r(n) = 0 \), that is, no cure is possible. Hence

\[
W(n|n') = \beta \delta_{n,n'+1} n' (\Omega - n')
\]  

(3.1)

The master equation is

\[
\dot{P}(n,t) = \beta(n - 1)(\Omega - n + 1)P(n + 1,t) - \beta n(\Omega - n)P(n,t)
\]  

\[= \beta \mathbf{E}^{-1} - 1) n(\Omega - n)P
\]  

(3.2)

The more general problem with arbitrary \( g(n) \) and \( r(n) = 0 \) has been treated by Weiss as a model for superradiance.\(^{14}\) Actually such problems can be solved without approximations in a more or less closed form, but the result is too involved to be of much use, unless \( g(n) \) is sufficiently simple.

One expects \( n \) to consist of a macroscopic part \( \Omega \phi(t) \) plus fluctuations of order \( \Omega^{1/2} \). That is, \( P(n,t) \) will be a sharp peak located roughly at \( \Omega \phi(t) \) with a width of order \( \Omega^{1/2} \) (see Fig. 2). Hence we set

\[
n = \Omega \phi(t) + \Omega^{1/2} x
\]  

(3.3)

where \( x \) is the new variable and \( \phi(t) \) will be chosen presently. We shall call \( \Omega \phi(t) \) the "macroscopic part" and \( \Omega^{1/2} x \) the "fluctuating part" of \( n \), and refer to the new variables as the "\( \Omega \) language." Accordingly the probability distribution of \( n \) now becomes a probability distribution \( \Pi \) of \( x \),

\[
P(n,t)\Delta n = \Pi(x,t)\Delta x
\]  

(3.4)

The following transformation formulas apply

\[
\frac{\partial \Pi}{\partial x} = \Omega^{1/2} \frac{\partial P}{\partial n}, \quad \frac{\partial \Pi}{\partial t} = \Omega^{1/2} \left\{ \Omega \frac{\partial \phi}{\partial t} \frac{\partial P}{\partial n} + \frac{\partial P}{\partial t} \right\}
\]
Hence*

\[ \Omega^{1/2} \frac{\partial P}{\partial t} = \frac{\partial \Pi}{\partial t} - \Omega^{1/2} \frac{d\varphi}{dt} \frac{\partial \Pi}{\partial x} \]  

(3.5)

Moreover one has

\[ \mathbf{E} = 1 + \Omega^{-1/2} \frac{\partial}{\partial x} + \frac{1}{2} \Omega^{-1} \frac{\partial^2}{\partial x^2} + \ldots \]  

(3.6)

Substitute the new variables in the master equation (3.2) and cancel an overall factor \( \Omega^{-1/2} \),

\[ \frac{\partial \Pi}{\partial t} - \Omega^{1/2} \frac{d\varphi}{dt} \frac{\partial \Pi}{\partial x} = \beta \Omega^2 \left\{ -\Omega^{-1/2} \frac{\partial}{\partial x} + \frac{1}{2} \Omega^{-1} \frac{\partial^2}{\partial x^2} \right\} \times (\varphi + \Omega^{-1/2} x)(1 - \varphi - \Omega^{-1/2} x) \Pi \]  

(3.7)

We absorb one factor \( \Omega \) into the time variable (and for convenience also the \( \beta \)) by setting

\[ \beta \Omega t = \tau \]  

(3.8)

Then the largest terms are

\[ -\Omega^{1/2} \frac{d\varphi}{d\tau} \frac{\partial \Pi}{\partial x} = -\varphi(1 - \varphi) \frac{\partial \Pi}{\partial x} \]  

They can be made to cancel by subjecting \( \varphi \) to the equation

\[ \frac{d\varphi}{d\tau} = \varphi(1 - \varphi) \]  

(3.9)

This equation determines how the macroscopic part of \( n \) varies with time. Translating back to the original variables it takes the form

\[ \frac{dn}{dt} = \beta n(\Omega - n) \]  

(3.10)

which appears to be identical with the macroscopic rate equation (2.12).

The terms of order \( \Omega^0 \) in (3.7) yield an equation for \( \Pi \),

\[ \frac{\partial \Pi}{d\tau} = - (1 - 2\varphi) \frac{\partial}{\partial x} x \Pi + \frac{1}{2} \varphi(1 - \varphi) \frac{\partial^2 \Pi}{\partial x^2} \]  

(3.11)

This is a Fokker–Planck equation whose coefficients involve \( \varphi \) and therefore depend on time. Observe, however, that the coefficient of the first term is

* It is possible to arrive at (3.5) without the intervention of the dubious symbol \( \partial P/\partial n \). Let \( t \) in (3.4) vary by \( \delta t \) and simultaneously \( x \) by \(- \Omega^{1/2} \varphi(t) \delta t \); this leads immediately to (3.5)
linear in $x$, and that the second term does not depend on $x$; we shall indicate these features of a Fokker–Planck equation by calling it linear. (Of course, all Fokker–Planck equations are linear in the unknown function—in this case $\Pi$.) Equation (3.11) governs the fluctuations in $n$ of order $\Omega^{1/2}$ about the macroscopic part.

The strategy for solving the master equation (3.2) with initial condition (1.7) now emerges. First solve (3.9) with initial value $\phi(0) = \frac{m}{\Omega}$. Then solve (3.11) with initial $\Pi(x, 0) = \delta(x)$. Then

$$P(n, t|m, 0) = \Omega^{-1/2} \Pi \left( \frac{n - \Omega \phi(\tau)}{\Omega^{1/2}}, \tau \right)$$

In this solution terms of relative order $\Omega^{-1/2}$ have been neglected.

**IV. THE GENERAL EXPANSION METHOD**

The basic idea is that there is a parameter $\Omega$ measuring the size of the system, such that for large $\Omega$ the fluctuations are relatively small. It is then possible to expand in descending powers of $\Omega$, as will be outlined in five steps.*

*First step*: specifying the dependence of the transition probabilities on $\Omega$. It is assumed that the way in which $W(n|n')$ depends on $\Omega$ has the following form:

$$W(n|n') = f(\Omega) \left[ \Phi_0 \left( \frac{n'}{\Omega}; n - n' \right) + \Omega^{-1} \Phi_1 \left( \frac{n'}{\Omega}; n - n' \right) + \cdots \right]$$ (4.1)

Each function $\Phi_j$ has a Taylor expansion with respect to its first argument, but is of course a discrete function of its second argument, which is the jump size. The factor $f(\Omega)$, usually some power of $\Omega$, is innocuous because it can be absorbed in the time variable. The jump moments (2.1) are transformed accordingly,

$$a_p(n) = f(\Omega) \alpha_p \left( \frac{n}{\Omega} \right)$$ (4.2)

In the following we suppose for simplicity that $\Phi_1, \Phi_2, \ldots$ vanish. They are not hard to include when they occur, as in Section IX, but cumbersome in the general treatment. When they do not vanish it is not strictly true that (2.3) is identical with the macroscopic law, inasmuch as $a_1$ involves higher orders in $1/\Omega$, which do not belong to a macroscopic description. The macroscopic law is determined by the first jump moment of $\Phi_0$ alone, but in the next approximation (2.4) both $\Phi_0$ and $\Phi_1$ have to be used for $a_1$.

* Previously we have used the Kramers–Moyal expansion as a convenient intermediate step, but we shall avoid it here, since its role has been misconstrued.
Substituting (4.1) in (1.6) and changing the summation variable from \( n' \) to \( v = n - n' \) one obtains for the master equation

\[
\frac{\partial P(n, t)}{\partial t} = f(\Omega) \sum_v \left\{ \Phi_0\left(\frac{n - v}{\Omega}; v\right)P(n - v, t) - \Phi_0\left(\frac{n}{\Omega}; v\right)P(n, t) \right\} \quad (4.3)
\]

Second step: postulating the way in which \( P \) depends on \( \Omega \). One expects \( P(n, t) \) to be a sharp peak located at some point \( \Omega \phi(t) \) with a width of order \( \Omega^{1/2} \). Hence one transforms the variable \( n \) to a new variable \( x \) as in (3.3). This transforms \( P(n, t) \) into \( \Pi(x, t) \) according to (3.4). Substitute this in the master equation:

\[
\frac{\partial \Pi}{\partial t} - \Omega^{1/2} \frac{d\phi}{dt} \frac{\partial \Pi}{\partial x} = f(\Omega) \left[ \sum_v \Phi_0(\phi(t) + \Omega^{-1/2}x - \Omega^{-1/2}v; v) \right.
\]

\[
\times \Pi(x - \Omega^{-1/2}v, t) - \sum_v \Phi_0(\phi(t) + \Omega^{-1/2}x; -v)\Pi(x, t) \right]
\]

(4.4)

The factor \( [\ ] \) vanishes to lowest order in \( \Omega^{-1/2} \). To obtain the next order it is convenient to write it in the form

\[
[\ ] = \sum_v \left\{ -\Omega^{-1/2}v \frac{\partial}{\partial x} + \frac{1}{2} \Omega^{-1}v^2 \frac{\partial^2}{\partial x^2} - \cdots \right\} \Phi_0(\phi + \Omega^{-1/2}x; v)\Pi(x, t)
\]

(4.5)

Third step: extracting the largest terms to obtain the macroscopic equation. The lowest order in \( [\ ] \) is \( \Omega^{-1/2} \); it can be combined with the term of order \( \Omega^{1/2} \) on the left if we define a scaled time \( \tau \) by*

\[
f(\Omega)\tau = \Omega \tau \quad (4.6)
\]

Then the largest terms are of order \( \Omega^{1/2} \) on both sides,

\[
-\Omega^{1/2} \frac{d\phi}{d\tau} \frac{\partial \Pi}{\partial x} = \Omega \left( -\Omega^{-1/2} \frac{\partial \Pi}{\partial x} \right) \sum_v v\Phi_0(\phi; v)
\]

Since both terms involve \( \Pi \) only through the factor \( \partial \Pi/\partial x \), it is possible to satisfy this equation by choosing for \( \phi \) a solution of

\[
\frac{d\phi}{d\tau} = \sum_v v\Phi_0(\phi, v) = \mu_1(\phi) \quad (4.7)
\]

* In many of our examples we shall find \( f(\Omega) = \Omega \) so that \( t = \tau \).
This is the equation for the macroscopic part of \( n \), that is, the macroscopic rate equation.

**Fourth step:** the next order determines the fluctuations. The terms of order \( \Omega^0 \) in (4.4) are

\[
\frac{\partial \Pi}{\partial \tau} = - \left\{ \sum_v v \Phi'_0(\varphi; \nu) \right\} \frac{\partial}{\partial x} x \Pi + \frac{1}{2} \left\{ \sum_v v^2 \Phi_0(\varphi; \nu) \right\} \frac{\partial^2 \Pi}{\partial x^2}
\]

\[
= - \alpha'_1(\varphi) \frac{\partial}{\partial x} x \Pi + \frac{1}{2} \alpha_2(\varphi) \frac{\partial^2 \Pi}{\partial x^2}
\]  

(4.8)

The prime indicates differentiation with respect to \( \varphi \). This is again a linear Fokker–Planck equation with time-dependent coefficients, which governs the fluctuations in \( n \) of order \( \Omega^{1/2} \) about the macroscopic part \( \Omega \).

**Final step:** collecting the results in order to solve (1.6) with initial condition (1.7). First solve (4.7) with initial condition

\[
\Omega \varphi(0) = m
\]

(4.9)

and call the solution \( \varphi(\tau | m/\Omega) \). Next solve (4.8) with initial condition

\[
\Pi(x, 0) = \delta(x)
\]

(4.10)

and call the solution \( \Pi(x, \tau | 0, 0) \). Then

\[
P(n, t | m, 0) = \Omega^{-1/2} \Pi \left( \frac{n - \Omega \varphi(\tau | m/\Omega)}{\Omega^{1/2}}, \tau \bigg| 0, 0 \right)
\]

(4.11)

Note that one has to the same order

\[
P(n, t | m, 0) = \Omega^{-1/2} \Pi \left( \frac{n - \Omega \varphi(\tau | m/\Omega) - c \Omega^{1/2}}{\Omega^{1/2}}, \tau \bigg| c, 0 \right)
\]

(4.12)

where \( c \) is an arbitrary number of order 1.

This program can be carried out by a number of integrations (see Appendix). It is simpler, however, and in many cases sufficient to determine only \( \langle n \rangle \) and \( \sigma_n^2 = \langle n^2 \rangle - \langle n \rangle^2 \) as functions of \( t \). The relevant formulas are derived in the next section.

Higher orders can be added and have the effect of modifying the equation for \( \Pi(x, t) \) (see Section VIII). However, we shall be mainly concerned with the approximation to order \( \Omega^0 \) as given here. This will be called the **linear noise approximation** since to this order the fluctuations are governed by the linear Fokker–Planck equation (4.8). It is the approximation on which the familiar theory of noise in electrical networks\(^{11} \) is based.
V. THE EQUATIONS FOR THE MOMENTS

Without actually solving (4.8) one may deduce directly from it (by multiplying with \( x \) and integrating)

\[
\frac{d}{d\tau} \langle x \rangle = \alpha_1(\varphi)\langle x \rangle + \alpha_2(\varphi)
\]  

(5.1)

Observe that this is identical with the "variational equation" belonging to (4.7), that is, the equation for the difference between two neighboring solutions of (4.7). The fact that this must be so can be gleaned from (4.12); a slight variation (of order \( \Omega^{-1/2} \)) in the initial value of \( \varphi \) can be compensated by the initial value of \( \langle x \rangle \). An important consequence is the following: Since the variational equation of (4.7) determines the stability of the macroscopic solution \( \varphi(t) \), it follows that the macroscopic stability also determines whether or not the average \( \langle x \rangle \) of the fluctuations grows with time.

One also deduces directly from (4.8)

\[
\frac{d}{d\tau} \langle x^2 \rangle = 2\alpha_1(\varphi)\langle x^2 \rangle + \alpha_2(\varphi)
\]  

(5.2)

In both (5.1) and (5.2) terms of order \( \Omega^{-1/2} \) have been neglected. With the choice of (4.9) or (4.10) for the initial values one has at \( t = 0 \)

\[
\langle x \rangle_0 = 0, \quad \langle x^2 \rangle_0 = 0
\]  

(5.3)

Hence \( \langle x \rangle \) remains zero at all \( t > 0 \), so that

\[
\langle n \rangle_t = \Omega\varphi(t) + O(1)
\]  

(5.4)

To the present order, therefore, the macroscopic part of \( n \) is also its average.

Furthermore, if the macroscopic solution \( \varphi(t) \) is stable, and therefore also (5.1), it follows from (5.2) that \( \langle x^2 \rangle \) remains finite as well. Consequently \( x \) remains of order unity at all times, which constitutes the \textit{a posteriori} justification of the Ansatz (3.3). Note that the stability is crucial for our approximation scheme: if \( \langle x^2 \rangle \) grows exponentially in time, the separation of powers of \( \Omega \) becomes invalid after a time of order long \( \Omega \). In Section XVIII we shall meet an example where \( \langle x^2 \rangle \) grows linearly with time.

It is possible to improve the equation for \( \langle n \rangle \) by one order without going beyond the linear noise approximation. To this end we rewrite the exact equation (2.2) in the \( \Omega \) language in order to display the powers of \( \Omega \),

\[
\frac{d}{d\tau} (\varphi + \Omega^{-1/2}\langle x \rangle) = \langle x_1(\varphi + \Omega^{-1/2}x) \rangle
\]

\[
= \alpha_1(\varphi) + \Omega^{-1/2}\alpha_1(\varphi)\langle x \rangle + \frac{1}{2}\Omega^{-1}\alpha_1(\varphi)\langle x^2 \rangle + O(\Omega^{-3/2})
\]
Since $\varphi$ obeys (4.7) by definition,

$$
\frac{d}{dt} \langle x \rangle = \alpha_1(\varphi)\langle x \rangle + \frac{1}{2}\Omega^{-1/2}\alpha_1(\varphi)\langle x^2 \rangle + \mathcal{O}(\Omega^{-1})
$$

(5.5)

Although this equation for $\langle x \rangle$ involves $\langle x^2 \rangle$ there is a factor $\Omega^{-1/2}$; hence the approximation (5.2) for $\langle x^2 \rangle$ suffices. Rewriting the result in the original variables we see that the two coupled equations (2.4) and (2.7) determine $\langle n \rangle$ to order $\Omega^0$ and $\sigma_n^2$ to order $\Omega^1$.

The conclusion reached here in a slightly devious manner can also be obtained by simply adding the next order correction to (4.8) and then computing the first and second moments to the desired order (compare Section VIII).

VI. ONE-STEP PROCESSES

One-step processes have been defined by processes that obey the master equation (2.8) or (2.10). However, it is necessary to specify in addition the range of $n$. There are three possibilities: (a) all integers, $-\infty < n < \infty$; (b) half-infinite range, $n = 0, 1, 2, \ldots$; (c) finite range, $n = 0, 1, 2, \ldots, N$. If the range consists of several intervals with gaps between them, a one-step process cannot have transitions between them, so that the process decomposes into several independent processes.

If $r(n)$ and $g(n)$ are constants and $n$ ranges from $-\infty$ to $+\infty$, the one-step process is identical with the (unsymmetric) random walk. The master equation can then easily be solved explicitly and no $\Omega$ expansion is needed. If $r$ and $g$ are constant and $n$ has a limited range, for example, $n = 0, 1, 2, \ldots, \infty$, then (2.8) cannot be valid for all $n$. It can at best hold for $n = 1, 2, \ldots$, whereas for $n = 0$ it must have a slightly different form. We shall then call the boundary at $n = 0$ artificial. The random walk with one or two artificial boundaries can still be solved explicitly and will therefore not be considered.

If $r(n)$ and $g(n)$ are linear functions of $n$, there must be at least one boundary to prevent them from becoming negative. Again this makes a modification of (2.8) necessary. The following particular case is of special interest and will be called a natural boundary. Suppose again $n = 0, 1, 2, \ldots$. Then $n = 0$ is a natural boundary if (Fig. 3)

(a) $r(0) = 0$ (6.1a)

(b) the modified equation at $n = 0$ is

$$
\dot{P}(0, t) = r(1)P(1, t) - g(0)P(0, t)
$$

(6.1b)

Note that this is identical with (2.8) for $n = 0$ if one knows that $P(-1, t) = 0$. 
Fig. 3. The one-step process with an absorbing boundary. The boundary is "natural" when \( r(0) = 0 \).

Although artificial boundaries often create considerable difficulties for solving the master equation\(^\text{17}\), natural boundaries do not—for the following reason. Consider the master equation (2.8) with natural boundary (6.1b). Now solve (2.8) for \( -\infty < n < \infty \), paying no attention to the boundary. Then take an initial state in which \( P(n, 0) = 0 \) for \( n < 0 \), for instance (1.7) with some nonnegative \( m \). It will now be true that \( P(n, t) \) for \( n < 0 \) remains zero at all \( t > 0 \); owing to (6.1a) no probability spills over to negative \( n \). Hence (6.1b) is automatically satisfied, since it is implied in (2.8) if one has \( P(-1, t) = 0 \).

An upper boundary \( n = N \) is called natural if \( g(N) = 0 \) and the modified equation for \( P(N, t) \) is obtained from (2.8) by setting \( P(N + 1, t) = 0 \). (In a way infinity might also be considered as a natural boundary.) The one-step process with linear or constant \( r(n) \) and \( g(n) \) and no other than natural boundaries can be solved explicitly, for example with the aid of generating functions.

If \( r(n) \) or \( g(n) \) or both are nonlinear functions, for instance polynomials, the definition (6.1) of a natural boundary remains valid. Explicit solutions of such master equations are rare, but it is always possible to find the stationary, that is, time-independent solution. For this purpose write (2.10) in the form

\[
0 = (E - 1) \{ r(n)P^{\text{st}}(n) - E^{-1}g(n)P^{\text{st}}(n) \} \tag{6.2}
\]

It follows that \( \{ \} \) must be constant

\[
r(n)P^{\text{st}}(n) - g(n - 1)P^{\text{st}}(n - 1) = J \tag{6.3}
\]

\( J \) is the net probability flow from \( n \) to \( n - 1 \). Using (6.3) one can construct the successive \( P^{\text{st}}(n) \), starting from a single one, for instance \( P^{\text{st}}(0) \), which then serves as a normalizing factor.

If there is a natural boundary, for instance at \( n = 0 \), one finds on substituting \( n = 0 \) in (6.3) that \( J \) must vanish:

\[
r(n)P^{\text{st}}(n) = g(n - 1)P^{\text{st}}(n - 1) \tag{6.4}
\]
It then follows directly that

\[ P^{\text{st}}(n) = \frac{g(n - 1)g(n - 2)\cdots g(0)}{r(n)r(n - 1)\cdots r(1)} P^{\text{st}}(0) \]  

(6.5)

The normalizing factor \( P^{\text{st}}(0) \) is subsequently found from

\[ \left[ P^{\text{st}}(0) \right]^{-1} = \sum_{n=0}^{N} \frac{g(n - 1)g(n - 2)\cdots g(0)}{r(n)r(n - 1)\cdots r(1)} \]  

(6.6)

When the upper bound \( N \) is infinite it may happen that the sum does not converge. In that case every solution \( P(n, t) \) continues to spread out indefinitely, in the same way as in the familiar random walk.

It should be emphasized that (6.4) is simply a mathematical identity for one-step processes. It has to be distinguished from detailed balance, which for one-step processes reads

\[ r(n)P^{\text{eq}}(n) = g(n - 1)P^{\text{eq}}(n - 1) \]  

(6.7)

Here \( P^{\text{eq}} \) is the thermal equilibrium distribution and is known \( \text{a priori} \) from the familiar phase space argument of equilibrium statistical mechanics. On the one hand, detailed balance is not restricted to one-step processes; on the other hand it only applies to closed physical systems, without magnetic field or overall rotation. The identity (6.4) also holds for open systems, for example, the photoconductor mentioned in the next section, and for population problems.

VII. SEMICONDUCTOR

As a second example of a nonlinear one-step process consider the following model of an intrinsic semiconductor. A crystal has a nearly empty conduction band and a nearly full valence band. Let \( n \) denote the number of electrons that by thermal fluctuations have been excited into the conduction band. The probability per unit time for an excitation to occur is \( g(n) = \beta \Omega \), where \( \Omega \) is the volume of the crystal and \( \beta \) a constant (see Fig. 4). The probability for a recombination is proportional to the number of excited electrons and to the density \( n/\Omega \) of the available holes: \( r(n) = \gamma n^2/\Omega \). Thus the macroscopic rate equation is

\[ \frac{dn}{dt} = \beta \Omega - \frac{\gamma}{\Omega} n^2 \]  

(7.1)

On the mesoscopic level the process is specified by the transition probabilities:

\[ W(n|n') = \beta \Omega \delta_{n,n-1} + \frac{\gamma}{\Omega} n^2 \delta_{n,n'} \]  

(7.2)
Fig. 4. Model for a semiconductor or a photoconductor.

This is actually of the form stipulated in (4.1)—with \( f(\Omega) = \Omega \) and vanishing \( \Phi_1, \Phi_2, \ldots \). The master equation for the probability \( P(n, t) \) of having \( n \) excited electrons is

\[
P(n, t) = \beta \Omega (E^{-1} - 1)P + \frac{\gamma}{\Omega} (E - 1)n^2 P
\]  

(7.3)

The range of \( n \) is \( \{0, 1, 2, \ldots\} \) and the equation has a natural boundary at \( n = 0 \). The stationary solution is found from (6.5) to be

\[
P^{st}(n) = \frac{\Omega^{2n}}{n!^2} \left( \frac{\beta}{\gamma} \right)^n P^{st}(0)
\]  

(7.4)

For large \( \Omega \) this represents a sharp peak whose position and width are determined by

\[
\langle (n - \langle n \rangle_{st})^2 \rangle_{st} = \frac{1}{2} \langle n \rangle_{st}^2 = \frac{1}{2} \Omega \sqrt{\beta/\gamma}
\]  

(7.5)

The distribution (7.4) must be the same as the Boltzmann distribution given by equilibrium statistical mechanics. Indeed, \( \Omega^{2n} \) is the phase volume of the \( n \) electrons and \( n \) holes, and each has a Gibbs factorial. Thus the remaining factor in (7.4) must be the energy exponential:

\[
\frac{\beta}{\gamma} = e^{-\epsilon/kT}
\]  

(7.6)

where \( \epsilon \) is the energy gap between both bands.

The same master equation (7.3), however, also describes a photoconductor. In that case \( \beta \) has an additional term due to excitation by incident photons, so that (7.6) is no longer true. We shall therefore not make use of it. Note, however, that it is necessary that the arrival times of the photons are uncorrelated (shot noise); otherwise their effect cannot be taken into account.
by a simple addition to the excitation probability but a much more elaborate
treatment is necessary.\textsuperscript{19}

The jump moments are in the $\Omega$ language:

$$\alpha_p(\varphi) = \beta + (-1)^p \gamma \varphi^2$$  \hfill (7.7)

Hence the macroscopic equation is, according to (4.7),

$$\dot{\varphi} = \beta - \gamma \varphi^2$$ \hfill (7.8)

This equation can readily be solved, but all we shall need to know is that
every solution tends to

$$\varphi^{st} = \sqrt{\frac{\beta}{\gamma}}$$ \hfill (7.9)

which is immediately seen from (7.8) [compare (7.5)]. The Fokker-Planck
equation for the fluctuations is, according to (4.8),

$$\frac{\partial \Pi}{\partial t} = 2\gamma \varphi \frac{\partial}{\partial \varphi} \varphi \Pi + \frac{1}{2} (\beta + \gamma \varphi^2) \frac{\partial^2 \Pi}{\partial \varphi^2}$$ \hfill (7.10)

One finds from this [compare (5.1) and (5.2)]

$$\frac{d}{dt} \langle x \rangle = -2\gamma \varphi \langle x \rangle$$ \hfill (7.11)

$$\frac{d}{dt} \langle x^2 \rangle = -4\gamma \varphi \langle x^2 \rangle + \beta + \gamma \varphi^2$$ \hfill (7.12)

The extended equation (5.5) for $\langle x \rangle$ is

$$\frac{d}{dt} \langle x \rangle = -2\gamma \varphi \langle x \rangle - \gamma \Omega^{-1/2} \langle x^2 \rangle$$ \hfill (7.13)

which together with (7.12) determines $\langle x \rangle$ to order $\Omega^{-1/2}$ and therefore $\langle n \rangle$
to order $\Omega^0$.

As the solution of these equations is rather laborious we shall merely
study the fluctuations in the stationary state. On substituting (7.9) in (7.11)
one finds

$$\langle x(t) \rangle = \langle x(0) \rangle e^{-2t \sqrt{\frac{\beta}{\gamma}}}$$ \hfill (7.14)

From (7.12) one obtains, in agreement with (7.5),

$$\langle x^2 \rangle^{st} = \frac{\beta + \gamma \varphi^2}{4\gamma \varphi} = \frac{1}{2} \sqrt{\frac{\beta}{\gamma}}$$ \hfill (7.15)
These data suffice to find the autocorrelation function of $x$,

$$\langle x(0)x(t) \rangle^st = \frac{1}{2} \sqrt{\frac{\beta}{\gamma}} \ e^{-2t\sqrt{\beta\gamma}}$$  \hspace{1cm} (7.16)

The autocorrelation function of $n$ is therefore

$$\langle \{n(0) - \langle n \rangle^st\} \{n(t) - \langle n \rangle^st\} \rangle^st = \frac{1}{2} \sqrt{\frac{\beta}{\gamma}} \ e^{-2t\sqrt{\beta\gamma}}$$  \hspace{1cm} (7.17)

where terms of order $Q_1I_2$ have been neglected.

This autocorrelation function consists of a single exponential, so that the spectral density of the fluctuations consists of a single Debye term with relaxation time $[2\sqrt{\beta\gamma}]^{-1}$. That result is not surprising since to this order the noise is treated in linear approximation. In the next section it will be seen that the higher orders give rise to additional exponentials involving other relaxation times.

**VIII. HIGHER-ORDER CORRECTIONS**

It is easy to work out the higher orders of (4.4) beyond the power $\Omega^0$ displayed in (4.8). The result is

$$\frac{\partial \Pi}{\partial t} = -\frac{\partial}{\partial x} \left\{ x_1x + \frac{1}{2} \Omega^{-1/2}x_2x^2 + \frac{1}{3!} \Omega^{-1}x_3x^3 + \cdots \right\} \Pi$$

$$+ \frac{1}{2} \frac{\partial^2}{\partial x^2} \left\{ x_2 + \Omega^{-1/2}x_2x + \frac{1}{2} \Omega^{-1}x_2x^2 + \cdots \right\} \Pi$$

$$- \frac{1}{3!} \Omega^{-1/2} \frac{\partial^3}{\partial x^3} \left\{ x_3 + \Omega^{-1/2}x_3x + \cdots \right\} \Pi$$

$$+ \frac{1}{4!} \Omega^{-1} \frac{\partial^4}{\partial x^4} \left\{ x_4 + \cdots \right\} \Pi + \cdots$$  \hspace{1cm} (8.1)

All functions $x$ have the argument $\varphi$, the prime indicates differentiation with respect to $\varphi$. All terms of order $\Omega^{-1}$ have been written down, and it is clear how the expansion continues: each order of $\Omega^{-1/2}$ adds one power of $x$ to the coefficients and at the same time an additional derivative appears. The nonlinear Fokker-Planck equation, which is so often used for describing nonlinear random processes in physics, contains all terms on the first two lines and ignores all other lines; clearly that is an inconsistent approximation, unfit to describe anything beyond the linear noise approximation (see Section XXII).

One cannot expect to solve the full equation (8.1), but it is remarkable that one can find the moments to any given order explicitly. For example, to
find \( \langle x \rangle \) to order \( \Omega^{-1/2} \) we write

\[
\frac{d}{dt} \langle x \rangle = \alpha'_1 \langle x \rangle + \frac{1}{2} \Omega^{-1/2} \alpha'_1 \langle x^2 \rangle + \mathcal{O}(\Omega^{-1}) \quad (8.2a)
\]

\[
\frac{d}{dt} \langle x^2 \rangle = 2 \alpha'_1 \langle x^2 \rangle + \nu_2 + \mathcal{O}(\Omega^{-1/2}) \quad (8.2b)
\]

These are the two equations already discussed in Sections II and V.

We shall now calculate the next correction to the autocorrelation function (7.16) or (7.17) of the model in the previous section. It is readily seen that the approximation (8.2) does not contribute owing to symmetry between positive and negative values of \( x \). Hence we have to go one step further to find how the nonlinearity affects the autocorrelation function and thereby the fluctuation spectrum. To save writing we rescale the variables,

\[
2t\sqrt{\beta \gamma} = \tau, \quad (\gamma/\beta)^{1/4} x = \epsilon, \quad (\gamma/\beta)^{1/4} \Omega^{-1/2} = \epsilon \quad (8.3)
\]

Then (8.1) to order \( \epsilon^2 \) takes the form

\[
\frac{\partial \Pi}{\partial \tau} = \frac{\partial}{\partial \xi} \left\{ \xi + \frac{1}{2} \epsilon^2 \xi^2 \right\} \Pi + \frac{1}{2} \frac{\partial^2}{\partial \xi^2} \left\{ 1 + \epsilon \xi + \frac{1}{2} \epsilon^2 \xi^2 \right\} \Pi
\]

\[
+ \frac{1}{3} \epsilon^2 \frac{\partial^3}{\partial \xi^3} \xi \Pi + \frac{1}{24} \epsilon^2 \frac{\partial^4 \Pi}{\partial \xi^4} \quad (8.4)
\]

The equations for the first four moments are to the required order

\[
\frac{d}{d\tau} \langle \xi \rangle = - \langle \xi \rangle - \frac{1}{2} \epsilon \langle \xi^2 \rangle + \mathcal{O}(\epsilon^3) \quad (8.5a)
\]

\[
\frac{d}{d\tau} \langle \xi^2 \rangle = -2 \langle \xi^2 \rangle - \epsilon \langle \xi^3 \rangle + 1 + \epsilon \langle \xi \rangle + \frac{1}{2} \epsilon^2 \langle \xi \rangle + \mathcal{O}(\epsilon^3) \quad (8.5b)
\]

\[
\frac{d}{d\tau} \langle \xi^3 \rangle = -3 \langle \xi^3 \rangle - \frac{3}{2} \epsilon \langle \xi^4 \rangle + 3 \langle \xi \rangle + 3 \epsilon \langle \xi^2 \rangle + \mathcal{O}(\epsilon^2) \quad (8.5c)
\]

\[
\frac{d}{d\tau} \langle \xi^4 \rangle = -4 \langle \xi^4 \rangle + 6 \langle \xi^2 \rangle + \mathcal{O}(\epsilon) \quad (8.5d)
\]

From this we first conclude

\[
\langle \xi^4 \rangle = \frac{3}{2} \langle \xi^2 \rangle^2 = \frac{3}{2} + \mathcal{O}(\epsilon) \quad (8.6a)
\]

\[
\langle \xi^3 \rangle = -\frac{3}{2} \epsilon - \frac{1}{2} \xi + \frac{1}{2} \epsilon = -\frac{1}{8} \xi + \mathcal{O}(\epsilon^2) \quad (8.6b)
\]

\[
\langle \xi^2 \rangle = \frac{1}{2} - \frac{1}{16} \epsilon^2 + \mathcal{O}(\epsilon^3) \quad (8.6c)
\]

\[
\langle \xi \rangle = -\frac{1}{2} \epsilon + \mathcal{O}(\epsilon^3) \quad (8.6d)
\]
Notice that to order $\Omega^{-1}$ it is no longer true that $\langle \xi \rangle = 0$, so that the macroscopic part $\Omega \varphi$ of $n$ is no longer identical with the average $\langle n \rangle$.

Subsequently solve the equations for the first three moments as functions of time to the desired order in $\epsilon$. The initial conditions are $\langle \xi \rangle_0 = \xi_0$, $\langle \xi^2 \rangle_0 = \xi_0^2$, $\langle \xi^3 \rangle_0 = \xi_0^3$. Finally multiply $\langle \xi(t) \rangle_0$ with $\xi_0$ and average $\xi_0$ over the stationary distribution. The result is the autocorrelation function:

$$\langle \xi_0 \langle \xi(t) \rangle \rangle \sim - \langle \xi^2 \rangle \langle \xi \rangle^2 = \frac{1}{2} (1 - \frac{1}{2} \epsilon^2) e^{-\left(1 - \frac{1}{2} \epsilon^2\right)^2} + \frac{1}{8} \epsilon^2 e^{-\left(2 + \epsilon^2\right)^2}$$

The coefficients and the exponents are correct to order $\epsilon^2$.

From this result one sees that the order $\Omega^{-1}$ lowers the rate of decay of the leading term, and modifies its coefficient. More strikingly, however, another exponential term appears which decays roughly twice as fast. Thus the nonlinearity gives rise to an additional Debye term in the fluctuation spectrum, which in principle could be observed. Higher orders give rise to a sequence of such terms.

The microscopic treatment of Bernard and Callen$^{20}$ led them to conclude that the nonlinearity does not affect the fluctuation spectrum at all. Admittedly, the present mesoscopic treatment is founded on the assumption that the master equation applies, but there are doubts about the applicability of the microscopic results as well.$^{5,21}$ It would be of great interest to perform an experimentum crucis.

IX. THE MALTHUS–VERHULST PROBLEM

In a population of $n$ individuals each individual has a probability $\alpha \, dt$ to die in the next $dt$, and a probability $\beta \, dt$ to give birth to an additional individual. Moreover the struggle for life gives rise to an additional death rate, which is proportional to the density $(n - 1)/\Omega$ of other individuals, where $\Omega$ is the amount of space or food available. The growth of the population is described by the Malthus–Verhulst equation,$^{22}$

$$\frac{dn}{dt} = (\beta - \alpha)n - \frac{\gamma}{\Omega} n(n - 1) \quad (9.1)$$

On the mesoscopic level one has a transition probability

$$W(n|n') = \alpha n^p \delta_{n,n-1} + \beta (n') \delta_{n,n'+1}$$

$$= \left\{ \alpha n' + \frac{\gamma}{\Omega} n'(n' - 1) \right\} \delta_{n,n-1} + \beta n' \delta_{n,n'+1} \quad (9.2)$$

It has the form (4.1) with $f(\Omega) = \Omega$ including a term

$$\Phi_1(n'/\Omega; n - n') = \gamma (n'/\Omega) \delta_{n,n'-1} \quad (9.3)$$
which has been underlined in (9.2). For this reason we shall carry out the expansion once more explicitly. The result will be that this term neither contributes to the macroscopic equation, nor to the linear noise approximation, but only to higher orders.

The master equation is

$$\dot{P}(n, t) = \alpha(E - 1)nP + \beta(E^{-1} - 1)nP + \frac{\gamma}{\Omega} (E - 1)n(n - 1)P$$  \hspace{1cm} (9.4)$$

Substitute the transformation (3.3), (3.4) using (3.6)

$$\frac{\partial\Pi}{\partial t} - \Omega^{1/2} \frac{d\varphi}{dt} \frac{\partial\Pi}{\partial x} = \alpha \Omega \left\{ \Omega^{-1/2} \frac{\partial}{\partial x} + \frac{1}{2} \Omega^{-1} \frac{\partial^2}{\partial x^2} \right\} (\varphi + \Omega^{-1/2}x) \Pi$$

$$+ \beta \Omega \left\{ -\Omega^{-1/2} \frac{\partial}{\partial x} + \frac{1}{2} \Omega^{-1} \frac{\partial^2}{\partial x^2} \right\} (\varphi + \Omega^{-1/2}x) \Pi$$

$$+ \gamma \Omega \left\{ \Omega^{-1/2} \frac{\partial}{\partial x} + \frac{1}{2} \Omega^{-1} \frac{\partial^2}{\partial x^2} \right\}$$

$$\times (\varphi + \Omega^{-1/2}x)(\varphi + \Omega^{-1/2}x - \Omega^{-1}) \Pi$$  \hspace{1cm} (9.5)$$

Collecting the terms of order $\Omega^{1/2}$ one obtains the macroscopic equation

$$-\dot{\varphi} = (\alpha - \beta)\varphi + \gamma \varphi^2$$  \hspace{1cm} (9.6)$$

It is the same equation as (9.1) with $n(n - 1)$ replaced by $n^2$. This difference is meaningless on the macroscopic level, because on this level one also ignores the difference between $\langle n^2 \rangle$ and $\langle n \rangle^2$, which is of the same order. It is therefore permissible, and in fact more consistent, to write $n^2$ in (9.1), as is usually done.

The solution of (9.6) is

$$\varphi(t) = \frac{\varphi(0)e^{(\beta - \alpha)t}}{1 + \varphi(0) \frac{\gamma}{\beta - \alpha} \{e^{(\beta - \alpha)t} - 1\}}$$  \hspace{1cm} (9.7)$$

For $\beta < \alpha$ it tends to zero: the population dies out because the death rate exceeds the birth rate. For $\beta > \alpha$ it tends to

$$\varphi_{\text{st}} = \frac{\beta - \alpha}{\gamma}$$  \hspace{1cm} (9.8)$$

which is a balance between the natural growth and the mutual interference.

The terms of order $\Omega^0$ in (9.5) yield the Fokker-Planck equation

$$\frac{\partial \Pi}{\partial t} = (\alpha - \beta + 2\gamma \varphi) \frac{\partial}{\partial x} x\Pi + \frac{1}{2} (\alpha \varphi + \beta \varphi + \gamma \varphi^2) \frac{\partial^2 \Pi}{\partial x^2}$$  \hspace{1cm} (9.9)$$
Note that the underlined term in (9.2) and (9.5) still does not contribute. From (9.9) one obtains for the first two moments

\[
\frac{d}{dt} \langle x \rangle = (\beta - \alpha - 2\gamma \phi) \langle x \rangle
\]

(9.10)

\[
\frac{d}{dt} \langle x^2 \rangle = 2(\beta - \alpha - 2\gamma \phi) \langle x^2 \rangle + (\alpha \phi + \beta \phi + \gamma \phi^2)
\]

(9.11)

In particular, after \( \phi \) has reached its stationary value (9.8) the fluctuations are determined by

\[
\frac{d}{dt} \langle x \rangle = -(\beta - \alpha) \langle x \rangle
\]

(9.12)

\[
\frac{d}{dt} \langle x^2 \rangle = -2(\beta - \alpha) \langle x^2 \rangle + 2\beta(\beta - \alpha)/\gamma
\]

(9.13)

Notice that whereas in the macroscopic equation (9.6) the quantities \( \alpha \) and \( \beta \) only occur in the combination \( \alpha - \beta \), (9.13) contains \( \beta \) separately. This demonstrates how the observation of fluctuations permits one to find more information about a system than mere macroscopic measurements. Of course this is well known; observation of Brownian motion or critical opalescence permits one to determine Avogadro's number.

In order to check the validity of the expansion in \( \Omega \) we have to investigate whether the fluctuations remain small of the order anticipated in (3.3). First, for \( \beta < \alpha \) there is one stationary value \( \phi_{st} = 0 \), and it is asymptotically stable since all other solutions tend to zero [see (9.7)]. It is evident from Fig. 5a that the fluctuations tend to decrease rather than to grow, and this is confirmed by the explicit calculation in the Appendix.

When \( \beta > \alpha \) the stationary solution \( \phi = 0 \) is unstable; all other solutions tend to (9.8). It is clear from Fig. 5b that the fluctuations may be magnified for some time, but will ultimately decrease again. Hence they cannot grow such that they violate the assumption they are of order \( \Omega^{1/2} \).

![Fig. 5a. The solutions of the macroscopic equation (9.6) for \( \beta < \alpha \).](image-url)
The fact that for different values of the parameters different stationary states prevail may be considered as a phase transition. This point of view will be amplified in Section XX, but it is useful to work out some details in the context of the present example.

In the previous section it was tacitly understood that $|\alpha - \beta|$ was of order unity. There is a transition region, however, where it is small of order $\Omega^{-1/2}$.

In that case the stable and unstable stationary states are not clearly separated, since their distance is of the same order as the fluctuations. In order to treat this we set $\beta - \alpha = \Omega^{-1/2} \Delta$, with constant $\Delta$, either positive or negative. Without loss of generality we also take for convenience $\beta + \alpha = 1$, $\gamma = 1$.

The transformed master equation (9.5) may now be written

$$\frac{\partial \Pi}{\partial t} - \Omega^{1/2} \frac{d\varphi}{dt} \frac{\partial \Pi}{\partial x} = -\Delta \varphi \frac{\partial \Pi}{\partial x} + \frac{1}{2} \varphi \frac{\partial^2 \Pi}{\partial x^2}$$

$$+ \Omega \left( \Omega^{-1/2} \frac{\partial}{\partial x} + \frac{1}{2} \Omega^{-1} \frac{\partial^2}{\partial x^2} \right) (\varphi + \Omega^{-1/2} x)^2 \Pi$$

(10.1)

The terms of order $\Omega^{1/2}$ yield

$$\frac{d\varphi}{dt} = -\varphi^2$$

(10.2)

This agrees with (9.6) since $\alpha$ and $\beta$ are now equal from a macroscopic point of view. The terms of order $\Omega^0$ yield

$$\frac{\partial \Pi}{\partial t} = \varphi \frac{\partial}{\partial x} (-\Delta + 2x) \Pi + \frac{1}{2} (\varphi + \varphi^2) \frac{\partial^2 \Pi}{\partial x^2}$$

(10.3)

Note that this equation is essentially different from (9.7) and cannot be obtained from it as a limiting case.
The solution of (10.2) with initial value $\varphi_0$ is

$$\varphi(t) = \frac{\varphi_0}{1 + t\varphi_0}$$

Thus the macroscopic part of $n$ tends to zero, but merely as $t^{-1}$ rather than exponentially. On the other hand, the average of the fluctuations obeys according to (10.3)

$$\frac{d}{dt} \langle x \rangle = \varphi(t)(\Delta - 2\langle x \rangle)$$

This is no longer identical with the variational equation associated with (10.2). The solution with initial value $\langle x \rangle_0 = 0$ is

$$\langle x \rangle_t = \frac{\Delta}{2} \left\{ 1 - \frac{1}{(1 + t\varphi_0)^2} \right\}$$

Hence it slowly tends to $\frac{1}{2}\Delta$ rather than to zero. In this case therefore, already in order $\Omega^0$ the average $\langle n \rangle$ does not coincide with the macroscopic part $\Omega \varphi$ of $n$.

One also finds after some algebra

$$\langle x^2 \rangle_t - \langle x \rangle_t^2 = \frac{1}{4} \left\{ 1 - \frac{1}{(1 + t\varphi_0)^4} \right\} + \frac{\varphi_0}{3} \frac{1}{1 + t\varphi_0} \left\{ 1 - \frac{1}{(1 + t\varphi_0)^3} \right\}$$

which tends to $\frac{1}{4}$. Thus when $t \gg \varphi_0^{-1}$ the distribution $P(n, t)$ consists of a peak with center $\frac{1}{2}\Omega^{1/2}\Delta$ and width $\frac{1}{2}\Omega^{1/2}$.

This cannot be the whole story, however, because according to this picture $n$ would also take negative values. The present model has the additional complication that one of the two stationary states also happens to be a boundary. The above solution can therefore only be trusted as long as the fluctuations do not reach that boundary:

$$\Omega \varphi(t) \gg \Omega^{1/2} \sqrt{\langle x^2 \rangle_t}$$

This condition amounts to $t \ll \Omega^{1/2}$. Thus in this case the expansion in powers of $\Omega^{-1/2}$ is no longer uniformly valid for all $t$, and cannot provide information about the behavior for $t \to \infty$.

It is obvious what that behavior is. Whenever by a fluctuation $n$ reaches the value zero the population has died out, so that $n$ remains zero ever after. No spontaneous generation occurs in the Malthus–Verhulst equation, in contrast with the semiconductor in Section VII. Thus $n = 0$ is an absorbing state and the Fokker–Plank equation (10.1) for $\Pi$ should be solved with an
absorbing boundary condition at \( n = 0 \). In terms of \( x \) the boundary moves as determined by

\[
x = -\Omega^{1/2} \varphi(t)
\]

Absorption at the boundary is represented by the condition that \( \Pi \) must vanish, hence one has to solve (10.1) with the boundary condition \( \Pi(-\Omega^{1/2} \varphi(t), t) = 0 \). Of course in principle this is also true for (9.9) in the previous section, but in that case \( P(0, t) \) is small of order \( \exp(-\Omega) \), so that error is negligible (compare Section XX).

\section*{XI. THE LANGEVIN APPROACH}

The success of Langevin's treatment of Brownian motion has led to the erroneous idea that all fluctuations can be treated in that way. The method consists of three steps.

(i) One supplements the macroscopic equation with a stochastic driving force. In the notation of Section II

\[
\frac{dn}{dt} = a_1(n) + l(t)
\]

This equation determines a stochastic process \( n(t) \), whose mean and variance are supposed to provide the macroscopic value and the fluctuations of \( n \), respectively. Of course (11.1) is moot unless one says something about \( l(t) \). First the usually tacit assumption is made that the statistical properties of \( l \) are independent of \( n(t) \).

(ii) Subsequently two more explicit assumptions are made:

\[
\langle l(t) \rangle = 0, \quad \langle l(t_1)l(t_2) \rangle = \Gamma \delta(t_1 - t_2)
\]

with constant \( \Gamma \). When \( a_1(n) \) is linear in \( n \) these properties are sufficient for computing the first two moments of \( n(t) \).\textsuperscript{23} If one is interested in higher moments, or when \( a_1(n) \) is nonlinear, one needs the higher moments of \( l(t) \) as well. They are ordinarily chosen by assuming \( l(t) \) to be Gaussian, that is, all higher cumulants of \( l(t) \) vanish, or all higher moments of \( l(t) \) factorize, for example,

\[
\langle l(t_1)l(t_2)l(t_3)l(t_4) \rangle = \langle l(t_1)l(t_2) \rangle \langle l(t_2)l(t_4) \rangle + \langle l(t_1)l(t_3) \rangle \langle l(t_2)l(t_4) \rangle + \langle l(t_1)l(t_4) \rangle \langle l(t_2)l(t_3) \rangle
\]

Whereas (11.2) makes physical sense for the force exerted by gas molecules on a Brownian particle, in other cases it is often doubtful, and of course (11.3) goes far beyond all physical intuition.
(iii) The macroscopic stationary value of \( n \) is found from \( a_1(n^n) = 0 \). It is stable if \( a'_1(n^n) < 0 \). One then obtains for the fluctuations in linear approximation:

\[
\frac{d\Delta n}{dt} = a'_1(n^n)\Delta n + l(t)
\]  

(11.4)

On solving this equation\(^{23}\) one finds in the limit \( t \to \infty \):

\[
\langle \Delta n^2 \rangle_\infty = \frac{\Gamma}{2} |x'_1(n^n)|
\]  

(11.5)

If one is dealing with a system in thermal equilibrium, (11.5) must be identical with the equilibrium fluctuation \( \langle \Delta n^2 \rangle^{eq} \) as known from equilibrium statistical mechanics. This identification determines \( \Gamma \), supposing that the macroscopic \( a_1(n) \) is known—which is the fluctuation-dissipation theorem. If one is dealing with a stationary state other than thermal equilibrium, no \( a \) priori knowledge of the mean square fluctuations is available and the value of \( \Gamma \) must be found in another way.

We shall now test the various assumptions on which this approach is based by comparing its results with those of the systematic expansion. Throughout we are only concerned with the weaker version (11.2) rather than (11.3). Moreover by not using (11.5) the discussion is not confined to thermal equilibrium.

On averaging (11.4) one obtains for the average of the fluctuations near the stationary state

\[
\frac{d\langle \Delta n \rangle}{dt} = a'_1(n^n)\langle \Delta n \rangle
\]  

(11.6)

This is identical with (5.1) when in the latter the stationary value of \( \phi \) is substituted. Also one finds in the usual way

\[
\frac{\langle (\Delta n)^2 \rangle}{\Delta t} = 2a'_1(n^n)\langle (\Delta n)^2 \rangle + \Gamma
\]  

(11.7)

This is identical with (5.2) if one takes \( \Gamma = a_2(n^n) \). Thus the Langevin approach yields equations for \( \langle \Delta n \rangle \) and \( \langle (\Delta n)^2 \rangle \) in the stationary state that are correct to the same approximation as (2.3), (2.7), that is, with neglect of \( \mathbf{c}(\Omega^0) \) and \( \mathbf{c}(\Omega^{1/2}) \), respectively.

However, let us consider the fluctuations around a nonstationary solution. On averaging (11.1) one obtains (2.2), which is exact, but not identical with the macroscopic equation (2.3) unless \( a_1(n) \) happens to be a linear function.
An equation for $\langle (\Delta n)^2 \rangle$ cannot even be deduced from (11.1) with (11.2). Actually the difficulty can be demonstrated on a simple linear example.

A radioactive sample consists of identical nuclei, each having a probability $\lambda$ per unit time to decay. The transition probability per unit time for the number $n$ of active nuclei is

$$W(n|n') = \lambda n' \delta_{n,n'-1}$$

(11.8)

Hence

$$a_\mu(n) = (-1)^\mu \lambda n$$

(11.9)

so that the exact equations (2.2) and (2.5) take the form

$$\frac{d}{dt} \langle n \rangle = -\lambda \langle n \rangle$$

$$\frac{d}{dt} \sigma_n^2 = \lambda \langle n \rangle - 2\lambda \sigma_n^2$$

(11.10)

Their solutions with the initial condition (1.7) are

$$\langle n \rangle_t = me^{-\lambda t}$$

$$\sigma_n^2(t) = me^{-\lambda t}(1 - e^{-\lambda t}) = \langle n \rangle_t(1 - e^{-\lambda t})$$

(11.11)

The Langevin approach, on the other hand, starts from the equation

$$\frac{dn}{dt} = -\lambda n + l(t)$$

(11.12)

with the solution

$$n(t) = me^{-\lambda t} + e^{-\lambda t} \int_0^t e^{\lambda t'} l(t') \, dt'$$

(11.13)

On averaging one duly finds (11.10). However, on squaring (11.13)

$$\langle n(t)^2 \rangle = m^2 e^{-2\lambda t} + \frac{\Gamma}{2\lambda} (1 - e^{-2\lambda t})$$

so that

$$\sigma_n(t)^2 = \frac{\Gamma}{2\lambda} (1 - e^{-2\lambda t})$$

(11.14)

Clearly it is not possible to choose the constant $\Gamma$ in order to find agreement with (11.11). A fortiori the fluctuation–dissipation value for $\Gamma$ cannot be
correct; in fact for the present case it would give \( \Gamma = 0 \). It is true that (11.14) reduces to (11.11) if one puts
\[
\Gamma = 2\lambda m (e^{\Delta t} + 1)^{-1}
\]
but that would make \( \Gamma \) depend not only on time, but also on the special solution considered. The conclusion is therefore that fluctuations around a time-dependent state cannot be treated by the Langevin method.

**XII. THE GENERATION AND RECOMBINATION CURRENTS**

Consider the one-step master equation [(2.8) or (2.10)]. The solution \( P(n, t) \) is the probability distribution of \( n \) at time \( t \), resulting from the random up and down jumps of \( n \). We now ask how these jumps are distributed in time. Rather than treating the general case we shall concentrate on the recombination events in the stationary state of the semiconductor model of Section VII. In a photoconductor these events may emit photons, which can be observed. In population problems the result might be of interest to morticians.

The recombination events constitute a random sequence of dots on the time axis. An appropriate tool for describing the statistical properties of such sequences is furnished by the hierarchy of distribution functions,
\[
fn(t_1, t_2, \ldots, t_n) \quad (n = 1, 2, 3, \ldots)
\]
They are defined as follows. First \( f_1(t_1) \ dt_1 \) is the probability for having an event in the time interval \((t_1, t_1 + dt_1)\), regardless of what happens outside that interval. The probability for having two or more events in \((t_1, t_1 + dt_1)\) is of higher order in \( dt \) and therefore negligible. Next \( f_2(t_1, t_2) \ dt_1 \ dt_2 \) is the joint probability for having one event in \((t_1, t_1 + dt_1)\) and another in \((t_2, t_2 + dt_2)\), regardless of all other events. Since this definition only holds for \( t_1 \neq t_2 \) we have to add that on integrating \( f_2 \) no extra contribution arises from \( t_1 = t_2 \); that is, \( f_2 \) contains no terms with \( \delta(t_1 - t_2) \).

The higher \( f_n \) are defined in the same way, but we shall only compute \( f_1 \) and \( f_2 \). It is often convenient to express the result in terms of the correlation functions \( g_n \), which are related to the \( f_n \) by the familiar cluster expansion,
\[
g_1(t_1) = f_1(t_1), \quad g_2(t_1, t_2) = f_2(t_1, t_2) - f_1(t_1)f_1(t_2)
\]
For independent events \( g_2 \) and the higher \( g_n \) vanish. By definition \( f_2 \) and \( g_2 \) are symmetric functions of \( t_1, t_2 \). When a sequence of events is stationary its \( f_1 \) is independent of time, and \( f_2 \) only depends on \( t_1 - t_2 \).

Now let \( P(n, t) \) be a solution of (2.10) and therefore describe a mesoscopic state of the system. The first term describes the recombination events. The probability for the occurrence of a jump from \( n \) to \( n - 1 \) in \((t_1, t_1 + dt_1)\) is
\[
r(n_1) \ dt_1 \ P(n_1, t_1)
\]
The probability per unit time for a recombination to occur is therefore

\[ f_1(t_1) = \sum_{n_1=1}^{\infty} r(n_1)P(n_1, t_1) = \langle r(n_1) \rangle_{t_1} \quad (12.1) \]

This is the average recombination current at time \( t_1 \).

In order to compute \( f_2(t_1, t_2) \) we have to find the joint probability for having a jump from \( n_1 \) to \( n_1 - 1 \) in \( (t_1, t_1 + dt_1) \) and subsequently a jump from \( n_2 \) to \( n_2 - 1 \) in \( (t_2, t_2 + dt_2) \). This is given by

\[ r(n_1) \ dt_1 P(n_1, t) \cdot r(n_2) \ dt_2 P(n_2, t_2 | n_1 - 1, t_1) \]

The last factor is the conditional probability (1.8), which describes the evolution from \( t_1 \) to \( t_2 \). It starts from the initial value \( n_1 - 1 \), because a recombination took place at \( t_1 \). This detail cannot be ignored as was first remarked by Ubbink.\(^{18}\) Thus we find

\[ f_2(t_1, t_2) = \sum_{n_1, n_2} r(n_1)r(n_2)P(n_1, t_1)P(n_2, t_2 | n_1 - 1, t_1) \quad (12.2) \]

Of course similar expressions obtain for the \( f_1 \) and \( f_2 \) of the generation events and it is easy to see how they generalize to higher \( f_n \). They are exact, but in order to evaluate them one needs to know the solutions of the master equation.

For more explicit results take the stationary state (7.4) of the semiconductor. According to (12.1) and (3.3)

\[ f_1 = \gamma \frac{\Omega}{n} \sum_{n=1}^{\infty} n^2 P^n(n) = \beta \Omega + o(\Omega^{-1}) \quad (12.3) \]

To show that the term of order \( \Omega^0 \) vanishes use has been made of

\[ \langle x \rangle^s = -\frac{1}{4} \Omega^{-1/2} \quad (12.4) \]

which follows from (8.6d) with the aid of (8.3).

The evaluation of (12.2) takes some algebra. We go to order \( \Omega \) and are careful to include all the necessary powers of \( \Omega \). The symbol \( \phi \) throughout denotes \( \phi^n = \sqrt{\beta/\gamma} \), and \( \Pi(x) \) is the stationary distribution.

\[ f_2(t_1, t_2) = \gamma^2 \Omega^2 \int \int \{ \phi + \Omega^{-1/2} x_2 \}^2 \Pi(x_2, t_2 | x_1 - \Omega^{-1/2}, t_1) \]

\[ \times \{ \phi + \Omega^{-1/2} x_1 \}^2 \Pi(x_1) \ dx_1 \ dx_2 \quad (12.5) \]
Begin by working out the first quadratic factor.

\begin{equation}
\begin{aligned}
f_2(t_1, t_2) &= \gamma^2 \Omega^2 \varphi^2 \int \{\varphi + \Omega^{-1/2}x_1\}^2 \Pi(x_1) \, dx_1 \\
&\quad + 2\gamma^2 \Omega^{3/2} \varphi \int \int x_2 \Pi(x_2, t_2 | x_1, t_1) \, dx_1 \, dx_2 \\
&\quad \times \{\varphi + \Omega^{-1/2}x_1\}^2 \Pi(x_1) \, dx_1 \, dx_2 \\
&\quad + \gamma^2 \Omega \int \int x_2 \Pi(x_2, t_2 | x_1, t_1) \varphi^2 \Pi(x_1) \, dx_1 \, dx_2 \\
&= \gamma^2 \Omega^2 \varphi^4 + 2\gamma^2 \Omega^{3/2} \varphi^3 \langle x \rangle^{st} + \gamma^2 \Omega \varphi^2 \langle x^2 \rangle^{st} \\
&\quad + 2\gamma^2 \Omega^{3/2} \varphi^3 \int \int x_2 \Pi(x_2, t_2 | x_1, t_1) \Pi(x_1) \, dx_1 \, dx_2 \\
&\quad + 4\gamma^2 \Omega \varphi^2 \int \int x_2 \Pi(x_2, t_2 | x_1, t_1) x_1 \Pi(x_1) \, dx_1 \\
&\quad + \gamma^2 \Omega \varphi^2 \langle x^2 \rangle^{st}
\end{aligned}
\end{equation}

The integral on the third line is given to sufficient approximation by (7.16); we set \( t_2 - t_1 = 2\tau \sqrt{\beta \gamma} \). The integral on the second line can be split into two parts. The first is with the aid of (12.4):

\[
\int \int x_2 \Pi(x_2, t_2 | x_1, t_1) \Pi(x_1) \, dx_1 \, dx_2 = \langle x \rangle^{st} = -\frac{1}{2} \Omega^{-1/2}
\]

The second part is to sufficient approximation:

\[
\int x_2 \Pi(x_2, t_2 | -\Omega^{-1/2}, t_1) \int \Pi(x_1) \, dx_1 \, dx_2 = -\Omega^{-1/2} \exp \left[-2\sqrt{\beta \gamma}(t_2 - t_1)\right]
\]

Collecting these results one obtains surprisingly

\begin{equation}
\begin{aligned}
f_2(t_1, t_2) &= \gamma^2 \Omega^2 \varphi^4 = \beta^2 \Omega^2
\end{aligned}
\end{equation}

Hence \( g_2 = 0 \); the recombination events are uncorrelated! It is not clear (to me) whether this could have been predicted on \textit{a priori} grounds.

\textbf{XIII. EXAMPLE OF A MULTIVARIATE MASTER EQUATION}

As a simple example in which two variables \( n, m \) occur, consider the following modification of the Malthus–Verhulst equation. A population consists of \( m \) males and \( n \) females. They have the same probability per unit time to die, both due to natural death and to the competition with the
surrounding population density \((n + m)/\Omega\). Their birth probability is for both sexes proportional with the number \(n\) of females.

\[
\begin{align*}
\dot{n} &= \beta n - \alpha n - \gamma m(n + m)/\Omega \\
\dot{m} &= \beta n - \alpha m - \gamma m(n + m)/\Omega
\end{align*}
\]  
(13.1a)

(13.1b)

It is possible, but unnecessarily cumbersome, to use different values for \(\alpha, \beta, \gamma\) in both equations.

The master equation for the joint distribution \(P(n, m, t)\) is

\[
\dot{P} = \beta(E_n^{-1} - 1)nP + \alpha(E_n - 1)nP + \frac{\gamma}{\Omega}(E_n - 1)n(n + m)P
\]

\[
+ \beta(E_m^{-1} - 1)nP + \alpha(E_m - 1)mP + \frac{\gamma}{\Omega}(E_m - 1)m(n + m)P
\]  
(13.2)

\(E_n\) and \(E_m\) are difference operators \((2.9)\) acting on \(n\) and \(m\), respectively. The ready-made equations derived in Sections IV and V cannot be used, but it is easy to adapt the \(\Omega\) expansion to this multivariate case.

Apply again the information \((3.3)\) combined with

\[
m = \Omega \psi(t) + \Omega^{1/2} y
\]

The new distribution \(\Pi(x, y, t)\) then obeys to order \(\Omega^0\)

\[
\frac{\partial \Pi}{\partial t} - \Omega^{1/2} \frac{d\phi}{dt} \frac{\partial \Pi}{\partial x} - \Omega^{1/2} \frac{d\psi}{dt} \frac{\partial \Pi}{\partial y}
\]

\[
= \beta \Omega \left( -\Omega^{-1/2} \frac{\partial}{\partial x} + \frac{1}{2} \Omega^{-1} \frac{\partial^2}{\partial x^2} \right) (\varphi + \Omega^{-1/2} x) \Pi
\]

\[
+ \alpha \Omega \left( \Omega^{-1/2} \frac{\partial}{\partial x} + \frac{1}{2} \Omega^{-1} \frac{\partial^2}{\partial x^2} \right) (\varphi + \Omega^{-1/2} x) \Pi
\]

\[
+ \gamma \Omega \left( \Omega^{-1/2} \frac{\partial}{\partial y} + \frac{1}{2} \Omega^{-1} \frac{\partial^2}{\partial y^2} \right)
\times (\varphi + \Omega^{-1/2} x)(\psi + \Omega^{-1/2} x + \Omega^{-1/2} y) \Pi
\]

\[
+ \beta \Omega \left( -\Omega^{-1/2} \frac{\partial}{\partial y} + \frac{1}{2} \Omega^{-1} \frac{\partial^2}{\partial y^2} \right) (\psi + \Omega^{-1/2} y) \Pi
\]

\[
+ \alpha \Omega \left( \Omega^{-1/2} \frac{\partial}{\partial y} + \frac{1}{2} \Omega^{-1} \frac{\partial^2}{\partial y^2} \right) (\psi + \Omega^{-1/2} y) \Pi
\]

\[
+ \gamma \Omega \left( \Omega^{-1/2} \frac{\partial}{\partial y} + \frac{1}{2} \Omega^{-1} \frac{\partial^2}{\partial y^2} \right)
\times (\psi + \Omega^{-1/2} y)(\varphi + \psi + \Omega^{-1/2} x + \Omega^{-1/2} y) \Pi
\]  
(13.3)
THE EXPANSION OF THE MASTER EQUATION

All terms of order $\Omega^{1/2}$ are either proportional to $\partial \Pi / \partial x$ or to $\partial \Pi / \partial y$. The coefficient of each of these derivatives vanishes if

$$- \frac{d \phi}{dt} = - \beta \phi + \alpha \phi + \gamma \eta \phi + \gamma \psi (\phi + \psi)$$  \hspace{1cm} (13.4a)

$$- \frac{d \psi}{dt} = - \beta \phi + \alpha \psi + \gamma \psi (\phi + \psi)$$  \hspace{1cm} (13.4b)

These are again the macroscopic rate equations (13.1). They can be solved explicitly; the result is sketched in Fig. 6. For our purpose it is sufficient to notice that, supposing $\beta > \alpha$, there is one stable stationary point

$$\phi^* = \psi^* = \frac{\beta - \alpha}{2 \gamma}$$  \hspace{1cm} (13.5)

The terms of order $\Omega^0$ yield a bivariate linear Fokker–Planck equation with time-dependent coefficients for $\Pi$

$$\frac{\partial \Pi}{\partial t} = \frac{\partial}{\partial x} \left\{ - \beta x + \alpha x + \gamma (\phi + \psi)x + \gamma \eta (x + y) \right\} \Pi$$

$$+ \frac{\partial}{\partial y} \left\{ - \beta x + \alpha x + \gamma (\phi + \psi)y + \gamma \psi (x + y) \right\} \Pi$$

$$+ \frac{1}{2} \left\{ \beta \phi + \alpha \phi + \gamma \eta (\phi + \psi) \right\} \frac{\partial^2 \Pi}{\partial x^2}$$

$$+ \frac{1}{2} \left\{ \beta \phi + \alpha \psi + \gamma \psi (\phi + \psi) \right\} \frac{\partial^2 \Pi}{\partial y^2}$$  \hspace{1cm} (13.6)

The general solution of such equations will be given in the next section.

Fig. 6. Hodogram of the solutions of the equations (13.4).
Here we shall only study the fluctuations in the stationary state. Substi-
tution of (13.5) reduces (13.6) to
\[
\frac{\partial \Pi}{\partial t} = \frac{1}{2} (\beta - \alpha) \frac{\partial}{\partial x} (x + y) \Pi + \frac{\partial}{\partial y} \left\{ -\frac{1}{2} (\beta + \alpha) x + \left( \frac{3}{2} \beta - \frac{1}{2} \alpha \right) y \right\} \Pi
\]
\[+ \frac{\beta}{2\gamma} (\beta - \alpha) \left( \frac{\partial^2 \Pi}{\partial x^2} + \frac{\partial^2 \Pi}{\partial y^2} \right) \]
(13.7)
The resulting equations for the first moments are
\[
\frac{d}{dt} \langle x \rangle = -\frac{1}{2} (\beta - \alpha) (\langle x \rangle + \langle y \rangle) \quad (13.8a)
\]
\[
\frac{d}{dt} \langle y \rangle = \frac{1}{2} (\beta + \alpha) \langle x \rangle - \left( \frac{3}{2} \beta - \frac{1}{2} \alpha \right) \langle y \rangle \quad (13.8b)
\]
The eigenvalues of the coefficient matrix are $-\beta$ and $-\beta + \alpha$, which confirms that the stationary solution (13.5) is asymptotically stable. The equations for the second moments are
\[
\frac{d}{dt} \langle x^2 \rangle = -(\beta - \alpha) (\langle x^2 \rangle + \langle xy \rangle) + \frac{\beta}{\gamma} (\beta - \alpha) \quad (13.9a)
\]
\[
\frac{d}{dt} \langle xy \rangle = \frac{1}{2} (\beta + \alpha) \langle x^2 \rangle + (2\beta - \alpha) \langle xy \rangle - \frac{1}{2} (\beta - \alpha) \langle y^2 \rangle \quad (13.9b)
\]
\[
\frac{d}{dt} \langle y^2 \rangle = (\beta + \alpha) \langle xy \rangle - (3\beta - \alpha) \langle y^2 \rangle + \frac{\beta}{\gamma} (\beta - \alpha) \quad (13.9c)
\]
They can be used for instance to find the equilibrium fluctuations
\[
\langle x^2 \rangle = \frac{7\beta^2 - 6\beta \alpha + \alpha^2}{4\gamma(2\beta - \alpha)} \quad (13.10a)
\]
\[
\langle xy \rangle = \frac{\beta^2 + 2\beta \alpha - \alpha^2}{4\gamma(2\beta - \alpha)} \quad (13.10b)
\]
\[
\langle y^2 \rangle = \frac{3\beta^2 - 2\beta \alpha + \alpha^2}{4\gamma(2\beta - \alpha)} \quad (13.10c)
\]
This result, together with the solutions of (13.8) permits us to determine the
two-time correlations; they will involve both exponential factors $e^{-\alpha t}$ and $e^{-(\beta - \alpha)t}$. 
XIV. SOLUTION OF THE MULTIVARIATE FOKKER–PLANCK EQUATION

The general linear Fokker–Planck equation with $K$ variables and time-dependent coefficients has the form

$$\frac{\partial \Pi}{\partial t} = - \sum_{i,j} \frac{\partial}{\partial x_i} A_{ij}(t)x_j \Pi + \frac{1}{2} \sum_{i,j} B_{ij}(t) \frac{\partial^2 \Pi}{\partial x_i \partial x_j} \quad (14.1)$$

The matrix $B$ is symmetric but in general $A$ is not. The first moments obey the equations

$$\frac{d}{dt} \langle x_i \rangle = \sum_j A_{ij}(t) \langle x_j \rangle \quad (14.2)$$

It is convenient to define an evolution matrix $V(t|t')$ by

$$\frac{dV(t|t')}{dt} = A(t)V(t|t') \quad V(t'|t') = 1 \quad (14.3)$$

Then the solution of (14.2) with given $\langle x_i \rangle$ at $t = 0$ is

$$\langle x_i \rangle_t = \sum_j V_{ij}(t|0) \langle x_j \rangle_0 \quad (14.4)$$

The second moments obey the set of equations

$$\frac{d}{dt} \langle x_i x_j \rangle = \sum_k A_{ik} \langle x_k x_j \rangle + \sum_k A_{jk} \langle x_i x_k \rangle + B_{ij} \quad (14.5)$$

Introducing the matrix $X_{ij}(t) = \langle x_i x_j \rangle$ one may write this equation in matrix notation:

$$\dot{X} = AX + X\tilde{A} + B \quad (14.6)$$

where $\tilde{A}$ is the transpose of $A$. Note that by construction $X$ must be symmetric and positive definite (or at least semidefinite). The problem is to find such an $X$ obeying (14.6) with arbitrary (symmetric, positive definite) initial value.

The resemblance of (14.6) with the Neumann equation for a quantum-mechanical density matrix suggests that one transforms $X$ by putting

$$X(t) = V(t|0)Y(t)V(t|0) \quad (14.7)$$

One then finds after some algebra

$$\dot{Y}(t) = V(t|0)^{-1}(\dot{X} - AX - X\tilde{A})\tilde{V}(t|0)^{-1}$$
$$\quad = V(t|0)^{-1}B(t)\tilde{V}(t|0)^{-1} \quad (14.8)$$
Hence

\[ Y(t) = X(0) + \int_0^t V(t'\mid 0)^{-1} B(t') \tilde{V}(t'\mid 0)^{-1} \, dt' \]

Thus we have found the solution of (14.6)

\[ X(t) = V(t\mid 0)X(0)\tilde{V}(t\mid 0) + \int_0^t V(t\mid t')B(t')\tilde{V}(t\mid t') \, dt' \]  

(14.9)

Clearly it is symmetric and positive definite when \( X(0) \) has these properties and when \( B(t) \) is positive definite. Thus a condition for our solution of (14.1) is that \( B \) is positive definite or at least semidefinite at all \( t \). What goes wrong when that condition is violated is exemplified by

\[ \frac{\partial \Pi(x, t)}{\partial t} = - \frac{\partial^2 \Pi(x, t)}{\partial x^2} \]

This amounts to solving the heat conduction equation towards negative times, which is not a correctly set problem in the sense of Hadamard.25

Finally we have to find the solution of (14.1). The transformation (14.7) amounts to introducing new variables,

\[ x_i = \sum_j V_{ij}(t\mid 0)y_j \]  

(14.10)

We transform the whole equation (14.1) to these new variables. The probability distribution of \( y \) is

\[ \Xi(y, t) = \Pi(x, t) \text{Det} \, V \]  

(14.11)

It is not necessary to carry out the transformation in detail, because we know beforehand that \( \Xi \) obeys an equation of the same type (14.1) and that the first-order derivatives vanish since the \( \langle y_i \rangle \) are constant by construction. Hence the equation for \( \Xi \) must have the form

\[ \frac{\partial \Xi}{\partial t} = \frac{1}{2} \sum_{i,j} C_{ij}(t) \frac{\partial^2 \Xi}{\partial y_i \partial y_j} \]  

(14.12)

The second-order derivatives can only stem from those in (14.1) and one must therefore have

\[ \sum_{ij} C_{ij} V_{ki} V_{ij} = B_{kl} \quad \text{or} \quad B = VC \tilde{V} \]  

(14.13)

Note that \( C \) is identical with the matrix in (14.8) so that

\[ \dot{Y} = C \]  

(14.14)
The form of (14.12) suggests that $\Xi$ is a Gaussian

$$
\Xi(y, t) = (2\pi)^{-1/2K}(\det R)^{-1/2} \exp \left[ -\frac{1}{2} \sum_{k, l} R_{kl}(t)(y_k - c_k)(y_l - c_l) \right] \quad (14.15)
$$

If that is so one knows that $R^{-1}$ is the correlation matrix of the $y$'s and hence identical with $Y$. To verify that (14.15) is a solution we substitute in (14.12)

$$
-\frac{1}{2} \frac{d}{dt} \log \det R = \frac{1}{2} \sum_{k, l} \dot{R}_{kl}(t)(y_k - c_k)(y_l - c_l)
$$

$$
= \frac{1}{2} \sum_{i, j} C_{ij} \left\{ \sum_{k, l} R_{ik}(y_k - c_k)R_{jl}(y_l - c_l) - R_{ij} \right\}
$$

For this to be an identity it is necessary that

$$
-\dot{R} = RCR
$$

$$
\frac{d}{dt} \log \det R = \text{Tr} CR
$$

The first one easily follows from (14.14). The second one is obtained by means of the Wronski identity:

$$
\det (Y + \dot{Y} dt) - \det Y = \det Y(\det(1 + Y^{-1}\dot{Y} dt) - 1)
$$

$$
= \det Y(\text{Tr} Y^{-1}\dot{Y}) dt
$$

This completes the solution of (14.1) or at least reduces it to solving (14.3). The final formula is obtained by restoring the variables $x_i$,

$$
\Pi(x, t) = (2\pi)^{-1/2K}(\det X)^{1/2} \exp \left[ -\frac{1}{2} \sum_{ij} (X^{-1})_{ij}(x_i - \langle x_i \rangle)(x_j - \langle x_j \rangle) \right] \quad (14.16)
$$

where $\langle x_i \rangle$ is obtained from (14.2) and $X(t)$ from (14.9). Of course this result can be checked by direct substitution in (14.1). In the case of a single variable ($K = 1$) the solution is the same as in the Appendix and in Ref. 4. In the case that $A$ and $B$ are independent of time the solution can be made more explicit as is shown in the next section. We have to add a remark here on stability.

Suppose $A(t)$ is such that all solutions of (14.2) tend to zero, strongly enough for the time integral of $\langle x_i \rangle$ to converge. According to (14.3) that implies that the operator $V(t | t')$ vanishes for $t \to \infty$ and fixed $t'$. Hence the

* An alternative way of solving (14.1) is by Fourier transformation, i.e., with the use of the characteristic function of $\Pi$.

† The precise condition is presumably that $\sum \langle x_i \rangle^2$ can be integrated from $t = 0$ to $t = \infty$. 
first term on the right of (14.9) tends to zero, which means that the correlations become independent of their initial values. Moreover, if $B(t)$ is bounded for $t \to \infty$, one may write for large $t$

$$X(t) = \int_0^\infty V(t|t - \tau)B(t - \tau)\tilde{V}(t|t - \tau) \, d\tau$$

(14.17)

Not only the initial value but also the initial time has disappeared from the formula; the fluctuations depend exclusively on the coefficient matrices of (14.1). This fact is of course taken for granted in many physical applications, but it should be borne in mind that it requires that all solutions of (14.2) tend to zero, which is tantamount to saying that the macroscopic solution is asymptotically stable. What happens when this is not so will be investigated in Section XVIII.

**XV. FOKKER–PLANCK EQUATION WITH CONSTANT COEFFICIENTS**

Take the same equation (14.1) but with $A$ and $B$ independent of $t$. The solution of (14.3) may then be written

$$V(t|t') = V(t - t') = e^{A(t-t')},$$

(15.1)

Hence the expression (14.9) for $X$ becomes

$$X(t) = e^{At}X(0)e^{At} + \int_0^t e^{A(t-t')}Be^{A(t-t')} \, dt'$$

(15.2)

It is clear that the stability conditions mentioned in the previous paragraph are obeyed if, and only if, each eigenvalue of $A$ has a negative real part. In that case $X(t)$ tends to the constant matrix

$$X(\infty) = \int_0^\infty e^{At}Be^{At} \, d\tau$$

(15.3)

This is a general formula for the stationary state fluctuations in the linear noise approximation, applicable to all systems that have a stable stationary state.

A method for actually evaluating (15.2), and hence also (15.3), is suggested by the following observation. If $A$ were symmetric one would perform an orthogonal transformation which diagonalizes $A$; as a result the matrix equation (15.2) would decompose in a set of uncoupled equations for the separate matrix elements of $X$ in this new representation. A similar approach is possible now, using a nonorthogonal transformation, owing to the
following fact. When $Q$ is a nonsingular matrix, (15.2) is invariant for the transformation

$$X' = QX, \quad B' = QB, \quad A' = QAQ^{-1}$$  \hspace{1cm} (15.4)

If a $Q$ can be found such that $A'$ is diagonal the problem is again solved.

Denote the eigenvalues of $A$ by $-\lambda_k$ and the corresponding left eigenvectors by $q_i^{(k)}$. There is no guarantee that the $q_i^{(k)}$ form a complete set of $K$ linearly independent vectors, but we assume that they do, because that suffices for our applications.* Then the diagonalizing matrix is $Q_{kj} = q_i^{(k)}$ whereas $Q^{-1}$ consists of the suitably normalized right eigenvectors $p_i^{(k)}$

$$(Q^{-1})_{ik} = p_i^{(k)}, \quad \sum_i q_i^{(k)} p_i^{(l)} = \delta_{kl}$$  \hspace{1cm} (15.4)

Thus we have the transformation formulas

$$A'_{kl} = \sum_{ij} q_i^{(k)} A_{ij} p_j^{(l)} = -\lambda_k \delta_{kl}$$  \hspace{1cm} (15.5)

$$X'_{kl} = \sum_{ij} q_i^{(k)} X_{ij} p_j^{(l)} = \tilde{q}^{(k)} \cdot X \cdot q^{(l)}$$  \hspace{1cm} (15.6)

In the new representation (15.2) reads

$$X'_{kl}(t) = X'_{kl}(0)e^{-(\lambda_k + \lambda_l)t} + \int_0^t e^{-\lambda_k(t-t')} B'_{kl} e^{-\lambda_l(t-t')} \, dt'$$  \hspace{1cm} (15.7)

The solution is simple:

$$X'_{kl}(t) = X'_{kl}(0)e^{-(\lambda_k + \lambda_l)t} + B'_{kl} \frac{1 - e^{-(\lambda_k + \lambda_l)t}}{\lambda_k + \lambda_l}$$  \hspace{1cm} (15.8)

When the denominator vanishes, the fraction is replaced by its limiting value, $t$. This cannot occur, however, when (15.1) is stable, since in that case all $\lambda_k$ have a positive real part and (15.8) tends to the limit

$$X_{kl}(\infty) = \frac{B_{kl}}{\lambda_k + \lambda_l}$$  \hspace{1cm} (15.9)

Finally one has to transform back to $X_{ij} \equiv \langle x_i x_j \rangle$. The result is an algebraic expression for the integral (15.3), or also a solution of (14.6) with the left-hand side zero.27 A special example of this result was obtained in (13.10).

* If they do not, $A$ can be transformed to the Jordan form, which makes the solution still possible but quite a bit more cumbersome.26
Chemical reactions provide a wide variety of applications. Consider a volume \( \Omega \) containing a mixture of different species of molecules, and suppose (i) that the mixture is sufficiently dilute to constitute an ideal gas, (ii) that it is homogeneously distributed* in \( \Omega \), and (iii) that the momenta are distributed according to Boltzmann with temperature \( T \). The concentrations of the various compounds, however, are not in thermal equilibrium and vary as a result of reactive collisions.

The simplest possible reaction is the transition between two isomers

\[
A + X \xrightarrow{\alpha} A + Y
\]

where \( A \) is a fixed catalyst needed for the collisions. The total number of molecules \( X \) and \( Y \) is a constant \( N \). If \( n \) is the number of \( X \), the number of reactive collisions from left to right is proportional with \( n \) and the fixed concentration of \( A \). Hence the macroscopic rate equation has the form

\[
\frac{dn}{dt} = -\alpha n + \beta (N - n)
\]

(16.2)

The transition probability per unit time is

\[
W(n|n') = \alpha n' \delta_{n,n'-1} + \beta (N - n') \delta_{n,n'+1}
\]

(16.3)

The master equation is therefore

\[
P(n, t) = \alpha (E - 1)nP + \beta (E^{-1} - 1)(N - n)P
\]

(16.4)

Note that there is a natural boundary at \( n = 0 \) (depletion of \( X \)) and another one at \( n = N \) (depletion of \( Y \)). This master equation is linear and can readily be solved by means of a generating function. Rather than writing the full solution we only note that according to (6.4) and (6.5)

\[
P^m(n) = (\alpha + \beta)^{-N} \binom{N}{n} \alpha^{N-n} \beta^n
\]

(16.5)

This is identical with the thermal equilibrium provided that

\[
\beta/\alpha = \exp (\sigma_x - \sigma_y)
\]

(16.6)

where \( \sigma_x, \sigma_y \) are the internal entropies of \( X \) and \( Y \).

The mixture of \( X \) and \( Y \) in the volume \( \Omega \) constituted a closed system since no molecules could enter or escape. In order to extend the theory to open systems one supposes that it is possible to inject molecules at a constant rate,

* Reactions that are not homogeneous in space are of considerable interest but will not be studied in this work.
or to extract them at a rate proportional to their number. This adds two new terms to the rate equation for \( n \),

\[
\frac{dn}{dt} = b - an + \cdots \quad (16.7)
\]

They correspond to two additional terms in the transition probability

\[
W(n|n') = b\delta_{n,n+1} + an'\delta_{n,n'-1} + \cdots \quad (16.8)
\]

In the master equation they give rise to the terms

\[
\dot{P}(n, t) = b(E^{-1} - 1)P + a(E - 1)nP + \cdots \quad (16.9)
\]

Of course it is understood that the injection and extraction do not violate assumptions (i), (ii), and (iii).

Another way of arriving at these equations is by taking a closed system which includes a reservoir of molecules that produce or absorb \( X \). To show this consider the reaction (16.1) in the limit \( N \to \infty, \beta \to 0 \) with constant \( \beta N = b \). Then (16.2) tends to (16.7), (16.3) to (16.8) and (16.4) to (16.9). Thus the molecules \( Y \) serve as a reservoir which creates, and annihilates molecules \( X \) according to the same equations as used for describing injection and extraction. The \( Y \) do not otherwise enter the picture and may be ignored. The remaining species \( X \) alone constitutes an open system, although \( X \) and \( Y \) together were a closed system.

This way of creating and annihilating \( X \) is not uniquely determined by the macroscopic equation. For instance the reaction

\[
Y \xrightarrow{\frac{1}{2} \beta} X + X
\]

would produce the same term \( b \) in (16.7) but not the same terms in (16.8) and (16.9). For this reason one usually includes in the reaction scheme the reactions with the reservoir molecules; they are indicated by the earlier letters of the alphabet \( A, B, \ldots \) and the numbers \( n_A, n_B, \ldots \) are then taken constant.

The Malthus–Verhulst problem can be interpreted as the following chemical reaction in an open system:

\[
X \xrightarrow{k_1} A
\]

\[
B + X \xrightarrow{k_2 \frac{k_1}{k_2}} X + X \quad (16.10)
\]

The amounts of \( A \) and \( B \) are supposed fixed, so that the only variable is the number \( n \) of molecules \( X \). The first line simulates the natural death: \( k_1 = \alpha \). The reverse process \( A \to X \) is supposed to be negligible, for instance because \( A \) is drained or undergoes further decays. The second line is an autocatalytic process. The arrow to the right simulates natural births proportional to the
existing population, \( k_2 n_B / \Omega = \beta \). The reverse process corresponds to the Verhulst term: \( k'_2 = \gamma \). This process was studied by Schlögl.\(^{30}\)

A reaction in a closed system having the same rate equation but a different master equation is\(^{34}\)

\[
X + Y \xrightarrow{k} 2X \tag{16.11}
\]

The concentrations of both compounds vary in time, but there is a conserved quantity \( n_X + n_Y = C \). Hence there is a single rate equation for \( n_X = n \),

\[
\frac{dn}{dt} = \frac{k}{\Omega} n(C - n) - \frac{k'}{\Omega} n^2 = \frac{kC}{\Omega} n - \frac{k + k'}{\Omega} n^2 \tag{16.12}
\]

\( \Omega \) is the volume in which the reacting mixture is enclosed. We have written \( n^2 \) rather than \( n(n - 1) \) in agreement with the remark in Section IX that the difference is negligible on the macroscopic scale. The equation is identical with the Malthus–Verhulst equation (9.1) with the identification

\[
kC/\Omega = \beta - \alpha, \quad k + k' = \gamma
\]

However, the master equation for (16.11) is

\[
\dot{P}(n, t) = \frac{k}{\Omega} (\mathbf{E}^{-1} - 1)n(C - n)P + \frac{k'}{\Omega} (\mathbf{E} - 1)n(n - 1)P \tag{16.13}
\]

Obviously this differs from (9.4) and we may expect that the fluctuations are different. In fact one finds

\[
\frac{\partial \Pi}{\partial t} = \{ -kc + 2(k - k')\varphi \} \frac{\partial}{\partial x} x\Pi + \frac{1}{2} \{ kc\varphi + (k' - k)\varphi^2 \} \frac{\partial^2 \Pi}{\partial x^2} \tag{16.14}
\]

with \( c = C/\Omega \). This is manifestly different from (9.5). The variance of the fluctuations in the stationary state is

\[
\sigma^2_n = \Omega \langle x^2 \rangle = \frac{k'kc}{(k' + k)(3k' - k)} \tag{16.15}
\]

which cannot even be expressed in \( \alpha, \beta, \gamma \).

**XVII. TWO-STEP CHEMICAL REACTIONS**

Consider the reaction scheme\(^{31,35}\)

\[
A \xrightarrow{k^{(1)}} X \xrightarrow{k^{(2)}} B \tag{17.1}
\]
The amounts of A and B are fixed and the reverse reactions are supposed to be negligible. The rate equation for the number of molecules X is

\[ \frac{dn}{dt} = k^{(1)}n_A - 2k^{(2)} \frac{n^2}{\Omega} \]  

(17.2)

This is the same as for the semiconductor, see (7.1) if

\[ k^{(1)}n_A/\Omega = \beta, \quad 2k^{(2)} = \gamma \]  

(17.3)

The master equation, however, has to take into account that the X can only disappear in pairs,

\[ \dot{\Pi}(n, t) = k^{(1)}n_A(E^{-1} - 1)P + \frac{k^{(2)}}{\Omega} (E^2 - 1)n(n - 1)P \]  

(17.4)

(Of course the number of pairs is \( \frac{1}{2}n(n - 1) \), but this \( \frac{1}{2} \) has been incorporated in the definition of \( k^{(2)} \).)

Expanding as usual and utilizing (17.3) as convenient abbreviations we get

\[ \frac{\partial \Pi}{\partial t} + \Omega^{1/2} \frac{d\varphi}{dt} \frac{\partial \Pi}{\partial x} = \Omega \beta \left( -\Omega^{-1/2} \frac{\partial}{\partial x} + \frac{1}{2} \Omega^{-1} \frac{\partial^2}{\partial x^2} \right) \Pi \]
\[ + \frac{1}{2} \gamma \Omega \left( 2\Omega^{-1/2} \frac{\partial}{\partial x} + 2\Omega^{-1} \frac{\partial^2}{\partial x^2} \right) \]
\[ \times (\varphi + \Omega^{-1/2}x)(\varphi + \Omega^{-1/2}x - \Omega^{-1}) \Pi \]  

(17.5)

The terms of order \( \Omega^{1/2} \) reproduce the macroscopic equation and those of order \( \Omega^0 \) yield

\[ \frac{\partial \Pi}{\partial t} = 2\gamma \varphi \frac{\partial}{\partial x} x\Pi + \left( \frac{1}{2} \beta + \gamma \varphi^2 \right) \frac{\partial^2 \Pi}{\partial x^2} \]  

(17.6)

Compare this width (7.10). In particular, to find the variance in the stationary state we substitute \( \varphi^* = \sqrt{\beta/\gamma} \) and obtain

\[ \sigma_n^2 = \Omega \langle x^2 \rangle^* = \frac{3}{4} \Omega \sqrt{\frac{\beta}{\gamma}} = \frac{3}{4} \langle n \rangle^* \]  

(17.7)

The factor 3/4 shows that the stationary distribution is narrower than a Poisson distribution. This fact has caused some misgivings about the correctness, but is actually easy to understand. The Poisson distribution is expected for independent particles, but the X molecules are not independent as they disappear in pairs. If at any time their number is slightly higher than the average they disappear more rapidly not only because there are more, but also because the probability for each molecule to disappear is enhanced.
by the higher number of others available to form a pair. A similar argument
applies when their number happens to be lower than average. Hence it was
to be expected that the distribution should be narrower than for molecules
that appear and disappear independently from each other.

On the other hand, when the molecules are created in pairs one expects that
the stationary distribution will be broader than Poisson. As an example
consider the reaction scheme

\[ \begin{align*}
A \xrightarrow{k^{(1)}} & \ 2X \\
X \xrightarrow{k^{(2)}} & \ B
\end{align*} \tag{17.8} \]

\( n_A \) and \( n_B \) are again fixed, and the master equation is with the abbreviations
\( k^{(1)} n_A / \Omega = \alpha \) and \( k^{(2)} = \beta \)

\[ \dot{P}(n, t) = \alpha \Omega (\mathbf{E}^{-2} - 1)P + \beta (\mathbf{E} - 1)nP \tag{17.9} \]

The expansion in \( \Omega \) yields

\[ \frac{\partial \Pi}{\partial t} - \Omega^{1/2} \frac{d\varphi}{dt} \frac{\partial \Pi}{\partial \varphi} = \alpha \Omega \left( -2 \Omega^{-1/2} \frac{\partial}{\partial \varphi} + 2 \Omega^{-1} \frac{\partial^2}{\partial \varphi^2} \right) \Pi \\
+ \beta \Omega \left( \Omega^{-1/2} \frac{\partial}{\partial \varphi} + \frac{1}{2} \Omega^{-1} \frac{\partial^2}{\partial \varphi^2} \right) (\varphi + \Omega^{-1/2} \varphi) \Pi \tag{17.10} \]

The terms of order \( \Omega^{1/2} \) give the macroscopic equation

\[ \frac{d\varphi}{dt} = 2\alpha - \beta \varphi \tag{17.11} \]

The terms of order \( \Omega^0 \) yield

\[ \frac{\partial \Pi}{\partial t} = \beta \frac{\partial}{\partial \varphi} \varphi \Pi + \left( 2\alpha + \frac{1}{2} \beta \varphi \right) \frac{\partial^2 \Pi}{\partial \varphi^2} \tag{17.12} \]

It is again easy to find \( \varphi_{\text{eq}} = 2\alpha / \beta \) and subsequently

\[ \sigma_n^2 = \Omega \langle x^2 \rangle_{\text{eq}} = \frac{3 \Omega \alpha}{\beta} = \frac{3}{2} \langle n \rangle_{\text{eq}} \tag{17.13} \]

This confirms the expectation.*

The reader will have no difficulty in treating the case that \( p \) molecules are
created simultaneously, while \( q \) are annihilated simultaneously. The master
equation is

\[ \dot{P} = \alpha \Omega (\mathbf{E}^{-p} - 1)P + \beta \Omega^{-q+1} (\mathbf{E}^q - 1)(n)_q P \tag{17.14} \]

* Actually the master equation (17.9) happens to be linear in \( n \) and can therefore be solved
exactly.
Here \((n)_q\) is an abbreviation (Pochhammer's symbol) for
\[
(n)_q = n(n - 1)(n - 2) \cdots (n - q + 1)
\]  
but in our approximation no error is committed on replacing it with \(n^q\). The result for the stationary distribution is
\[
\sigma_n^2 = \frac{p + q}{2q} \langle n \rangle^{st}
\]
In particular for \(p = q\) one recovers the Poisson-like result. In fact one can verify by direct substitution in (17.14) that the Poisson distribution with \(\langle n \rangle = \Omega(x/\beta)^p\) is an exact solution.

**XVIII. FLUCTUATIONS ABOUT A LIMIT CYCLE**

As an example we take a reaction scheme whose macroscopic behavior has been analyzed by Glansdorff and Prigogine,\textsuperscript{28} Nicolis,\textsuperscript{36} and others\textsuperscript{37,38}

\[
\begin{align*}
\text{A} & \quad \longrightarrow \quad \text{X} \\
2\text{X} + \text{Y} & \quad \longrightarrow \quad 3\text{X} \\
\text{B} + \text{X} & \quad \longrightarrow \quad \text{Y} + \text{D} \\
\text{X} & \quad \longrightarrow \quad \text{E}
\end{align*}
\]  
(18.1)

There are two variables \(n_X\) and \(n_Y\); they transport matter from the reservoir A into E and from B into D. These flows are supposed to be sufficiently strong for the reverse reactions to be negligible. With a suitable choice of units of \(t\) and \(\Omega\) the master equation takes the form
\[
\dot{P}(n_X, n_Y, t) = \Omega x (E_X^{-1} - 1)P + \Omega^{-2}(E_X^{-1}E_Y - 1)n_X^2 n_Y P + (E_X^{-1} - 1)n_X P + \beta(E_X E_Y^{-1} - 1)P
\]  
(18.2)

In order to expand in \(\Omega\) we set
\[
n_X = \Omega \phi(t) + \Omega^{1/2} x, \quad n_Y = \Omega \psi(t) + \Omega^{1/2} y
\]  
(18.3)

The macroscopic equations so obtained are
\[
\begin{align*}
\phi &= x + \phi^2 \psi - \beta \psi - \phi \\
\psi &= \beta \phi - \phi^2 \psi
\end{align*}
\]  
(18.4a, 18.4b)

A careful study of these equations\textsuperscript{36,37} shows that the solutions do not tend to a stationary point but to a limit cycle (see Fig. 7). Thus there is a periodic

* Note added in proof. After this article was submitted, a paper by K. Tomita, T. Ohta, and H. Tomita appeared [Progr. Theor. Phys., 52, 1744, (1974)], which covers the same ground as our Section XVIII.
solution with period $T$, or rather a family of such solutions because the phase is arbitrary. All other solutions tend to one of these as $t \to \infty$. We propose to investigate the fluctuations about this periodic solution of the macroscopic equations.

The terms of $\Omega^0$ in the expansion of (18.2) yield a Fokker–Planck equation for $\Pi$, which is of the form (14.1) with

$$A(t) = \begin{pmatrix} 2\phi\psi - 1 - \beta & \phi^2 \\ -2\phi\psi + \beta & -\phi^2 \end{pmatrix} \quad (18.5)$$

$$B(t) = \begin{pmatrix} \phi^2\psi + \beta\phi + \phi + \alpha & -\phi^2\psi - \beta\phi \\ -\phi^2\psi - \beta\phi & \phi^2\psi + \beta\phi \end{pmatrix} \quad (18.6)$$

According to the general scheme of Section XIV one obtains the correlation matrix $X(t)$ from (14.9) after solving the linear equation (14.3) for $V(t|t')$. The condition that $B$ should be positive definite is duly satisfied. The behavior for $t \to \infty$, however, will be different from the behavior in the case of a limit point.

Suppose that the system has evolved long enough, so that $(\phi, \psi)$ has approached the limit cycle within a distance of order $\Omega^{-1/2}$. Thus we have to substitute in $A(t)$ and $B(t)$ the periodic solution with some value of the phase. It follows that $A(t)$ is periodic with period $T$, rather than constant. This fact by itself does not exclude the possibility that the solution $V(t|t')$ of (14.3) tends to zero but we shall show that in fact it does not.

It is convenient to write $\phi_1, \phi_2$ instead of $\phi, \psi$ and to employ the symbol $\phi$ for this two-component vector. The macroscopic equations (18.4) may then be written

$$\dot{\phi}_i(t) = f_i(\phi(t)) \quad (18.7)$$
Of course it is also true that
\[ \phi_i(t + \delta t) = f_i(\phi(t + \delta t)) \]  
(18.8)
and therefore
\[ \dot{\phi}_i(t) = \sum_j \frac{\partial f_j(\phi)}{\partial \phi_j} \dot{\phi}_j(t) \]  
(18.9)

As mentioned in Section V the matrix of the derivatives is just \( A_{ij} \). Hence we have found one solution of (14.2), namely, \( \dot{\phi}_i(t) \). Now substitute for \( \phi \) a periodic solution \( \phi^k \) with some prescribed phase. Then (18.9) shows that there is one solution of (14.2) that does not go to zero since it is periodic in \( t \). Hence the equation (14.17) for large \( t \) cannot be correct for fluctuations about \( \phi^k \).

In order to investigate the modifications needed to deal with this lack of stability we construct a new coordinate system for the neighborhood of the limit cycle. In each point \( P \) of the limit cycle a natural base vector \( \mathcal{g}^{(1)} \) is defined by the tangent (Fig. 8):
\[ \mathcal{g}_j^{(1)} = \dot{\phi}_j \]  
(18.10)
This equation may also serve to define the length of \( \mathcal{g}^{(1)} \), since \( ||\dot{\phi}_j|| \) has a positive lower bound along the cycle. By construction we have according to (18.9)
\[ \dot{\mathcal{g}}_i^{(1)} = \sum_j A_{ij} \mathcal{g}_j^{(1)} \]  
(18.11)

Next a suitable direction for the other base vector \( \mathcal{g}^{(2)} \) has to be found. From each point \( Q \) in the vicinity of \( P \), but not on the cycle itself, a solution curve starts out that approaches the cycle. Compare this solution with the one that starts out from \( P \) at the same time. If the direction \( PQ \) is almost

Fig. 8. The new base vectors along the limit cycle.
parallel to \( g^{(1)} \) the former will end up by being slightly ahead in time with respect to the latter. If \( PQ \) is almost antiparallel to \( g^{(1)} \) it will end up by being behind. In between there is a direction where both solutions will ultimately coincide, not merely their orbits. We take \( g^{(2)} \) in this direction and choose some arbitrary length for it. Then we know

\[
\varphi(t|P) - \varphi(t|P + \epsilon g^{(2)}) \to 0 \quad (18.12)
\]

where the notation introduced below (4.9) is employed.

The variables \( x_j \), which describe the fluctuations about the macroscopic value \( \varphi_j^p(t) \), are now transformed by expressing them as linear combinations of the base vectors \( g^{(a)} \) attached to the point \( P = \varphi_j^h(t) \),

\[
x_k = \sum_{a=1}^{2} x_a' g^{(a)}_j = \sum_a \Theta_{ja} x_a' \quad (18.13)
\]

The moments transform accordingly,

\[
\langle x_j \rangle = \sum_a \Theta_{ja} \langle x_a' \rangle \quad X = \Theta X' \Theta \quad (18.14)
\]

In the primed variables (14.2) becomes

\[
\frac{d}{dt} \langle x_a' \rangle = \sum_\beta A'_{a\beta}(t) \langle x_\beta' \rangle \quad (18.15)
\]

where the matrix \( A' \) is

\[
A' = \Theta^{-1} A \Theta - \Theta^{-1} \dot{\Theta} \quad (18.16)
\]

The solution of (18.15) may again be expressed in an evolution matrix \( V'(t|t') \), which is easily seen to be

\[
V'(t|t') = \Theta(t)^{-1} V(t|t') \Theta(t') \quad (18.17)
\]

Owing to the choice of the new base, \( A' \) and \( V' \) have special properties. The primed components of \( g^{(1)} \) are simply \((1,0)\); hence (18.10) translates into \( 0 = A'(t)(1,0) \). This says that \((1,0)\) is invariant in time and therefore \( V'(t|t')(1,0) = (1,0) \), so that \( V' \) must have the form

\[
V'(t|t') = \begin{pmatrix} 1 & V'_{12} \\ 0 & V'_{22} \end{pmatrix} \quad (18.18)
\]

Moreover, according to (18.12) one has \( V'(t|t')(0,1) \to 0 \) so that

\[
\lim_{t \to \infty} V'_{12}(t|t') = \lim_{t \to \infty} V'_{22}(t|t') = 0 \quad (18.19)
\]

How fast these elements go to zero depends on how fast the solutions of the macroscopic equations tend to the limit cycle; we shall assume here that they are integrable.
In the new variables equation (14.9) for the second moments takes the form

$$X'(t) = V'(t|0)X(0)V'(0) + \int_0^t V'(t|t')B'(t')V'(t|t') \, dt$$  \hspace{1cm} (18.20)

where we have defined $B'(t)$ by

$$B(t) = \Theta(t)B'(t)\tilde{\Theta}(t)$$  \hspace{1cm} (18.21)

As $t \to \infty$ one obtains for the (1, 1)-element

$$X_{11}'(t) = X_{11}'(0) + \int_0^t B_{11}'(t') \, dt \hspace{1cm} (18.22)$$

Thus the initial value does not disappear, but even a term that grows roughly linearly with $t$ appears, and some finite additional terms. That means that fluctuations directed along the limit cycle are not damped but grow roughly like the displacement of a Brownian particle. When $X_{11}'$ has become of order $\Omega$ the whole limit cycle is covered; the time needed for that is also of order $\Omega$.

The other matrix elements of $X(t)$ obey equations of the same type as (14.17) and therefore tend to finite values. This means that fluctuations away from the limit cycle do not grow.

Thus the probability distribution $P(n, t)$ for large $t$ spreads out over a narrow strip covering the limit cycle. As it is no longer a sharp peak, no position can be ascribed to it, and no macroscopic part $\Omega \Phi$ of $n$ exists. Hence no macroscopic equation of motion can be formulated that describes the behavior during times of order $\Omega$, because after such times the position on the limit cycle is no longer determined. The same effect is known in electrical engineering as "phase slip."

**XIX. THE RANDOM WALK PICTURE**

The following way of visualizing a given one-step process is suggestive but, as will be shown presently, somewhat misleading. Take a line with a coordinate $s$ on it and mark the points $s = nt$. Define a "potential energy" $U(s)$ by putting

$$P^s(n) = \text{const} \, e^{-U(n\epsilon)}$$  \hspace{1cm} (19.1)

The given one-step process may then be visualized as a random walk on the point set \{ne\} under influence of a force with potential $U(s)$; the factor $kT$ has been absorbed in $U$. Of course this model does not uniquely specify the
jump probabilities $r(n)$ and $g(n)$, but expresses them in terms of a single function $k(n),\)

\[ r(n) = k(n)e^{U(n)}, \quad g(n) = k(n + 1)e^{U(n)} \]  

(19.2)

For example, in the much studied diode circuit\(^{40,41}\) of Fig. 9, where $n$ is the number of excess electrons on the left condenser plate, one has $U = e^2n^2/2kTC$. When $n > 0$, the electrons that jump from left to right only face the potential threshold formed by the work function of the diode plate so that $r(n)$ is a constant given by Richardson’s formula. The electrons jumping from right to left, however, face both the work function and the potential difference; one then has

\[ r(n) = A, \quad g(n) = A \exp \left[ -\frac{e^2}{kTC} \left( n + \frac{1}{2} \right) \right] \]  

(19.3)

For $n < 0$ the roles of $r$ and $g$ are interchanged.

The function $U$ does not always have such a concrete meaning, but this analogy with a potential energy makes the picture suggestive. If it has a single minimum, as in the diode example, one sees how the particles jump back and forth with a preference for lower energies, and in so doing establish a one-humped $P^\ast$ (see Fig. 10a). Now suppose that $U$ has two minima (Fig. 10b). Then $P^\ast$ has one hump in the lower minimum, and an additional one in the other minimum only if it is as low or almost as low. If it is possible to alter the relative depths of the two minima by varying an external parameter, one may make the hump of $P^\ast$ jump from one minimum to the other, and so create a phase transition. If the two minima have equal depth, and one varies a second parameter in order to reduce the maximum between them,
eventually a critical point will be reached where both minima merge into a single flat minimum (Fig. 10c). The fluctuations in the flat minimum are exceptionally large, as computed in Section XXI. In cases with more than one variable a great variety of phenomena may be visualized in this way.\textsuperscript{10,41}

Of course all this is nothing but interpreting (19.1). In addition, however, the picture also demonstrates some well-known features that are not implied in the stationary distribution. A metastable state occurs when the lower minimum is gradually raised above the other one, because the probability has to be transferred across the potential maximum. The rate at which this happens is mainly determined by the density of the initial distribution at the maximum, which is roughly

\[
\exp \left[ - (U_{\text{max}} - U_{\text{min}}) \right]
\]

This may easily be less than the rate at which the external parameter varies. Landauer\textsuperscript{43} has pointed out that flip-flop circuits, tunnel diodes, and in fact all memory-storing elements are examples of this situation and only retain their memory thanks to the smallness of (19.4).

Another time-dependent phenomenon that can be visualized by the random walk picture is the critical slowing down. When the minimum of \( U \)
is flat the difference between the jump probabilities towards and away from the minimum is small, which makes the distribution slow in moving to its equilibrium.

It has already been tacitly assumed that \( \epsilon \) is small and that \( U \) can be expressed as a smooth function of a continuous variable \( s \). We now also set

\[
\begin{align*}
  r(n) = \rho(s), \quad g(n) = \gamma(s), \quad k(n) = \kappa(s), \quad \epsilon P(n, t) = p(s, t) 
\end{align*}
\]

(19.5)

Then the master equation (2.8) may be expanded in \( \epsilon \):

\[
\frac{\partial p(s)}{\partial t} = \epsilon \frac{\partial}{\partial s} \{ \rho(s) - \gamma(s) \} p + \frac{1}{2} \epsilon^2 \frac{\partial^2}{\partial s^2} \{ \rho(s) + \gamma(s) \} p + O(\epsilon^3) \tag{19.6}
\]

Using (19.2) and setting \( \epsilon^2 t = \tau \) one obtains the nonlinear Fokker–Planck equation:

\[
\frac{\partial p(s)}{\partial \tau} = -\frac{\partial}{\partial s} \kappa'(s)e^{U(s)}p + \frac{\partial^2}{\partial s^2} \kappa(s)e^{U(s)}p \tag{19.7a}
\]

Instead of \( \kappa \) one may also use the function \( \lambda(s) = \kappa e^{U} \) to specify the jump probabilities,

\[
\frac{\partial p(s)}{\partial \tau} = \frac{\partial}{\partial s} \left( \lambda U' - \lambda' \right)p + \frac{\partial^2 p}{\partial s^2} \tag{19.7b}
\]

Notice that the average of \( s \) obeys

\[
\frac{d}{d\tau} \langle s \rangle = \langle \kappa'(s)e^{U(s)} \rangle = -\langle \lambda(s)U'(s) \rangle + \langle \lambda'(s) \rangle \tag{19.8}
\]

The fact that this is not equal to \(-\langle U'(s) \rangle\), let alone \(-U'(\langle s \rangle)\), as one might expect from a macroscopic point of view, has given rise to much discussion.\(^{40,49}\)

This confusion is caused by the inherent weakness of the random walk model. Although the model is appropriate for describing an actual random walk in an external potential field,\(^{45}\) in general it does not apply to the fluctuations of a macroscopic quantity in a many-body system. The formal expansion in \( \epsilon \) is not an expansion in an actual physical quantity like \( 1/\Omega \). This is already apparent because for small \( \epsilon \) the width of the stationary distribution does not become small, since it is solely determined by \( U(s) \) according to (19.1). The correct expansion is obtained by first identifying \( \epsilon \) with \( 1/\Omega \) (so that \( s \) becomes the intensive variable corresponding to the extensive variable \( n \)), and then defining the function \( U(s) \) by

\[
P^n(n) = \exp \left[ -\frac{1}{\epsilon} U(s) \right] = \exp \left[ -\Omega U(s) \right] \tag{19.9}
\]
rather than by (19.1). For instance in the diode circuit\textsuperscript{40} the capacity \( C \) has the role of \( \Omega = 1/e, \ s = n/C = V/e \) represents the voltage, and

\[ P^\text{th}(n) = \text{const exp} \left[ -\frac{1}{e} \frac{e^2 s^2}{2kT} \right] \]

The reason why (19.9) is correct rather than (19.1) can be understood as follows. The mesoscopic description of a many-body system in terms of a master equation is based on the assumption that on a suitable coarse-grained level the state of the system can be described by a single variable \( n \) or \( s \) (or by a number of such variables, but much less than the number of microscopic variables). The other degrees of freedom merely cause the random jumps of this single variable, called the “order parameter.” The probability distribution (19.1) is the partition function for fixed \( s \). Hence \( U(s) \) is the free energy, which is of course proportional to \( \Omega \). To obtain a correct expansion in \( 1/\Omega \) one must display this factor explicitly as in (19.9).*

The modification (19.9) changes the expansion radically, undermines the validity of (19.6) and (19.7), and largely spoils the suggestive random walk picture, but it gives the results correctly. As an additional benefit we found that the \( \Omega \) expansion leads to a Fokker–Planck equation that can be solved generally, whereas (19.7) can be solved only in very few cases.

Kubo has applied the same idea to the time-dependent solutions of the master equation.\textsuperscript{10} He substituted

\[ P(n, t) = \exp [-\Omega U(s, t)] \]

and solved the resulting equation for \( U \) in successive powers of \( 1/\Omega \). The results were, of course, the same as those obtained by the \( \Omega \) expansion of \( P \) itself, as described in the present article.

**XX. PHASE TRANSITIONS**

The random walk picture is particularly attractive for illustrating phase transitions, as in Fig. 10b. In this section we describe a few examples. They have been amply discussed in the literature; our purpose is merely to emphasize the role of the parameter \( \Omega \).

The Weiss theory of ferromagnetism\textsuperscript{†} provides a simple model for a phase transition. The free energy per particle, at a fixed value \( s \) of the magnetization, is, in proper units,

\[ N\{\frac{1}{2} + s\} \log\left(\frac{1}{2} + s\right) + \left(\frac{1}{2} - s\right) \log\left(\frac{1}{2} - s\right) - 2Ks^2 - 2Hs \]  

(20.1)

* The new function \( U \) as it appears in (19.9) may involve higher orders in \( 1/\Omega \), but we shall ignore this complication here.

† Also called Bragg–Williams approximation (see Ref. 46).
$N$ is the number of dipoles and serves as size parameter $\Omega$. $H$ is the external field and the interaction energy among the particles is represented by the Weiss field $K_s$. Thus (19.9) holds, where $U(s)$ is the expression \{ \} in (20.1). If for simplicity one takes $H = 0$ it is easily seen that $U(s)$ looks like Fig. 10a for $|K| < 1$ and like Fig. 10b for $|K| > 1$.

In order to endow this model with a temporal behavior, Ruijgrok and Tjon considered a collection of $N$ spins, each having an up and a down state. The number of up spins minus the number of down spins is $2n = 2N_s$. The canonical distribution is, supposing $N$ even,

$$P^{eq}(n) = \text{const} \left( \frac{N}{\frac{1}{2}N + n} \right) \exp \left[ \frac{2K}{N} n^2 + 2Hn \right] \quad (20.2)$$

which for large $N$ is the same as (20.1). The variable $n$ performs a random walk between $\frac{1}{2}N$ and $-\frac{1}{2}N$. The jump probabilities are taken in agreement with (20.2) and (6.4) to be

$$r(n) = (\frac{1}{2}N + n) \exp \left[ -\frac{2K}{N} n - H \right]$$

$$g(n) = (\frac{1}{2}N - n) \exp \left[ \frac{2K}{N} n + H \right]$$

It should be clear that they are not derived from any microscopic Hamiltonian, but are postulated as part and parcel of the model, in the spirit of Glauber's model. Otherwise one would rather expect to find transition probabilities similar to (19.3).

If one now lets $N$ grow, the function $U(s)$ remains the same, while the steps in the $s$-scale tend to zero. However, the equilibrium distribution (19.9) also changes and becomes very sharp. As a consequence the fluctuations of $s$ become small and therefore $U(s)$ may well be approximated near its minima by parabolas. This explains why the linear noise approximation holds and why the low-lying eigenvalues of the master equation are equidistant. Thus in the systematic expansion the nonlinear Fokker–Planck equation (19.7) never comes in, and efforts to solve it appear to be irrelevant.

As a second example we take the much discussed laser. At a low pumping rate the radiation is weak, proportional to the pumping, and incoherent, but at a certain threshold value the self-multiplication of photons suddenly starts. For single-mode operation the following master equation for the number $n$ of photons has been derived.

$$\dot{P}(n, t) = (E^{-1} - 1) \frac{A(n + 1)}{1 + (n + 1)/\Omega} P + C(E - 1)nP \quad (20.3)$$
The constant $A$ is the linear gain, $C$ the loss rate per phonon, $\Omega$ the volume of the cavity, in such units as to make the "saturation parameter" equal to $1/\Omega$. The stationary solution is given by (6.5):

$$P^{st}(n) = \frac{\text{const}}{\Gamma(n + \Omega + 1)} \left( \frac{A}{C} \right)^n \approx \text{const} \exp \left[ -\Omega \left\{ (s + 1) \log (s + 1) - s - s \log \frac{A}{C} \right\} \right]$$

where $s = \epsilon n = n/\Omega$. This is again of the form (19.9); the function $U(s)$ is the expression \{ \} and is sketched in Fig. 11. For $A > C$ there is a potential minimum at $s = (A - C)/C$, corresponding to a macroscopic number of photons. The variance of the photon number is

$$\langle (\Delta n)^2 \rangle = \Omega^2 \langle (\Delta s)^2 \rangle = \Omega^2 [\Omega U''(s)]^{-1} = \frac{\Omega A}{C} = \langle n \rangle + \Omega$$

which shows again that the fluctuations in $s$ are small for large $\Omega$.

![Fig. 11. The distribution given by (20.4).](image)

As a third example remember that the Malthus–Verhulst equation in Section IX showed a phase transition at $\beta = \alpha$. We now compute the stationary solution. The general formula (6.5) yields $P^{st}(n) = \delta_{n,0}$. In fact this is the sole time-independent solution, because for every other solution probability constantly leaks from $n = 1$ into $n = 0$ without returning. That is, the probability for a population to die out tends to unity, just as a gambler will always be ruined in the long run.* In practice, of course this probability is often so small that an almost stationary or metastable state

* This complication does not arise if there is an additional creation or immigration term as in (16.7). The stationary distribution for that case$^{50}$ has the same form (20.6).
prevails. Such a distribution must obey (6.4) except for small $n$ and is therefore

$$P_{ms}(n) = \frac{g(n - 1)g(n - 2)\cdots g(n_0)}{r(n)r(n - 1)\cdots r(n_0 + 1)} P_{ms}(n_0)$$

$$= \text{const} \frac{1}{n! \Gamma(n + \frac{\alpha \Omega}{\gamma})} \left( \frac{\beta \Omega}{\gamma} \right)^n$$

$$\cong \text{const} \exp \left[ -\Omega \left\{ \left( s + \frac{\alpha}{\gamma} \right) \log \left( s + \frac{\alpha}{\gamma} \right) - s - s \log \frac{\beta}{\gamma} \right\} \right] \quad (20.6)$$

The function $U(s)$ given by \{ \} is the same as in (20.4) and as sketched in Fig. 11. For $\beta > \alpha$ it has a minimum in agreement with (9.8). The leakage into $n = 0$ is roughly of order $\theta(\Omega)$.

$$\frac{P_{ms}(1)}{P_{ms}(\text{max})} = \exp \left[ -\Omega \{ U(1) - U(\text{min}) \} \right] \quad (20.7)$$

For $\beta < \alpha$ the minimum of $U$ lies at $n = 1$; the leak is not small and therefore the solution (20.6) becomes meaningless.

Our last example will be discussed in more detail because it serves for computing critical fluctuations in the next section. Consider the following autocatalytic chemical reaction, studied by Schlögl.\textsuperscript{30}

$$3X \xrightarrow{\frac{1}{3}} A + 2X$$

$$X \xrightarrow{\frac{\beta}{\gamma}} B \quad (20.8)$$

The macroscopic rate equation for the concentration $\phi$ of $X$ is

$$\dot{\phi} = -\phi^3 + 3\phi^2 - \beta\phi + \gamma \equiv \alpha_1(\phi) \quad (20.9)$$

The units of time and of $\Omega$ are chosen in such a way that the first two coefficients are 1 and 3.

For $\beta > 3$ (Fig. 12a) there is a single stationary state, which is asymptotically stable because $\alpha_1(\phi) < 0$.

For $\beta < 3$ and sufficiently large $\gamma$ (Fig. 12b) there is still a single stationary state with $\alpha_1(\phi^*) < 0$. There is an interval where $\alpha_1(\phi) > 0$, but nevertheless the fluctuations cannot grow indefinitely, because $\phi$ passes through that interval in a finite time and they remain of order $\Omega^{1/2}$. A similar situation occurred in the initial stage of the population growth in Section IX.

For $\beta < 3$ and small $\gamma$ (Fig. 12c) there are three stationary states, two of them stable. Hence the situation inside the reaction vessel is not uniquely determined by the externally imposed conditions. It may even happen that
both situations coexist in different regions of the vessel, but fluctuations in such inhomogeneous states are outside the scope of this article.

For $\beta < 3$ and one intermediate $\gamma$ (Fig. 12d) there is one stationary state $\phi^s$ that is stable and another one $\phi^u$ with $\alpha'_s(\phi^u) = 0$ and $\alpha'_u(\phi^u) > 0$. The equation for a small deviation $\delta \phi = \phi - \phi^u$ is

$$\frac{d}{dt} \delta \phi = \frac{1}{2} \alpha''(\phi^u)(\delta \phi)^2$$

(20.10)

A negative deviation decays slowly like $1/t$ rather than exponentially. A positive deviation, however, grows. Hence the fluctuations will grow in the

Fig. 12a. The function $\alpha_s(\phi)$ given in (20.9) for $\beta > 3$.

Fig. 12b. The function $\alpha_s(\phi)$ in (20.9) for $\beta < 3$ and sufficiently large $\gamma$.

Fig. 12c. The function $\alpha_s(\phi)$ in (20.9) for $\beta < 3$ and small $\gamma$. 
positive direction and ultimately cause a transition from \( \varphi^{us} \) into \( \varphi^{st} \). The \( \Omega \) expansion can describe the onset of this transition, but not the entire transition, since it breaks down by the time the fluctuations have reached a macroscopic size.

For \( \beta = 3, \gamma = 1 \) (Fig. 12e) there is a single stationary state, \( \varphi^{st} \), and \( \alpha'(\varphi^{st}) = \alpha''(\varphi^{st}) = 0 \), which is analogous to the critical point in fluids or magnetic systems. In the next section we compute the fluctuations about \( \varphi^{st} \) and \( \varphi^{us} \).

XXI. CRITICAL FLUCTUATIONS

The expansion method is based on the idea that the fluctuations in \( n \) are small and remain small, that is, of order \( \Omega^{1/2} \) as compared to the macroscopic part of order \( \Omega \). It is therefore necessary to verify \textit{a posteriori} that this is so (see Section V). Whereas the diffusion term in the Fokker–Planck equation (4.8) tends to make the fluctuations grow proportionally with \( \sqrt{\alpha_2 t} \), it is counteracted by the "convection term," which tries to squeeze them back towards equilibrium at the rate \( |\alpha'_2| \). Roughly speaking one has therefore\(^9\)

\[
\frac{\langle \Delta x^2 \rangle}{\Delta t} = \alpha_2 - 2|\alpha'_2|\langle x^2 \rangle
\]

(21.1)
which leads to the estimate

$$\langle x^2 \rangle = \frac{\alpha_2}{2|\alpha'_1|} \tag{21.2}$$

In equilibrium $\alpha'_1$ and $\alpha_2$ are constant; then this estimate is exact and expresses the connection between dissipation and fluctuations.

This argument assumes that $\alpha'_1(\phi) < 0$, which is also the condition for the macroscopic solution $\phi$ to be asymptotically stable. In the multivariate case too it was found that asymptotic stability of the macroscopic equation guaranteed that the fluctuations remain of order $\Omega^{1/2}$. This condition is not satisfied in the case of a limit cycle (Section XVIII); as a consequence the fluctuations grow and render the formal expansion invalid. The equations indicate that this happens, but do not describe the behavior after a time $t \sim \Omega$.

An even more disastrous example is provided by the Lotka–Volterra equations for two species, one of which feeds on the other. The rate equations are

$$\dot{n} = -\alpha n + \gamma \Omega^{-1} nm \tag{21.3a}$$
$$\dot{m} = \beta m - \delta \Omega^{-1} nm \tag{21.3b}$$

The same equations (with $\gamma = \delta$) also describe the chemical reaction

$$X \xrightarrow{\alpha} A$$
$$B + Y \xrightarrow{\beta} 2Y \tag{21.4}$$
$$X + Y \xrightarrow{\gamma} 2X$$

The macroscopic equations do not have a proper limit cycle, but all solutions are periodic.* Hence the fluctuations grow unchecked not only along the solution curves, but also perpendicular to them.

Returning to the one-variable case, one sees that difficulties arise when $\alpha'_1 > 0$. The convection term then magnifies the fluctuations exponentially rather than counteracting the diffusive tendency; the expansion breaks down at $t \sim \log \Omega$. When, however, $\alpha'_1 = 0$ the asymptotic stability of the macroscopic equations, and thereby the growth of the fluctuations, is determined by the higher derivatives of $\alpha_1$.

Suppose that the first jump moment $\alpha_1(\phi)$ has a zero $\phi^n$ where the first $q - 1$ derivatives vanish while $\alpha^{(q)}(\phi^n) \neq 0$. In order to compute the fluctuation we start from the general master equation (4.3). Instead of the

* This follows from the fact that there is a constant of the motion: $-\alpha \log m - \beta \log n + \Omega^{-1}(\gamma m + \delta n) = C$. The solution curves in $m, n$ space are determined by this equation.
transformation (4.6) we set more generally

\[ n = \Omega \varphi(\tau) + \Omega^\mu x \]  

(21.5)

The constant \( \mu \) is still adjustable, subject to the condition \( 0 < \mu < 1 \). The transformed master equation is

\[
\frac{\partial \Pi}{\partial \tau} - \Omega^{1-\mu} \frac{d\varphi}{d\tau} \frac{\partial \Pi}{\partial x} = - \Omega^{1-\mu} \frac{\partial}{\partial x} \alpha_1(\varphi + \Omega^{\mu-1} x) \Pi \\
+ \frac{1}{2} \Omega^{1-2\mu} \frac{\partial^2}{\partial x^2} \alpha_2(\varphi + \Omega^{\mu-1} x) \Pi \\
- \frac{1}{3!} \Omega^{1-3\mu} \frac{\partial^3}{\partial x^3} \alpha_3(\varphi + \Omega^{\mu-1} x) \Pi + \cdots 
\]  

(21.6)

The largest terms are of order \( \Omega^{1-\mu} \); they are made to vanish by choosing for \( \varphi \) a solution of (4.7). Take the stationary solution \( \varphi^* \); the fluctuations are then determined by

\[
\frac{\partial \Pi}{\partial \tau} = - \frac{1}{q!} \alpha_1^{(q)}(\varphi^*) \Omega^{1-q-1} \frac{\partial}{\partial x} x^q \Pi \\
+ \frac{1}{2} \alpha_2(\varphi^*) \Omega^{1-2\mu} \frac{\partial^2 \Pi}{\partial x^2} + \text{higher orders} 
\]  

(21.7)

In order to obtain an equation that causes \( x \) to remain of order unity we determine \( \mu \) by equating both powers of \( \Omega \),

\[
(1 - \mu)(1 - q) = 1 - 2\mu, \quad \mu = \frac{q}{q + 1} 
\]  

(21.8)

The result is

\[
\frac{\partial \Pi}{\partial \tau} = \Omega^{-(q-1)/(q+1)} \left[ - \frac{1}{q!} \alpha_1^{(q)} \frac{\partial}{\partial x} x^q \Pi + \frac{1}{2} \alpha_2 \frac{\partial^2 \Pi}{\partial x^2} \right] 
\]  

(21.9)

The reader will remember that \( \alpha_1^{(q)} \) and \( \alpha_2 \) are taken at \( \varphi^* \). Observe that the fluctuations vary on a slower time scale than the macroscopic quantity \( \varphi \). Another difference with the normal case is that the equation for the average

\[
\frac{d}{d\tau} \langle x \rangle = \Omega^{-(q-1)/(q+1)} \frac{1}{q!} \alpha_1^{(q)} \langle x^q \rangle 
\]  

(21.10)

is no longer just the variational equation associated with the macroscopic equation.

When \( q \) is even (as in Fig. 12d) it appears from (21.10) that positive fluctuations grow and negative ones decay, or vice versa, depending on the sign of
\( \alpha_1^{(q)} \). According to the equation they even become infinite after a time of order 
\[ \tau \sim \Omega^{(q-1)/(q+1)} \]
but of course the expansion breaks down as soon as they are 
of order \( x \sim \Omega^{1-\mu} = \Omega^{1/(q+1)} \), because then the distinction (21.5) between 
a macroscopic and a mesoscopic part becomes blurred.

The first interesting value of \( q \) is 3, which occurs at the critical point in 
Fig. 12e. Equation (21.9) then becomes
\[
\frac{\partial \Pi}{\partial \tau} = \Omega^{-1/2} \left[ -\frac{1}{6} \alpha_1^{''} \frac{\partial}{\partial x} x^3 \Pi + \frac{1}{2} \alpha_2 \frac{\partial^2 \Pi}{\partial x^2} \right]
\]
(21.11)

If \( \alpha_1^{''} < 0 \) the value of \( x \) remains finite, that is, the fluctuations in \( n \) remain of 
order \( \Omega^{3/4} \). More precisely, \( \Pi \) tends to
\[
\Pi(x, \infty) = \frac{1}{\Gamma(\frac{1}{3})} \left( \frac{4|\alpha_1^{''}|}{3\alpha_2} \right)^{1/4} \exp \left[ -\frac{|\alpha_1^{''}|}{12\alpha_2} x^4 \right]
\]
(21.12)
The rate of approach, however, is of order \( \Omega^{-1/2} \), which exhibits the critical 
slowing down.

XXII. CONCLUSIONS

The \( \Omega \) expansion is the appropriate way for solving master equations 
of the type (1.6) when the numbers involved are so large that the fluctuations 
are relatively small. It dispenses with \textit{ad hoc} assumptions about the smallness 
of certain terms, which gave rise to controversies and paradoxes.

The terms of order \( \Omega^{1/2} \) yield the macroscopic equation, which may well 
be nonlinear. Terms of relative order \( \Omega^{-1} \), which are sometimes included in 
the macroscopic equation, are left out, as they should be in the thermo-
dynamic limit.

The terms of order \( \Omega^0 \) give rise to a linear Fokker–Planck equation with 
time-dependent coefficients, describing the fluctuations around the macro-
scopric behavior (linear noise approximation). A nonlinear Fokker–Planck 
equation is merely an artifact, due to making \textit{ad hoc} approximations rather 
than a systematic expansion.

Fluctuations in a stationary state are in this approximation described by 
the familiar linear Fokker–Planck equation with constant coefficients and 
therefore constitute an Ornstein–Uhlenbeck process. Alternatively they may 
be treated by a Langevin equation, but the Langevin approach fails for 
fluctuations around time-dependent states.

In the next order the average value of \( n \) no longer coincides with the 
macroscopic value as determined by the macroscopic equation. The average 
does not obey an equation by itself, but can be found by solving the coupled 
equation (2.4) and (2.7).

In this and higher orders the Fokker–Planck equation is modified: 
additional powers are added in its coefficients, but at the same time higher
derivatives appear. As a consequence the stationary distribution need no longer be Gaussian, and the autocorrelation function of the fluctuations is no longer a single exponential. Nevertheless the moments can be computed successively.

The multivariate master equation can be treated in the same way. The only difficulty is solving the resulting set of coupled macroscopic equations. Once this is done the fluctuations can be found (in the linear noise approximation).

With slight modifications the $\Omega$ expansion also applies to fluctuations around critical points and other special solutions of the macroscopic equations, as long as they are asymptotically stable, so that the fluctuations remain small.

Exceptions are the solutions that are unstable (as occurring in phase transitions) or merely orbitally stable (such as limit cycles). The fluctuations around such solutions grow so as to obliterate the distinction between the macroscopic value and the mesoscopic fluctuations. In these cases the $\Omega$ expansion is valid only for a limited time and can at best indicate what happens after that.

These conclusions remain true in many cases having a continuous variable $q$ in lieu of the discrete $n$, but that is not the subject of this article.

**APPENDIX**

In order to carry out the program of Section IV one first has to solve (4.7) with given initial $\varphi(0)$. This can be done by a quadrature

$$\tau = \int_{\varphi(0)}^{\varphi(t)} \frac{d\varphi}{\alpha_1(\varphi)}$$

(A.1)

followed by an inversion to find $\varphi(\tau)$ as a function of $\tau$. We suppose this done and write for the solution $\varphi$, rather than $\varphi(\tau)$.

Next one may solve (5.1) by using $\varphi$, as the independent variable instead of $\tau$

$$\frac{d\langle x \rangle}{d\varphi} = \frac{\alpha_1(\varphi)}{\alpha_1(\varphi)} \langle x \rangle$$

(A.2)

The solution is given by

$$\frac{\langle x \rangle}{\alpha_1(\varphi)} = \frac{\langle x \rangle_0}{\alpha_1(\varphi_0)}$$

(A.3)

Alternatively one may write

$$\langle x \rangle = \frac{\langle x \rangle_0 \phi}{\phi_0}$$

(A.4)
which shows more clearly that \( \langle x \rangle \to 0 \) when \( \varphi \) tends to a limiting value \( \varphi^* \), so that \( \dot{x} \) tends to zero.

In the same way (5.2) is solved with the result

\[
\frac{\langle x^2 \rangle_t}{\alpha_1(\varphi_t)^2} = \frac{\langle x^2 \rangle_0}{\alpha_1(\varphi_0)^2} + \int_{\varphi_0}^{\varphi_t} \frac{\alpha_2(\varphi)}{\alpha_1(\varphi)^3} d\varphi
\]  

(A.5)

Again, when \( \varphi_t \to \varphi^* \) one has of course \( \alpha_1(\varphi^*) = 0 \) so that

\[
\langle x^2 \rangle_t \to \alpha_2(\varphi^*) \lim_{\varphi \to \varphi^*} \frac{\alpha_1(\varphi)^2}{\alpha_1(\varphi)^3} d\varphi
\]

\[
\to \frac{\alpha_2(\varphi^*)}{\alpha_1(\varphi^*)} \lim_{\varphi \to \varphi^*} (\varphi_t - \varphi^*)^2 \int_{\varphi_0}^{\varphi_t} \frac{d\varphi}{(\varphi - \varphi^*)^3}
\]

\[
\to \frac{1}{2} \frac{\alpha_2(\varphi^*)}{|\alpha_1(\varphi^*)|}
\]  

(A.6)

This expression is of course the same as the one obtained directly from (5.2) with \( \varphi = \varphi^* \) and identifying the left-hand side with zero.

Having found \( \langle x \rangle_t \) and \( \langle x^2 \rangle_t \) one obtains the solution of (4.8) by taking for \( \Pi \) a Gaussian with these values of \( \langle x \rangle \) and \( \langle x^2 \rangle \). The fact that it obeys (4.8) is explicitly verified for the multivariate case in Section XIV and need not be repeated here. We only note that the next order equation (5.5) can also be solved in this way with the following result:

\[
\frac{\langle x \rangle_t}{\alpha_1(\varphi_t)} = \frac{\langle x \rangle_0}{\alpha_1(\varphi_0)} + \frac{1}{2} \Omega^{-1/2} \times \left[ \frac{\alpha_1(\varphi_t) - \alpha_1(\varphi_0)}{\alpha_1(\varphi_0)^2} \langle x^2 \rangle_0 + \int_{\varphi_0}^{\varphi_t} \frac{\alpha_1'(\varphi) - \alpha_1'(\varphi_0)}{\alpha_1(\varphi)^3} \alpha_2(\varphi) d\varphi \right]
\]

In the case of the Malthus–Verhulst equation the solution (A.1) is given in (9.7). Substitution in (A.3) gives

\[
\langle x \rangle_t = \langle x \rangle_0 e^{(\beta - \alpha)t} \left[ 1 + \varphi(0) \frac{\gamma}{\beta - \alpha} \{ e^{(\beta - \alpha)t} - 1 \} \right]^{-2}
\]

Regardless of \( \langle x \rangle_0 \) this actually tends to zero both for \( \beta > \alpha \) and for \( \beta < \alpha \). In a similar way it is possible to compute \( \langle x^2 \rangle_t \) from (9.11) using (A.5), but the resulting expression is too long to be reproduced here.
References


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