Fluctuations in continuous systems

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Abstract

The density of particles in space or in phase space varies in time through continuous flow and through discrete jumps, due to collisions, chemical reactions, radioactive decay, or births and deaths [41]. A method is developed for computing the fluctuations arising from the random jumps and influenced by the flow. It equally applies to equilibrium, stationary states, and time-dependent situations, both in linear and nonlinear systems. The method is based on the master equation and does not require the additional assumptions needed in the Langevin approach, although the correct form of the Langevin force can be deduced from it *a posteriori*. Factorial cumulants turn out to be a convenient tool. As applications the fluctuations inherent in the diffusion process are computed and a controversial chemical reaction is discussed.

I. INTRODUCTION

The validity of macroscopic transport laws or rate equations is subject to restrictions due to the discrete nature of matter. The deviations from the macroscopic behavior show up in the fluctuations. In principle all the properties of the fluctuations are implied in the microscopic equations of motion of all particles, but the road from the microscopic starting point to the observed fluctuations is long and hazardous. It involves all the difficulties connected with the appearance of irreversibility, and cannot, therefore, dispense with some statistical assumptions. In practice one starts from an intermediate level of description, more detailed than the macroscopic picture, but less detailed than the microscopic one, and introduces some suitable and physically reasonable assumptions and the behavior on that level. "Mesoscopic" is a suitable term for this approach.

One way of computing fluctuations mesoscopically consists in supplementing the macroscopic equations with a fluctuating Langevin term and to assume certain stochastic properties for it. However, the physical justification for these assumptions is dubious, unless the Langevin term corresponds to an actual force, as in the Brownian particle case. Moreover, in time-dependent situations the Langevin method can only be saved by allowing the stochastic properties to depend on the special solution under consideration [1].

More satisfactory is the mesoscopic approach that starts by setting up the master equation. It equally applies to stationary and time-dependent states, and often requires no other assumptions than have already been used in establishing the macroscopic equations. All that is needed is to frame them in an equation for the probability distribution, *i.e.*, the master equation. One is then left, however, with the problem of solving it.

Exact solutions of the master equation are available only in a restricted class of problems, to be identified in Section II as *linear* master equations. For nonlinear master equations an approximation method has been developed, which produces solutions in the form of an expansion in reciprocal powers of a parameter measuring the size of the system. This method was first formulated for the case of a single fluctuating variable [2–7], and subsequently generalized to the multivariate case [1, 8]. The present article studies the case of infinitely many variables; more precisely, the case that the fluctuating quantity is itself a function of a continuously varying coordinate. Examples of such quantities are the density of particles in space, or in one-particle phase space.

Density fluctuations in equilibrium can of course be calculated by the usual phase-space methods [9], an example is given in Section VII. Non-equilibrium fluctuations in continuous systems have appeared in the literature in connection with various special problems, such as the derivation of the Onsager relations [10, 11], the theory of noise in electron beams [12], and the study of hot electrons [13–16]; and in connection with transport processes [17, 18], generalized hydrodynamics [19, 20], the electromagnetic field in dielectrics [21], lasers [22], chemical reactions [23–26], and even sociology [22, 27]. Often, however, recourse had to be taken to *ad hoc* methods, involving assumptions and simplifications (optimistically called "approximations"), which sometimes lead to ambiguities and controversies [23–26, 28–31]. A more general systematic, if rather cumbersome, treatment was given by Van Vliet [32], who obtained several of the results that will be derived here. His method, however, is based on the Langevin equation and is therefore subject to the objections mentioned above. Recently Mori [33] has treated continuous variables by Fourier transformation, which reduces them to a set of discrete variables.

Our aim is to provide a general method that permits to calculate without ambiguities the fluctuations in stationary or time-dependent states of linear and nonlinear systems with continuous variables, using no other assumptions than those that are indispensable for a mesoscopic treatment. Of course this does not mean that we claim to have resolved *all* difficulties; in particular the effect of space charge has yet to be worked out.

The difficulty in dealing with continuous variables is that discrete jumps, such as excitations or reactive collisions, have to be combined with the continuous change due to flow in the phase space. Although it is actually

possible to construct a master equation that incorporates both aspects, in this article a simpler approach will be used, based on the moments. The first and second moments vary as a consequence of both discrete and continuous processes. It is therefore possible to compute the two kinds of processes separately and compound them in the equations for the moments.

Our interest is focused on the correlations of the fluctuations at two points in space or phase space *at the same time*. We do not consider the correlations at different times needed to find the fluctuation spectrum. In stationary states, they can be derived from our results using the linearized macroscopic equations together with Onsager's assumption [10, 11].

The equation for the first moment (or average) is the macroscopic equation. The second moments (also called density-density or two-point correlation functions) describe the fluctuations. Equivalent with the second moments, but often more convenient, are the variance and covariance (also called the second cumulants). Even more convenient in many cases, however, are the less familiar 'factorial cumulants', whose properties are reviewed in Appendix A.

Our strategy will be, rather than to formulate the method in full generality, to demonstrate it on a series of examples so as to introduce successively various generalizations and complications. Inevitably the examples are somewhat artificial, because most applications to realistic cases require lengthier calculations than would be consistent with the purpose of elucidating the basic idea of the method. Yet the derivation of the fluctuating term inherent in the diffusion equation (see Section V) and the calculation of the fluctuations in the much discussed chemical reaction in Sections VIII and IX may be of practical interest.

II. DECAY WITH DIFFUSION

The aim of this section is to demonstrate the method of compounding moments on the simplest possible example. Take the dissociation of a chemical compound, or the decay of a radioactive isotope

$$X \xrightarrow{a} A, \tag{1}$$

where A is inert and does not enter into the equations. Let $N = 0, 1, 2, \ldots$ be the number of active particles X. The macroscopic rate equation for N as a function of t is of course

$$\frac{dN}{dt} = -aN. \tag{2}$$

This is an approximate law, valid when the number N is large. The actual value of N fluctuates around the value given by Eq. 2 owing to the statistical character of the individual decays. In order to describe the fluctuations one introduces the probability $P(N, t)$ for having N active particles at time t. The evolution of P is governed by the master equation

$$\frac{dP}{dt} = a \cdot (N+1) \cdot P(N+1, t) - a \cdot N \cdot P(N, t), \tag{3}$$

which is simply a gain-loss equation for the probability. For its justification *one only needs the same assumption on which Eq. 2 is based*: each active particle has a fixed probability a per unit time to decay. Of course this implies the Markov property in that a does not depend on the history of the particle.

The object is to solve Eq. 3 with arbitrary initial value $P(N, 0)$; it clearly suffices to take

$$P(N, 0) = \delta_{N, N_0}, \quad N_0 \text{ arbitrary}. \tag{4}$$

The solution will determine the average of N, and the fluctuations around it at all $t > 0$. For our purposes it suffices to find the average and the mean square of N.

It is convenient to introduce the step operator $\mathbf{E}$ defined by

$$\mathbf{E}f(N) = f(N+1),$$

for an arbitrary function f. Note the identity

$$\sum_{N=0}^{\infty} g(N)\mathbf{E}f(N) = \sum_{N=0}^{\infty} f(N)\mathbf{E}^{-1}g(N) \tag{5}$$

for any pair of functions f, g such that the sum converges and that $g(0) = 0$. With the aid of this step operator the master equation Eq. 3 may be written

$$\frac{dP}{dt} = a \cdot (\mathbf{E} - 1) \cdot N \cdot P. \tag{6}$$

By definition the master equation is linear in the unknown function P. The term "linear" is therefore available and will now be used to denote the fact that the coefficients in Eqs. 3 and 6 are linear functions of N. Linear master equations describe collections of particles that do not interfere with each other – neither by actual forces nor through Bose or Fermi statistics. It will appear that they lead to linear macroscopic laws.

From a mathematical point of view linear master equations have an especially simple structure and can be solved by deriving equations from them for the successive moments. Multiply Eq. 6 with N and sum over all N, using Eq. 5,

$$\frac{d\langle N \rangle}{dt} = a \sum_{N=0}^{\infty} N(\mathbf{E} - 1)NP = a\langle N(\mathbf{E}^{-1} - 1)N \rangle$$

$$= -a\langle N \rangle. \tag{7}$$

The angular brackets denote averages. Thus the rate of change of the first moment depends on that same moment alone. In fact, Eq. 7 is nothing but the macroscopic Eq. 2.

Similarly one finds for the second moment

$$\frac{d\langle N^2 \rangle}{dt} = 2a\langle N^2 \rangle + a\langle N \rangle. \qquad (8)$$

Thus its rate of change depends on the first and second moments, but not on higher ones, so that the Eqs. 7 and 8 can be solved successively.

The solutions corresponding to the initial condition Eq. 4 are

$$\langle N \rangle_t = N_0 e^{-at},$$
$$\langle N^2 \rangle_t = N_0^2 e^{-2at} + N_0 e^{-at}(1 - e^{-at}). \qquad (9)$$

Hence it is possible to obtain the desired information, without determining the complete solution of the master equation. For nonlinear master equations a different approach will be needed (Section VIII).

Although the two moments contain all the information we need, two auxiliary quantities will be useful. The variance or second cumulant, to be denoted by double brackets

$$\langle\langle N^2 \rangle\rangle = \langle N^2 \rangle - \langle N \rangle^2,$$

describes more directly the fluctuations and vanishes for the initial state Eq. 4. The 'factorial cumulant' will be denoted by square brackets and is defined by (compare Appendix A)

$$\left[N^2 \right] = \langle\langle N^2 \rangle\rangle - \langle N \rangle = \langle N^2 \rangle - \langle N \rangle^2 - \langle N \rangle.$$

It often obeys a simpler equation than either the second moment or the variance. For instance in the present case

$$\frac{d\langle\langle N^2 \rangle\rangle}{dt} = -2a\langle\langle N^2 \rangle\rangle + a\langle N \rangle,$$
$$\frac{d\left[N^2 \right]}{dt} = -2a\left[N^2 \right].$$

The fact that the latter equation does not have an inhomogeneous term will turn out to greatly simplify the solution in more complicated examples. So far only the total number N of active particles X has been studied. In order to take into account the location in space, we subdivide the total volume in cells Δ, labelled with a Greek subscipt[42]. The number of particles in each cell λ is N_λ and P is a function of the set $\{N_\lambda\}$. The time dependence will no longer be indicated explicitly. The master equation is

$$\frac{dP(\{N_\lambda\})}{dt} = a \sum_\lambda (\mathbf{E}_\lambda - 1) N_\lambda P, \qquad (10)$$

where $\mathbf{E}_\lambda$ is the step operator acting on the variable N_λ.

The equations for the first two moments are again obtained by multiplying Eq. 10 with N_α and $N_\alpha N_\beta$ respec-

Each cell is a well-mixed reaction vessel

$$N_\lambda \rightarrow N_\mu$$

$$\left(N_\lambda + 1, N_\mu \right) \xrightarrow{\ w_{\mu\lambda} N_\lambda\ } \left(N_\lambda, N_\mu + 1 \right)$$
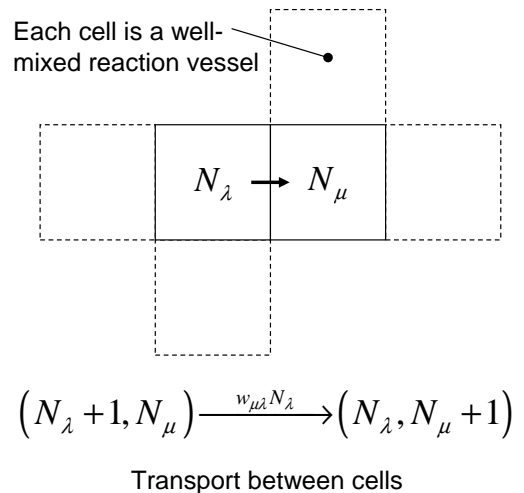
Transport between cells

FIG. 1: **Subdivision of the total volume.** The total volume is subdivided into cells of volume Δ. The number of particles in each cell λ is N_λ. We assume the volume is *small enough* that each cell is a well-mixed, homogeneous reaction vessel; however, we assume N_λ is *large enough* that the linear noise approximation can be applied within each cell.

tively and summing over all $\{N_\lambda\}$,

$$\frac{d\langle N_\alpha \rangle}{dt} = -a\langle N_\alpha \rangle, \qquad (11)$$

and,

$$\frac{d\langle N_\alpha N_\beta \rangle}{dt} = -2a\langle N_\alpha N_\beta \rangle + a\langle N_\alpha \rangle \delta_{\alpha,\beta}. \qquad (12)$$

These equations might be surmised directly from Eqs. 7 and 8 by noting that there is no statistical correlation among the cells, since so far no transport of particles between them has been included. The solutions can again easily be obtained, but that is not our aim.

The following step is the elimination of the cells by going to a continuous description. Denote the density N_α/Δ in cell α by $n(\mathbf{r})$, where $\mathbf{r}$ is the position vector of that cell. Then Eq. 11 may be written, on dividing by Δ,

$$\frac{d\langle n(\mathbf{r}) \rangle}{dt} = -a\langle n(\mathbf{r}) \rangle. \qquad (13)$$

Of course the functions $n(\mathbf{r})$ and $\langle n(\mathbf{r}) \rangle$ are only defined on a coarse-grained grid in $\mathbf{r}$-space, determined by the cell size Δ. In the present example, however, this is not a restriction, because Δ may be taken as small as one wishes. As a check we integrate over an arbitrary volume V to obtain for the number N_V of active particles in V

$$\langle N_V \rangle = \int_V \langle n(\mathbf{r}) \rangle d\mathbf{r}.$$

When V is taken to be one of the cells, integration of Eq. 13 reproduces Eq. 11.

Similarly we divide Eq. 12 by Δ^2. The Kronecker symbol $\delta_{\alpha,\beta}$ considered as a function of β, vanishes everywhere except inside the cell α. Hence its integral over space equals Δ, so that $\delta_{\alpha,\beta}/\Delta$ may be identified with Dirac's delta function. Thus Eq. 12 becomes

$$\frac{d\langle n(\mathbf{r}_1)n(\mathbf{r}_2)\rangle}{dt} = -2a\langle n(\mathbf{r}_1)n(\mathbf{r}_2)\rangle + a\langle n(\mathbf{r}_1)\rangle\delta(\mathbf{r}_1 - \mathbf{r}_2). \tag{14}$$

This can again be justified by integrating $\mathbf{r}_1$ over cell α and $\mathbf{r}_2$ over cell β, which reproduces Eq. 12.

The second cumulant now turns into a mixed cumulant or covariance matrix

$$\langle\langle N_\alpha N_\beta\rangle\rangle = \langle N_\alpha N_\beta\rangle - \langle N_\alpha\rangle\langle N_\beta\rangle,$$

which obeys

$$\frac{d\langle\langle N_\alpha N_\beta\rangle\rangle}{dt} = -2a\langle\langle N_\alpha N_\beta\rangle\rangle + a\langle N_\alpha\rangle\delta_{\alpha,\beta}. \tag{15}$$

The factorial cumulant is by definition

$$[N_\alpha N_\beta] = \langle\langle N_\alpha N_\beta\rangle\rangle - \langle N_\alpha\rangle\delta_{\alpha,\beta}, \tag{16}$$

and obeys

$$\frac{d[N_\alpha N_\beta]}{dt} = -2a[N_\alpha N_\beta].$$

Again on dividing by Δ^2, one has in the continuous description

$$\frac{d\langle\langle n(\mathbf{r}_1)n(\mathbf{r}_2)\rangle\rangle}{dt} = -2a\langle\langle n(\mathbf{r}_1)n(\mathbf{r}_2)\rangle\rangle + a\langle n(\mathbf{r}_1)\rangle\delta(\mathbf{r}_1 - \mathbf{r}_2),$$

$$\frac{d[n(\mathbf{r}_1)n(\mathbf{r}_2)]}{dt} = -2a[n(\mathbf{r}_1)n(\mathbf{r}_2)]. \tag{17}$$

Either of these equations may be used instead of Eq. 14.

Having obtained the rates at which the first and second moments vary as a result of the decay process, we may now combine them with those caused by transport. Suppose the particles X are molecules in a fluid in which they diffuse with diffusion coefficient D. If only diffusion took place without decay, one would expect

$$\frac{d\langle n(\mathbf{r})\rangle}{dt} = D\nabla^2\langle n(\mathbf{r})\rangle, \tag{18}$$

and

$$\frac{d\langle n(\mathbf{r}_1)n(\mathbf{r}_2)\rangle}{dt} = D(\nabla_1^2 + \nabla_2^2)\langle n(\mathbf{r}_1)n(\mathbf{r}_2)\rangle, \tag{19}$$

where ∇_1^2 and ∇_2^2 are Laplace operators with respect to $\mathbf{r}_1$ and $\mathbf{r}_2$. Eq. 19, however, is not correct, because it treats diffusion as a deterministic process. Actually the diffusion Eq. 18 is the average of a random process. Hence

the diffusion is a source of additional noise, which has to be taken into account by adding suitable terms to Eq. 19. This will be done in Section V and we here anticipate the result: *The noisy character of diffusion is correctly taken into account by writing instead of Eq. 19*

$$\frac{d[n(\mathbf{r}_1)n(\mathbf{r}_2)]}{dt} = D(\nabla_1^2 + \nabla_2^2)[n(\mathbf{r}_1)n(\mathbf{r}_2)]. \tag{20}$$

Finally, we obtain the combined effect of decay and diffusion by adding the effects that each of them has on the moments. First Eqs. 13 and 18 lead to

$$\frac{d\langle n(\mathbf{r})\rangle}{dt} = -a\langle n(\mathbf{r})\rangle + D\nabla^2\langle n(\mathbf{r})\rangle, \tag{21}$$

which is the macroscopic equation. Secondly, we compound Eqs. 17 and 20 to obtain

$$\frac{d[n(\mathbf{r}_1)n(\mathbf{r}_2)]}{dt} =$$
$$-2a[n(\mathbf{r}_1)n(\mathbf{r}_2)] + D(\nabla_1^2 + \nabla_2^2)[n(\mathbf{r}_1)n(\mathbf{r}_2)]. \tag{22}$$

The Eqs. 21 and 22 together describe the macroscopic behavior of the density of particles X and the fluctuations around it caused by the statistical character of the individual decay events, and by the stochastic nature of diffusion.

It is easy to solve Eqs. 21 and 22 for given initial density $n_0(\mathbf{r})$. The solution of Eq. 21 is readily seen to be

$$\langle n(\mathbf{r})\rangle_t = \frac{e^{-at}}{(4\pi Dt)^{3/2}}\int \exp\left[-\frac{(\mathbf{r} - \mathbf{r}')^2}{4Dt}\right]n_0(\mathbf{r})d\mathbf{r}. \tag{23}$$

Similarly the general solution of Eq. 22 is

$$[n(\mathbf{r}_1)n(\mathbf{r}_2)]_t = \frac{e^{-2at}}{(4\pi Dt)^3}\times$$
$$\times\int\exp\left[-\frac{(\mathbf{r}_1 - \mathbf{r}_1')^2 + (\mathbf{r}_2 - \mathbf{r}_2')^2}{4Dt}\right][n(\mathbf{r}_1')n(\mathbf{r}_2')]_0 d\mathbf{r}_1' d\mathbf{r}_2'.$$

As the initial value is given by $[n(\mathbf{r}_1)n(\mathbf{r}_2)]_0 = -\delta(\mathbf{r}_1 - \mathbf{r}_2)n_0(\mathbf{r}_1)$, this reduces to

$$[n(\mathbf{r}_1)n(\mathbf{r}_2)]_t = \frac{-e^{-2at}}{(4\pi Dt)^3}\exp\left[-\frac{r_1^2 + r_2^2}{4Dt}\right]\times$$
$$\times\int\exp\left[-\frac{(\mathbf{r}'^2 - (\mathbf{r}_1 + \mathbf{r}_2)\cdot\mathbf{r}')}{2Dt}\right]n_0(\mathbf{r}')d\mathbf{r}' =$$
$$= \frac{-e^{-2at}}{(4\pi Dt)^3}\exp\left[-\frac{(\mathbf{r}_1 - \mathbf{r}_2)^2}{8Dt}\right]\times$$
$$\times\int\exp\left[-\frac{\mathbf{r}'^2}{2Dt}\right]n_0\left(\mathbf{r}' + \frac{\mathbf{r}_1 + \mathbf{r}_2}{2}\right)d\mathbf{r}'. \tag{24}$$

The covariance of the fluctuations $\langle\langle n(\mathbf{r}_1)n(\mathbf{r}_2)\rangle\rangle$ is obtained by multiplying Eq. 23 with $\delta(\mathbf{r}_1 - \mathbf{r}_2)$ and adding it to Eq. 24. We give the result for the special case

$n_0(\mathbf{r}) = \text{constant} = n_0$:

$$\langle\langle n(\mathbf{r}_1)n(\mathbf{r}_2)\rangle\rangle = \delta(\mathbf{r}_1 - \mathbf{r}_2)n_0 e^{-at} - \\ - \frac{n_0 e^{-2at}}{(8\pi Dt)^{3/2}} \exp\left[-\frac{(\mathbf{r}_1 - \mathbf{r}_2)^2}{8Dt}\right].$$

Obviously, it only depends on $|\mathbf{r}_1 - \mathbf{r}_2|$. On integrating over $\mathbf{r}_1$ and $\mathbf{r}_2$, one obtains the variance in the total number N in agreement with Eq. 9.

III. THE DENSITY IN PHASE SPACE

In this section the method of compounding moments is extended to a distribution in phase space, rather than just coordinate space. As an example, we consider the same decay process described by Eq. 1, but suppose that the particles move freely in space rather than diffusing in a medium. It is then necessary to distinguish them according to both their position $\mathbf{r}$ and velocity $\mathbf{v}$. Accordingly, the cells Δ are now six-dimensional cells in the one-particle phase space and $n(\mathbf{r}, \mathbf{v})$ is the phase space density. For chemical reactions, it is more realistic to allow a to depend on $|\mathbf{v}| = v$. The master equation for the cell distribution then takes the form

$$\frac{dP(\{N_\lambda\})}{dt} = \sum_\lambda a_\lambda (\mathbf{E}_\lambda - 1) N_\lambda P. \qquad (25)$$

The equations for the moments are

$$\frac{d\langle N_\alpha\rangle}{dt} = -a_\alpha\langle N_\alpha\rangle,$$
$$\frac{d\langle N_\alpha N_\beta\rangle}{dt} = -(a_\alpha + a_\beta)\langle N_\alpha N_\beta\rangle + a_\alpha\langle N_\alpha\rangle\delta_{\alpha,\beta}.$$

In the continuous description, one now has to put

$$\frac{\delta_{\alpha,\beta}}{\Delta} \mapsto \delta(\mathbf{r}_1 - \mathbf{r}_2)\delta(\mathbf{v}_1 - \mathbf{v}_2).$$

The result is

$$\frac{d\langle n(\mathbf{r}, \mathbf{v})\rangle}{dt} = -a(v)\langle n(\mathbf{r}, \mathbf{v})\rangle,$$
$$\frac{d\langle n(\mathbf{r}_1, \mathbf{v}_1)n(\mathbf{r}_2, \mathbf{v}_2)\rangle}{dt} = -[a(v_1) + a(v_2)]\langle n(\mathbf{r}_1, \mathbf{v}_1)n(\mathbf{r}_2, \mathbf{v}_2)\rangle \\ + a(v_1)\langle n(\mathbf{r}_1, \mathbf{v}_1)\rangle\delta(\mathbf{r}_1 - \mathbf{r}_2)\delta(\mathbf{v}_1 - \mathbf{v}_2).$$

The last equation again simplifies when written in terms of the factorial cumulant:

$$\frac{d[n(\mathbf{r}_1, \mathbf{v}_1)n(\mathbf{r}_2, \mathbf{v}_2)]}{dt} = -[a(v_1) + a(v_2)][n(\mathbf{r}_1, \mathbf{v}_1)n(\mathbf{r}_2, \mathbf{v}_2)]. \qquad (26)$$

These equations are now compounded with the flow term:

$$\frac{d\langle n(\mathbf{r}, \mathbf{v})\rangle}{dt} = -\mathbf{v}\cdot\nabla - a(v)\langle n(\mathbf{r}, \mathbf{v})\rangle, \qquad (27)$$

and,

$$\frac{d\langle n(\mathbf{r}_1, \mathbf{v}_1)n(\mathbf{r}_2, \mathbf{v}_2)\rangle}{dt} = \\ (-\mathbf{v}_1\cdot\nabla_1 - \mathbf{v}_2\cdot\nabla_2)\langle n(\mathbf{r}_1, \mathbf{v}_1)n(\mathbf{r}_2, \mathbf{v}_2)\rangle \\ - [a(v_1) + a(v_2)]\langle n(\mathbf{r}_1, \mathbf{v}_1)n(\mathbf{r}_2, \mathbf{v}_2)\rangle \\ + a(v_1)\langle n(\mathbf{r}_1, \mathbf{v}_1)\rangle\delta(\mathbf{r}_1 - \mathbf{r}_2)\delta(\mathbf{v}_1 - \mathbf{v}_2). \qquad (28)$$

In this case, the flow terms are to be inserted in the second moment rather than in the factorial cumulant, since the flow in phase space is a deterministic process and not an additional noise source (Appendix B). The Eqs. 27 and 28 determine the average flow in phase space and the fluctuations around it.

It is again somewhat simpler to rewrite Eq. 28 in terms of the factorial cumulant. After some algebra, one finds the simple result

$$\frac{d[n(\mathbf{r}_1, \mathbf{v}_1)n(\mathbf{r}_2, \mathbf{v}_2)]}{dt} = \\ -[\mathbf{v}_1\cdot\nabla_1 + \mathbf{v}_2\cdot\nabla_2 + a(v_1) + a(v_2)][n(\mathbf{r}_1, \mathbf{v}_1)n(\mathbf{r}_2, \mathbf{v}_2)]. \qquad (29)$$

On comparing this with Eq. 26 one observes that actually no error is made by inserting the flow terms in the equation for the factorial cumulant instead of in the equation for the second moment. This fact is proved generally in Appendix B.

The solution of Eq. 27 with given initial $n_0(\mathbf{r}, \mathbf{v})$ is

$$\langle n(\mathbf{r}, \mathbf{v})\rangle_t = e^{-a(v)t}n_0(\mathbf{r} - \mathbf{v}t, \mathbf{v}).$$

Similarly, Eq. 29 is solved by

$$[n(\mathbf{r}_1, \mathbf{v}_1)n(\mathbf{r}_2, \mathbf{v}_2)]_t = e^{-[a(v_1) + a(v_2)]t}[n(\mathbf{r}_1, \mathbf{v}_1)n(\mathbf{r}_2, \mathbf{v}_2)]_0 \\ = e^{-2a(v_1)t}n_0(\mathbf{r} - \mathbf{v}t, \mathbf{v})\delta(\mathbf{r}_1 - \mathbf{r}_2)\delta(\mathbf{v}_1 - \mathbf{v}_2).$$

Hence the correlation of the fluctuations in phase space are

$$\langle\langle n(\mathbf{r}_1, \mathbf{v}_1)n(\mathbf{r}_2, \mathbf{v}_2)\rangle\rangle_t = \\ \left[1 - e^{-a(\mathbf{v}_1)t}\right]\langle n(\mathbf{r}_1, \mathbf{v}_1)\rangle\delta(\mathbf{r}_1 - \mathbf{r}_2)\delta(\mathbf{v}_1 - \mathbf{v}_2).$$

It appears that there is no correlation between the fluctuations at different points of phase space. The reason is again that the particles do not interfere with one another.

IV. FLUCTUATIONS IN A STATIONARY STATE

The decay process in the previous two sections tends to an equilibrium in which everything vanishes. In this

section we add a term that creates particles, so as to make a nontrivial stationary state possible. The scheme is

$$B \xrightarrow{b} X, \quad X \xrightarrow{a} A, \qquad (30)$$
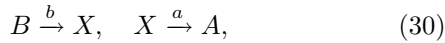
and may be regarded as an intermediate stage of the radioactive decay process, or as a chemical reaction. B is supposed to be a reservoir of practically constant amount. The probability per unit time that an X is created in a given cell λ is proportional to the amount of B in that cell and will therefore be denoted by $b\Delta$. The master equation describing Eq. 30 is

$$\frac{dP(\{N_\lambda\})}{dt} = b\Delta \sum_\lambda \left(\mathbf{E}_\lambda^{-1} - 1 \right) P + a \sum_\lambda \left(\mathbf{E}_\lambda - 1 \right) N_\lambda P.$$
$$(31)$$

where N_λ is again the number of particles X in cell λ.

As this master equation is still linear it is possible to derive from it equations for separate moments

$$\frac{d\langle N_\alpha \rangle}{dt} = b\Delta - a\langle N_\alpha \rangle,$$
$$\frac{d\langle N_\alpha N_\beta \rangle}{dt} = b\Delta \left[\langle N_\alpha \rangle + \langle N_\beta \rangle + \delta_{\alpha,\beta} \right]$$
$$+ a \left[-2\langle N_\alpha N_\beta \rangle + \langle N_\alpha \rangle \delta_{\alpha,\beta} \right].$$

In terms of the variance, the latter equation simplifies to

$$\frac{d\langle\langle N_\alpha N_\beta \rangle\rangle}{dt} = -2a\langle\langle N_\alpha N_\beta \rangle\rangle + \left[b\Delta + a\langle N_\alpha \rangle \right] \delta_{\alpha,\beta},$$

and in terms of the factorial cumulant as

$$\frac{d[N_\alpha N_\beta]}{dt} = -2a[N_\alpha N_\beta]. \qquad (32)$$

Dividing by Δ and Δ^2, respectively, and using the continuous notation, one gets

$$\frac{d\langle n(\mathbf{r}) \rangle}{dt} = b - a\langle n(\mathbf{r}) \rangle,$$
$$\frac{d[n(\mathbf{r}_1)n(\mathbf{r}_2)]}{dt} = -2a[n(\mathbf{r}_1)n(\mathbf{r}_2)].$$

Now suppose again that the reaction occurs in a diffusive medium and add the corresponding terms:

$$\frac{d\langle n(\mathbf{r}) \rangle}{dt} = b - a\langle n(\mathbf{r}) \rangle + D\nabla^2 \langle n(\mathbf{r}) \rangle, \qquad (33)$$

$$\frac{d[n(\mathbf{r}_1)n(\mathbf{r}_2)]}{dt} = \left\{ -2a + D \left(\nabla_1^2 + \nabla_2^2 \right) \right\} [n(\mathbf{r}_1)n(\mathbf{r}_2)].$$
$$(34)$$

These equations are easily solved; the solution of Eq. 33 is obtained from Eq. 23 by replacing n with $n-b/a$, while the solution of Eq. 34 is identical with Eq. 24. However,

we here merely want to study the stationary state given by

$$\langle n(\mathbf{r}) \rangle^{st} = b/a,$$
$$D \left(\nabla_1^2 + \nabla_2^2 \right) [n(\mathbf{r}_1)n(\mathbf{r}_2)]^{st} = -2a[n(\mathbf{r}_1)n(\mathbf{r}_2)]^{st}.$$

It is easily seen that the latter equation has only one solution that does not grow exponentially for large $|\mathbf{r}_1|$ or $|\mathbf{r}_2|$, $viz.$,

$$[n(\mathbf{r}_1)n(\mathbf{r}_2)]^{st} = 0.$$

The variance of the fluctuations is therefore

$$\langle\langle n(\mathbf{r}_1)n(\mathbf{r}_2) \rangle\rangle^{st} = \langle n(\mathbf{r}_1) \rangle^{st} \delta(\mathbf{r}_1 - \mathbf{r}_2). \qquad (35)$$

Conclusions: (i) in the stationary state, there is no correlation between fluctuations at different points in space; (ii) the diffusion does not affect the fluctuations; (iii) the fluctuations are the same as for a Poisson distribution, in agreement with the fact that the individual particles X are independent of each other.

Instead of diffusion we now suppose that the particles move freely in space, as in Section III, and accordingly study the phase space density $n(\mathbf{r}, \mathbf{v})$. The probability for generating a particle will depend on v, but it will still be supposed independent of $\mathbf{r}$. In lieu of the two Eqs. 33 and 34, we now have

$$\frac{d\langle n(\mathbf{r}, \mathbf{v}) \rangle}{dt} = b(v) - a(v)\langle n(\mathbf{r}, \mathbf{v}) \rangle - \mathbf{v} \cdot \nabla \langle n(\mathbf{r}, \mathbf{v}) \rangle,$$
$$\frac{d[n(\mathbf{r}_1, \mathbf{v}_1)n(\mathbf{r}_2, \mathbf{v}_2)]}{dt} = - \left\{ a(v_1) + a(v_2) \right\} [n(\mathbf{r}_1, \mathbf{v}_1)n(\mathbf{r}_2, \mathbf{v}_2)]$$
$$- \left\{ \mathbf{v}_1 \cdot \nabla_1 + \mathbf{v}_2 \cdot \nabla_2 \right\} [n(\mathbf{r}_1, \mathbf{v}_1)n(\mathbf{r}_2, \mathbf{v}_2)].$$

The solutions can again be obtained explicitly, but we are interested in the stationary state,

$$\langle n(\mathbf{r}, \mathbf{v}) \rangle^{st} = b(v)/a(v),$$
$$[n(\mathbf{r}_1, \mathbf{v}_1)n(\mathbf{r}_2, \mathbf{v}_2)]^{st} = 0,$$
$$\langle\langle n(\mathbf{r}_1, \mathbf{v}_1)n(\mathbf{r}_2, \mathbf{v}_2) \rangle\rangle^{st} = \langle n(\mathbf{r}_1, \mathbf{v}_1) \rangle^{st} \delta(\mathbf{r}_1 - \mathbf{r}_2)\delta(\mathbf{v}_1 - \mathbf{v}_2).$$

This result is similar to Eq. 35 and shows that the propagation does not affect the fluctuations in the stationary state.

V. DIFFUSION AS A STOCHASTIC PROCESS

Let coordinate space be subdivided in cells Δ, and suppose that there is a probability $w_{\lambda\mu}dt$ for a particle that is in cell μ at time t to find itself in cell λ at time $t + dt$. The probability distribution $P(\{N_\lambda\})$ of the occupation numbers of the cells obeys the master equation

$$\frac{dP(\{N_\lambda\})}{dt} = \sum_{\lambda,\mu} w_{\lambda\mu} \left(\mathbf{E}_\lambda^{-1}\mathbf{E}_\mu - 1 \right) N_\mu P. \qquad (36)$$

The first moment obeys the equation, *cf.* Eq. 5,

$$\frac{d\langle N_\alpha\rangle}{dt} = \sum_{\lambda,\mu} w_{\lambda\mu}\langle N_\mu\left(\mathbf{E}_\lambda\mathbf{E}_\mu^{-1} - 1\right)N_\alpha\rangle$$

$$= \sum_{\lambda,\mu} w_{\lambda\mu}\langle N_\mu\left(N_\alpha + \delta_{\alpha,\lambda} - \delta_{\alpha,\mu} - N_\alpha\right)$$

$$= \sum_\mu w_{\alpha\mu}\langle N_\mu\rangle - \sum_\lambda w_{\lambda\alpha}\langle N_\alpha\rangle. \qquad (37)$$

To save writing, we define the matrix

$$\mathbb{W}_{\alpha\beta} = w_{\alpha\beta} - \delta_{\alpha,\beta}\sum_\lambda w_{\lambda\alpha}.$$

Then the equation for the first moments may be written

$$\frac{d\langle N_\alpha\rangle}{dt} = \sum_\mu \mathbb{W}_{\alpha\mu}\langle N_\mu\rangle. \qquad (38)$$

For the second moments one finds in a similar way

$$\frac{d\langle N_\alpha N_\beta\rangle}{dt} = \sum_\mu \mathbb{W}_{\alpha\mu}\langle N_\mu N_\beta\rangle + \sum_\mu \mathbb{W}_{\beta\mu}\langle N_\alpha N_\mu\rangle$$

$$-w_{\alpha\beta}\langle N_\beta\rangle - w_{\beta\alpha}\langle N_\alpha\rangle$$

$$+\delta_{\alpha,\beta}\left\{\sum_\mu w_{\alpha\mu}\langle N_\mu\rangle + \sum_\lambda w_{\lambda\alpha}\langle N_\alpha\rangle\right\}. \qquad (39)$$

Again this equation simplifies when written in the factorial cumulant

$$\frac{d[N_\alpha N_\beta]}{dt} = \sum_\mu \mathbb{W}_{\alpha\mu}[N_\mu N_\beta] + \sum_\mu \mathbb{W}_{\beta\mu}[N_\alpha N_\mu]. \quad (40)$$

Writing $\mathbb{W}$ for the matrix $\mathbb{W}_{\alpha\beta}$ and Θ for the matrix $[N_\alpha N_\beta]$ one has the matrix equation

$$\frac{d\Theta}{dt} = \mathbb{W}\cdot\Theta + \Theta\cdot\widetilde{\mathbb{W}}, \qquad (41)$$

where the tilde denotes transposition.

In order to go to the continuous description one should realize that $w_{\lambda\mu}$ is proportional to the size of the receiving cell λ and therefore involves a factor Δ. This provides the $d\mathbf{r}$ needed to change the sums into integrals

$$w_{\lambda\mu} \rightarrow w(\mathbf{r}|\mathbf{r}'),$$

$$\mathbb{W}_{\lambda\mu} \rightarrow \left\{w(\mathbf{r}|\mathbf{r}') - \delta(\mathbf{r}-\mathbf{r}')\int d\mathbf{r}''w(\mathbf{r}''|\mathbf{r})\right\}d\mathbf{r}. \quad (42)$$

The Eqs. 37 and 38 therefore become

$$\frac{d\langle n(\mathbf{r})\rangle}{dt} = \int w(\mathbf{r}|\mathbf{r}')\langle n(\mathbf{r}')\rangle d\mathbf{r}' - \langle n(\mathbf{r})\rangle\int w(\mathbf{r}'|\mathbf{r})d\mathbf{r}'$$

$$= \int \mathbb{W}(\mathbf{r}|\mathbf{r}')\langle n(\mathbf{r}')\rangle d\mathbf{r}'. \qquad (43)$$

Similarly, Eq. 40 takes the form

$$\frac{d[n(\mathbf{r}_1)n(\mathbf{r}_2)]}{dt} = \int \mathbb{W}(\mathbf{r}_1|\mathbf{r}')[n(\mathbf{r}')n(\mathbf{r}_2)]d\mathbf{r}'+$$

$$\int \mathbb{W}(\mathbf{r}_2|\mathbf{r}')[n(\mathbf{r}_1)n(\mathbf{r}')]d\mathbf{r}', \qquad (44)$$

which is also covered by the symbolic form Eq. 41.

In order that the average density obeys the diffusion equation, one must identify the operator $\mathbb{W}$ in Eq. 43 with $D\nabla^2$. It then follows from Eq. 44 that $[n(\mathbf{r}_1)n(\mathbf{r}_2)]$ obeys

$$\frac{d[n(\mathbf{r}_1)n(\mathbf{r}_2)]}{dt} = D\left(\nabla_1^2 + \nabla_2^2\right)[n(\mathbf{r}_1)n(\mathbf{r}_2)]. \qquad (45)$$

This is the result anticipated in Section II. Note that Eq. 19 is indeed incorrect since the second moment obeys the more complicated Eq. 39. The equivalent equation in continuous variables is most easily obtained from Eq. 45:

$$\left(\frac{d}{dt} - D\nabla_1^2 - D\nabla_2^2\right)\langle n(\mathbf{r}_1)n(\mathbf{r}_2)\rangle =$$

$$2D\nabla_1\cdot\nabla_2\left\{\delta(\mathbf{r}_1-\mathbf{r}_2)\langle n(\mathbf{r}_1)\rangle\right\}. \qquad (46)$$

In Appendix C, this result is shown to agree with the one obtained by Van der Ziel [34] and Van Vliet [32] using the Langevin approach.

Electrical conduction is often described as a diffusion process biased by an external field $\mathbf{F}$. The macroscopic equation for the electron density then obeys

$$\frac{\partial n}{\partial t} = -\mu\mathbf{F}\cdot\nabla n + D\nabla^2 n, \qquad (47)$$

where μ is the mobility while charge effects have been ignored. The same Eqs. 43 and 44 remain valid, but the operator $\mathbb{W}$ is now to be identified with $-\mu\mathbf{F}\cdot\nabla + D\nabla^2$. Hence

$$\frac{d[n(\mathbf{r}_1)n(\mathbf{r}_2)]}{dt} = \qquad (48)$$

$$\left\{-\mu\mathbf{F}\cdot(\nabla_1 + \nabla_2) + D\left(\nabla_1^2 + \nabla_2^2\right)\right\}[n(\mathbf{r}_1)n(\mathbf{r}_2)].$$

Rewriting this in terms of the second moment, one obtains

$$\left\{\frac{d}{dt} + \mu\mathbf{F}\cdot(\nabla_1 + \nabla_2) - D\left(\nabla_1^2 + \nabla_2^2\right)\right\}\langle n(\mathbf{r}_1)n(\mathbf{r}_2)\rangle =$$

$$= -2D\nabla_1\cdot\nabla_2\delta(\mathbf{r}_1-\mathbf{r}_2)\langle n(\mathbf{r}_1)\rangle. \qquad (49)$$

The term on the right is identical with the one in Eq. 46. The fact that it turns out to be unaffected by the field constitutes a justification for considering it as a noise source inherent in the diffusion process. The mathematical reason why the field does not affect the noise is seen on performing the algebra that leads from Eq. 48 to Eq. 49: the field term in $\mathbb{W}$ involves a first rather than a second derivative.

VI.   CONDUCTION BETWEEN ELECTRODES

The Eqs. 47 and 48 for electrical conduction cannot be solved unless one knows the boundary conditions. They are provided by the nature of the surface of the material, in particular the electrodes attached to it. For simplicity, we take a long wire with two electrodes at the ends, so that the problem may be treated as one-dimensional. The precise effect of the electrodes depends on the physical properties of the contact. We choose a simple model for it in order to minimize the algebra [35].

The wire, $0 < x < L$, is subdivided in cells of length Δ with occupation numbers N_λ with $\lambda = 0, 1, 2, \ldots, \Lambda = L/(N+1)$. The probability distribution $P(\{N_\lambda\})$ obeys a master equation whose main term is Eq. refeq:5.1, where $\mathbb{W}$ has the form given in Eq. 47. Additional terms arise from the ends, where the electrons spill over from the electrodes and also disappear into them. The end at $x = 0$ is responsible for two additional terms in the master equation,

$$\Delta \sum_\lambda b_\lambda \left(\mathbf{E}_\lambda^{-1} - 1 \right) P + \sum_\lambda a_\lambda \left(\mathbf{E}_\lambda - 1 \right) N_\lambda P.$$

They are similar to Eq. 31, but the coefficients for creation and annihilation now depend on the cell λ, and are practically zero for cells that are more than a few mean free paths from the end. Adding the same term for the other end one obtains the master equation,

$$\frac{dP(\{N_\lambda\})}{dt} = \sum_{\lambda,\mu} w_{\lambda\mu} \left(\mathbf{E}_\lambda^{-1} \mathbf{E}_\mu - 1 \right) N_\mu P +$$

$$+ \Delta \sum_\lambda b_\lambda \left(\mathbf{E}_\lambda^{-1} - 1 \right) P + \sum_\lambda a_\lambda \left(\mathbf{E}_\lambda - 1 \right) P +$$

$$+ \Delta \sum_\lambda b_{\Lambda-\lambda} \left(\mathbf{E}_\lambda^{-1} - 1 \right) P + \sum_\lambda a_{\Lambda-\lambda} \left(\mathbf{E}_\lambda - 1 \right) P. \quad (50)$$

First compute the rate equation for the average:

$$\frac{d\langle N_\alpha \rangle}{dt} = \sum_\mu \mathbb{W}_{\alpha\mu} \langle N_\mu \rangle + \Delta \left(b_\alpha - b_{\Lambda-\alpha} \right) -$$

$$- \left(a_\alpha - a_{\Lambda-\alpha} \right) \langle N_\alpha \rangle.$$

In the continuous description

$$\frac{\partial \langle n(x) \rangle}{\partial t} = -\mu F \frac{\partial \langle n(x) \rangle}{\partial x} + D \frac{\partial^2 \langle n(x) \rangle}{\partial x^2} + \quad (51)$$

$$+ \{ b(x) + b(L-x) \} - \{ a(x) + a(L-x) \} \langle n(x) \rangle.$$

In principle this equation can be solved if $b(x)$ and $a(x)$ are known. It is clear, however, that the precise form of these functions cannot be important for the actual problem since they have a very short range. To utilize that fact, we let their range go to zero, and at the same time increase their magnitude so as to not loose their

effect altogether. More precisely, we take

$$b(x) = \frac{1}{\varepsilon} b, \quad a(x) = \frac{1}{\varepsilon} a, \quad (0 < x < \varepsilon);$$

$$b(x) = 0, \quad a(x) = 0, \quad (x > \varepsilon). \quad (52)$$

In the limit $\varepsilon \to 0$, the result is that the boundary values of $\langle n(x) \rangle$ are fixed,

$$\langle n(0) \rangle = \langle n(L) \rangle = b/a \equiv \rho.$$

Thus the effect of our electrodes on the macroscopic diffusion equation is simply to assign fixed boundary values to $n(x)$.

The solution of the equation consisting of the first line of Eq. 51 with boundary values ρ is a standard problem. The stationary solution is particularly easy: clearly $\langle n(x) \rangle = \rho$ obeys all requirements. That gives for the average electrical current the familiar expression

$$\langle J \rangle = e\mu F \langle n \rangle - eD \frac{d\langle n \rangle}{dx} = e\mu F \rho.$$

Of course, knowing that the Boltzmann equilibrium $\langle n \rangle = \exp(eFx/kT)$ carries no current, one finds from this $eD = \mu kT$. (This should have come out automatically if we had specified the transition probabilities $w_{\lambda\mu}$ in Section V, but we have taken a shortcut by replacing the matrix $\mathbb{W}$ rightaway with the differential operator Eq. 51.)

The fluctuations obey Eq. 48, or, in one dimension,

$$\frac{\partial [n(x_1)n(x_2)]}{\partial t} = \quad (53)$$

$$\left\{ -\mu F \left(\frac{\partial}{\partial x_1} + \frac{\partial}{\partial x_2} \right) + D \left(\frac{\partial^2}{\partial x_1^2} + \frac{\partial^2}{\partial x_2^2} \right) \right\} [n(x_1)n(x_2)].$$

To find the proper boundary conditions, however, it is necessary to return to the master equation, (50). One obtains in the usual way, compare Eqs. 40 and 32,

$$\frac{d[N_\alpha N_\beta]}{dt} = \sum_\mu \mathbb{W}_{\alpha\mu}[N_\mu N_\beta] + \sum_\mu \mathbb{W}_{\beta\mu}[N_\alpha N_\mu] -$$

$$- \left(a_\alpha + a_\beta + a_{\Lambda-\alpha} + a_{\Lambda-\beta} \right) [N_\alpha N_\beta].$$

Even without writing this out in the continuous description one sees that the last term has no effect unless at least one of the cells $\alpha, \beta, \Lambda - \alpha, \Lambda - \beta$ is near a boundary. In that case, however, the corresponding coefficient is large, of order α/ε, so that $[N_\alpha N_\beta]$ goes to zero in a negligibly short time. Hence the limit $\varepsilon \to 0$ in Eq. 52 leads to the boundary values

$$[n(0)n(x)] = [n(L)n(x)] = 0.$$
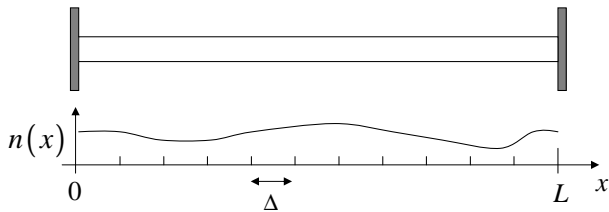
Clearly, with these boundary conditions the only station-

FIG. 2: **Electron transport through a long wire.** The wire is subdivided into cells of length Δ. The number of particles in each cell λ is N_λ. In the continuum limit ($\Delta \to 0$), we replace the discrete index λ by a continuously varying function $N_\lambda \to n(x)$.

ary solution of Eq. 53 is $[n(x_1)n(x_2)] = 0$, so that

$$\langle n(x_1)n(x_2)\rangle = \langle n(x_1)\rangle\langle n(x_2)\rangle +$$
$$\delta(x_1 - x_2)\langle n(x_1)\rangle. \tag{54}$$

This result states that the density fluctuations are Poissonian, just as in Eq. 35. This is not surprising because the electrons are injected and retrieved by the electrodes independently from each other.

Thus we have found the two-point correlations of the density of the charge carriers. Unfortunately this is not enough information for finding the current fluctuations. For that purpose, one needs to know the velocity of the carriers, so that a description in terms of the phase space density $n(\mathbf{r}, \mathbf{v})$ is required. Its average obeys the flow equation

$$\left(\frac{\partial}{\partial t} + \mathbf{v} \cdot \frac{\partial}{\partial \mathbf{r}} + \mathbf{F} \cdot \frac{\partial}{\partial \mathbf{v}}\right)\langle \mathbf{n}(\mathbf{r}, \mathbf{v})\rangle = \mathbb{W}\langle \mathbf{n}(\mathbf{r}, \mathbf{v})\rangle,$$

where $\mathbb{W}$ is the same collision operator as in the next section. It would therefore be necessary to tie up this flow in $\mathbf{r}, \mathbf{v}$ space with the diffusion equation in $\mathbf{r}$ space. This task will not be undertaken here [15, 16, 36].

VII. THE LORENTZ GAS

Consider a classical gas of N identical and independent molecules, each endowed with a velocity $\mathbf{v}$ and having a probability $w(\mathbf{v}'|\mathbf{v})$ per unit time to change to a velocity $\mathbf{v}'$ through the collision with a scatterer. At first we suppose that the scatterers are also moving, so that in general $|\mathbf{v}'| \neq |\mathbf{v}|$. The macroscopic equation for the density $n(\mathbf{v})$ in velocity space is

$$\frac{n(\mathbf{v})}{dt} = \int \{w(\mathbf{v}|\mathbf{v}')n(\mathbf{v}') - w(\mathbf{v}'|\mathbf{v})n(\mathbf{v})\} \, d\mathbf{v}'$$
$$= \int \mathbb{W}(\mathbf{v}|\mathbf{v}')n(\mathbf{v}')d\mathbf{v}'. \tag{55}$$

This model has been employed for describing electrons in a semiconductor. The solution for given initial density

$n_0(\mathbf{v})$ is formally

$$n(\mathbf{v}, t) = e^{\mathbb{W}t}n_0(\mathbf{v}).$$

As Eq. 55 has the form of a master equation, we may conclude that all solutions tend to an equilibrium, which is the same function $\chi(\mathbf{v})$ for all, apart from normalization. Taking $\chi(\mathbf{v})$ normalized according to $\int \chi(\mathbf{v})d\mathbf{v} = 1$, one has

$$e^{\mathbb{W}t}n_0 \to \chi(\mathbf{v})N = \chi(\mathbf{v})\int n_0(\mathbf{v}')d\mathbf{v}'. \tag{56}$$

Thus $e^{\mathbb{W}t}$ tends to a projection operator[43].

In order to describe the fluctuations around the macroscopic behavior, we subdivide velocity space in cells Δ, with occupation numbers N_λ and write the master equation for $P(\{N_\lambda\})$. It is the same Eq. 36 as for diffusion in coordinate space. Hence, we can skip the various steps and write immediately the equation for the average by substituting $\mathbf{v}$ for $\mathbf{r}$ in Eq. 43:

$$\frac{d\langle n(\mathbf{v})\rangle}{dt} = \int \mathbb{W}(\mathbf{v}|\mathbf{v}')\langle n(\mathbf{v}')\rangle d\mathbf{v}'. \tag{57}$$

This has, of course, the same form as the macroscopic Eq. 55. Similarly, the equation for the fluctuations can be read off from Eq. 44:

$$\frac{d[n(\mathbf{v}_1)n(\mathbf{v}_2)]}{dt} = \int \mathbb{W}(\mathbf{v}_1|\mathbf{v}')[n(\mathbf{v}')n(\mathbf{v}_2)]d\mathbf{v}' +$$
$$\int \mathbb{W}(\mathbf{v}_2|\mathbf{v}')[n(\mathbf{v}_1)n(\mathbf{v}')]d\mathbf{v}'. \tag{58}$$

In principle this solves the problem of finding the fluctuations in an arbitrary time-dependent situation, but one cannot make the result more explicit without solving Eq. 57. The fluctuations in equilibrium, however, can readily be found. Equation 58 has the from of Eq. 41 and is solved formally by

$$\Theta(t) = e^{\mathbb{W}t} \cdot \Theta(0) \cdot e^{\widetilde{\mathbb{W}}t}, \tag{59}$$

as can be checked by direct substitution. With the aid of Eq. 56, one obtains for $t \to \infty$,

$$\Theta(\infty) = [n(\mathbf{v}_1)n(\mathbf{v}_2)]^{eq}$$
$$= \chi(\mathbf{v}_1)\chi(\mathbf{v}_2)\int\int[n(\mathbf{v}_1')n(\mathbf{v}_2')]_0 d\mathbf{v}_1'd\mathbf{v}_2'$$
$$= \chi(\mathbf{v}_1)\chi(\mathbf{v}_2)N.$$

This gives for the covariance of the density

$$\langle\langle n(\mathbf{v}_1)n(\mathbf{v}_2)\rangle\rangle^{eq} = N\{\delta(\mathbf{v}_1 - \mathbf{v}_2)\chi(\mathbf{v}_1) - \chi(\mathbf{v}_1)\chi(\mathbf{v}_2)\}, \tag{60}$$

and for the second moment

$$\langle n(\mathbf{v}_1)n(\mathbf{v}_2)\rangle^{eq} = \left(1 - N^{-1}\right)\langle n(\mathbf{v}_1)\rangle^{eq}\langle n(\mathbf{v}_2)\rangle^{eq}$$
$$+\delta(\mathbf{v}_1 - \mathbf{v}_2)\langle n(\mathbf{v}_1)\rangle^{eq}. \quad (61)$$

This result agrees with the familiar counting argument of equilibrium statistical mechanics. Subdivide velocity space cells λ each having an *a priori* probability p_λ. Distribute the N molecules over these cells; the probability for a set of occupation numbers $\{N_\lambda\}$ with $\sum N_\lambda = N$ is

$$\frac{N!}{N_1!N_2!\dots} p_1^{N_1}p_2^{N_2}\dots = N!\prod_\lambda \frac{p_\lambda^{N_\lambda}}{N_\lambda!}.$$

The average $\langle N_\alpha\rangle$ is Np_α and

$$\langle N_\alpha N_\beta\rangle = N(N - 1)p_\alpha p_\beta + \delta_{\alpha,\beta}Np_\alpha$$
$$= \left(1 - N^{-1}\right)\langle N_\alpha\rangle\langle N_\beta\rangle + \delta_{\alpha,\beta}\langle N_\alpha\rangle,$$

in agreement with Eq. 61. It deviates from the Poisson distribution because of the term involving $1/N$, which is due to the fact that the total number is fixed.

Now modify the model by supposing that *the scatterers are fixed.* As a consequence no transitions occur with $|\mathbf{v}| \neq |\mathbf{v}'|$, so that

$$\mathbb{W}(\mathbf{v}|\mathbf{v}') = v^{-2}\delta(v - v')\mathbb{W}_v(\Omega/\Omega'), \quad (62)$$

where Ω is the unit vector $\mathbf{v}/v$. The factor v^{-2} has been inserted for convenience in normalizing, in agreement with

$$\delta(\mathbf{v} - \mathbf{v}') = v^{-2}\delta(v - v')\delta(\Omega - \Omega'). \quad (63)$$

We set $N_v dv$ for the number of molecules between v and $v + dv$,

$$N_v = \int \delta(v - |\mathbf{v}|)n(\mathbf{v}')d\mathbf{v}'.$$

Equation 55 no longer has a unique equilibrium solution, because the collisions are unable to alter N_v. Assuming that there are no other constants of the motion, one has an equilibrium distribution $\chi(v, \Omega)$ for each separate v, which is of course independent of the direction Ω, and properly normalized equals $1/4\pi$. Hence in the limit

$$e^{\mathbb{W}t}n_0(\mathbf{v}) \rightarrow \frac{N_v}{4\pi} = \frac{1}{4\pi}\int \delta(v - v')n_0(\mathbf{v}')d\mathbf{v}',$$

which is again a projection operator. Thus $\langle n(\mathbf{v})\rangle(t)$ tends to $\langle n(\mathbf{v})\rangle_\infty = N_v/4\pi$.

The solution Eq. 59 for Θ yields in the limit

$$[n(\mathbf{v}_1)n(\mathbf{v}_2)]_\infty =$$
$$= -(4\pi)^{-2}\int \delta(v_1 - v_1')d\mathbf{v}_1' \times$$
$$\times \int \delta(v_2 - v_2')\delta(\mathbf{v}_1' - \mathbf{v}_2')n_0(\mathbf{v}_2')d\mathbf{v}_2'$$
$$= -(4\pi)^{-2}\delta(v_1 - v_2)N_{v_1}.$$

Hence, using Eq. 63,

$$\langle\langle n(\mathbf{v}_1)n(\mathbf{v}_2)\rangle\rangle_\infty =$$
$$-(4\pi)^{-2}\delta(v_1 - v_2)N_{v_1} + (4\pi)^{-1}\delta(\mathbf{v}_1 - \mathbf{v}_2)N_{v_1}$$
$$= \delta(v_1 - v_2)N_{v_1}\left\{\frac{1}{4\pi v_1^2} - \frac{1}{(4\pi)^2}\right\}$$

This is analogous to Eq. 60, but has a delta function in front, which states that no correlation exists between subspaces belonging to different v. The second moment can also be written in a form analogous to Eq. 61,

$$\langle n(\mathbf{v}_1)n(\mathbf{v}_2)\rangle_\infty = \left\{1 - \frac{\delta(v_1 - v_2)}{N_{v_1}}\right\}\langle n(\mathbf{v}_1)\rangle_\infty\langle n(\mathbf{v}_2)\rangle_\infty +$$
$$+\delta(\mathbf{v}_1 - \mathbf{v}_2)\langle n(\mathbf{v}_1)\rangle_\infty.$$

VIII. THE EXPANSION OF THE NONLINEAR MASTER EQUATION

So far the examples were all governed by *linear* master equations, from which closed equations could be obtained for the first and second moments. As a first example of a *nonlinear* master equation, we take the simple chemical reaction

$$B \xrightarrow{b} X, \quad 2X \xrightarrow{a} A. \quad (64)$$
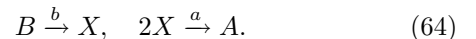
The compound A may well be the molecule X_2, provided that it doeas not dissociate again. This model has been the subject of some discussion [23–26, 30, 31]. In the homogeneous stationary state, the fluctuations in the total number of molecules X can be calculated without difficulty [1], but we are now in a position to compute the spatial correlations between local fluctuations as well.

Subdivide the total volume into cells Δ, with occupation numbers N_λ. The master equation for the probability distribution $P(\{N_\lambda\})$ is, ignoring transport,

$$\frac{dP(\{N_\lambda\})}{dt} = b\Delta\sum_\lambda \left(\mathbf{E}_\lambda^{-1} - 1\right)P - a\Delta^{-1}\sum_\lambda \left(\mathbf{E}_\lambda^2 - 1\right)N_\lambda^2 P. \quad (65)$$

The first term is the same as in Eq. 31. The second term expresses that each of the N_λ molecules X in cell λ has a probability proportional to N_λ/Δ to meet another one in unit time. Actually, the number of pairs is of course $\frac{1}{2}N_\lambda(N_\lambda - 1)$, but the 1 is immaterial in the order that we

shall calculate [1, 30], and the factor $\frac{1}{2}$ has been absorbed in the reaction rate a, as is customary when using the law of mass action.

It is no longer possible to deduce from Eq. 65 exact closed equations for the first and second moments, but an expansion method exists, based on the idea that N_λ is large [2–8]. Note that it is therefore no longer possible to take Δ arbitrarily small. Nevertheless, the language of continuously varying variables can be employed, provided one does not claim to describe fluctuations on a scale comparable with the mean distance between molecules X. We shall now demonstrate this expansion method on Eq. 65.

Transform from the variable N_λ to new variables ζ_λ by setting

$$N_\lambda = \Delta\phi_\lambda(t) + \Delta^{1/2}\zeta_\lambda, \qquad (66)$$

where the ϕ_λ are functions of time to be specified presently. This substitution decomposes N_λ into a *macroscopic part* proportional to Δ and a *fluctuating part* proportional to $\Delta^{1/2}$. In chemical parlance, ϕ_λ is the concentration.

The distribution function $P(\{N_\lambda\}, t)$ transforms into a distribution $\Pi(\{\zeta_\lambda\}, t)$,

$$P(\{\Delta\phi_\lambda(t) + \Delta^{1/2}\zeta_\lambda\}, t) = \Delta^{-\frac{\Lambda}{2}}\Pi(\{\zeta_\lambda\}, t).$$

The factor $\Delta^{-\frac{\Lambda}{2}}$, where Λ is the total number of cells, is needed for the normalization of Π, but will not enter into the equations. The operator $\mathbf{E}_\lambda$ acting on an arbitrary function $f(\zeta_\lambda)$ takes the form

$$\mathbf{E}_\lambda f(\zeta_\lambda) = f(\zeta_\lambda + \Delta^{-1/2})$$
$$= \left\{ 1 + \Delta^{-1/2}\frac{\partial}{\partial\zeta_\lambda} + \frac{\Delta^{-1}}{2}\frac{\partial^2}{\partial\zeta_\lambda^2} + \dots \right\} f(\zeta_\lambda).$$

Rewriting the master Eq. 65 in the new variables one obtains

$$\frac{\partial\Pi}{\partial t} - \Delta^{1/2}\sum_\lambda \frac{d\phi_\lambda}{dt}\frac{\partial\Pi}{\partial\zeta_\lambda} = b\Delta^{\frac{1}{2}}\sum_\lambda \left\{ -\frac{\partial}{\partial\zeta_\lambda} + \frac{\Delta^{-\frac{1}{2}}}{2}\frac{\partial^2}{\partial\zeta_\lambda^2} \right\}\Pi$$
$$+ a\Delta^{\frac{1}{2}}\sum_\lambda 2\left\{ \frac{\partial}{\partial\zeta_\lambda} + \Delta^{-\frac{1}{2}}\frac{\partial^2}{\partial\zeta_\lambda^2} \right\}\left(\phi_\lambda + \Delta^{-\frac{1}{2}}\zeta_\lambda \right)^2 \Pi. \qquad (67)$$

It is now possible to separate the successive orders in Δ. Terms of higher order in $\Delta^{-\frac{1}{2}}$ than we are interested in have already been omitted in Eq. 67.

The terms of order $\Delta^{\frac{1}{2}}$ are all proportional to $\partial\Pi/\partial\zeta_\lambda$ and can therefore be caused to cancel by setting (for each λ)

$$-\frac{d\phi_\lambda}{dt} = -b + 2a\phi_\lambda^2. \qquad (68)$$

These equations determine the behavior of the macro-

scopic parts of the N_λ and, in fact, they are the familiar rate equations for Eq. 64. Remember that transfer between cells is till ignored.

The terms of order Δ^0 in Eq. 67 are

$$\frac{\partial\Pi}{\partial t} = \frac{b}{2}\sum_\lambda \frac{\partial^2\Pi}{\partial\zeta_\lambda^2} + 2a\sum_\lambda \phi_\lambda^2 \frac{\partial^2\Pi}{\partial\zeta_\lambda^2} +$$
$$+ 4a\sum_\lambda \phi_\lambda \frac{\partial}{\partial\zeta_\lambda}\zeta_\lambda\Pi. \qquad (69)$$

This is a multivariate Fokker-Planck equation with coefficients that are linear in the ζ_λ, but will depend on time through the ϕ_λ. It can readily be solved [44], but all we need are the first and second moments of the ζ_λ. One easily deduces

$$\frac{d\langle\zeta_\alpha\rangle}{dt} = -4a\phi_\alpha(t)\langle\zeta_\alpha\rangle, \qquad (70)$$

and

$$\frac{d\langle\zeta_\alpha\zeta_\beta\rangle}{dt} = -4a\left(\phi_\alpha + \phi_\beta\right)\langle\zeta_\alpha\zeta_\beta\rangle + \delta_{\alpha,\beta}\left(b + 4a\phi_\alpha^2\right). \qquad (71)$$

In Eq. 69 – and therefore also in Eqs. 70 and 71 – terms of relative order $\Delta^{-\frac{1}{2}}$ are neglected. Note that Eq. 70 is the *variational equation* associated with Eq. 68.

Substituting the results Eqs. 68 and 70 into Eq. 66, one obtains

$$\frac{d\langle N_\alpha\rangle}{dt} = b\Delta - 2a\Delta^{-1}\langle N_\alpha\rangle^2 + O(\Delta^0). \qquad (72)$$

In addition, one obtains from Eq. 71 an equation, which is more easily written in terms of the covariance matrix,

$$\frac{d\langle\langle N_\alpha N_\beta\rangle\rangle}{dt} = -4a\Delta^{-1}\left(\langle N_\alpha\rangle + \langle N_\beta\rangle\right)\langle\langle N_\alpha N_\beta\rangle\rangle +$$
$$+ \delta_{\alpha,\beta}\left\{ b\Delta + 4a\Delta^{-1}\langle N_\alpha\rangle^2 \right\} + O(\Delta^{\frac{1}{2}}).$$

In order to write these equations with continuous space variables so as to eliminate the cell size Δ, we divide them by Δ and Δ^2, respectively, and obtain

$$\frac{d\langle n(\mathbf{r})\rangle}{dt} = b - 2a\langle n(\mathbf{r})\rangle^2, \qquad (73)$$

and

$$\frac{d\langle\langle n(\mathbf{r}_1)n(\mathbf{r}_2)\rangle\rangle}{dt} = -4a\left\{\langle n(\mathbf{r}_1)\rangle + \langle n(\mathbf{r}_2)\rangle\right\}\langle\langle n(\mathbf{r}_1)n(\mathbf{r}_2)\rangle\rangle +$$
$$+ \delta(\mathbf{r}_1 - \mathbf{r}_2)\left\{ b + 4a\langle n(\mathbf{r}_1)\rangle^2 \right\}. \qquad (74)$$

The equation for the factorial cumulant is

$$\frac{d[n(\mathbf{r}_1)n(\mathbf{r}_2)]}{dt} = -4a\left\{\langle n(\mathbf{r}_1)\rangle + \langle n(\mathbf{r}_2)\rangle\right\}[n(\mathbf{r}_1)n(\mathbf{r}_2)] -$$
$$- 2a\delta(\mathbf{r}_1 - \mathbf{r}_2)\langle n(\mathbf{r}_1)\rangle^2. \qquad (75)$$

It is clear that the use of factorial cumulants no longer gives a worthwhile simplification. Yet Eq. 75 is needed for our next task: compounding these equations with the flow.

IX. THE SAME REACTION WITH DIFFUSION

It is now possible to add the effect of transfer between cells. We take the case of transfer by diffusion. These Eqs. 73 and 75 are now extended into

$$\frac{d\langle n(\mathbf{r})\rangle}{dt} = b - 2a\langle n(\mathbf{r})\rangle^2 + D\nabla^2\langle n(\mathbf{r})\rangle, \qquad (76)$$

and,

$$\frac{d[n(\mathbf{r}_1)n(\mathbf{r}_2)]}{dt} =$$
$$\{(-4a\langle n(\mathbf{r}_1)\rangle + \langle n(\mathbf{r}_2)\rangle) + D\left(\nabla_1^2 + \nabla_2^2\right)\} \times$$
$$\times [n(\mathbf{r}_1)n(\mathbf{r}_2)] - 2a\delta(\mathbf{r}_1 - \mathbf{r}_2)\langle n(\mathbf{r}_1)\rangle^2. \qquad (77)$$

As it does not seem possible to obtain explicit solutions of these time dependent equations, we must apply them to the *homogeneous stationary* case. First one gets from Eq. 76

$$\langle n\rangle^{st} = \sqrt{b/2a},$$

so that Eq. 77 becomes

$$D\left(\nabla_1^2 + \nabla_2^2\right)[n(\mathbf{r}_1)n(\mathbf{r}_2)] =$$
$$4\sqrt{2ab}\,[n(\mathbf{r}_1)n(\mathbf{r}_2)] + b\delta(\mathbf{r}_1 - \mathbf{r}_2). \qquad (78)$$

A particular solution can be obtained by taking for $[n(\mathbf{r}_1)n(\mathbf{r}_2)]$ a function $\vartheta(\mathbf{r})$ of $\mathbf{r} = \mathbf{r}_1 - \mathbf{r}_2$ alone, which then must satisfy

$$\nabla^2\vartheta(\mathbf{r}) = \kappa^2\vartheta(\mathbf{r}) + (b/2D)\delta(\mathbf{r}),$$

where $\kappa^2 = 2\sqrt{2ab}/D$. Consequently

$$\vartheta(\mathbf{r}) = -\frac{b}{8\pi D}\,\frac{e^{-\kappa r}}{r}, \qquad (79)$$

is a solution of Eq. 78. Other solutions differ from it by a solution of the homogeneous equation and it is easily seen that they have to be excluded since they grow exponentially with $|\mathbf{r}_1|$ and $|\mathbf{r}_2|$.

Thus the fluctuations in the homogeneous stationary state are determined by Eq. 79, or in the more familiar terms of the covariance

$$\langle\langle n(\mathbf{r}_1)n(\mathbf{r}_2)\rangle\rangle = -\frac{b}{8\pi D}\,\frac{e^{-\kappa|\mathbf{r}_1-\mathbf{r}_2|}}{|\mathbf{r}_1-\mathbf{r}_2|} + \sqrt{\frac{b}{2a}}\,\delta(\mathbf{r}_1 - \mathbf{r}_2)$$
$$= \langle n\rangle^{st}\left\{\delta(\mathbf{r}_1 - \mathbf{r}_2) - \frac{1}{4}\,\frac{\kappa^2 e^{-\kappa|\mathbf{r}_1-\mathbf{r}_2|}}{4\pi|\mathbf{r}_1-\mathbf{r}_2|}\right\}. \qquad (80)$$

The rate at which a given molecule X disappears into A is seen from Eq. 72 to be $2a\langle n\rangle^{st} = \sqrt{2ab}$. Hence κ^{-1} measures the distance over which it diffuses before decayng. We now apply Eq. 80 to two limiting cases.

Take a region V whose linear dimensions are much larger than κ^{-1}. The variance of the number N_V of molecules X in that region is

$$\langle\langle N_V^2\rangle\rangle^{st} = \int_V\int_V\langle\langle n(\mathbf{r}_1)n(\mathbf{r}_2)\rangle\rangle d\mathbf{r}_1 d\mathbf{r}_2 \qquad (81)$$

$$= \langle n\rangle^{st}\left\{1 - \frac{1}{4}\right\}V = \frac{3}{4}\langle N_V\rangle^{st}. \qquad (82)$$

This is the same result as obtained by Nitzan and Ross [30], and by Kuramoto [26], although they did not use a systematic expansion. The effect of diffusion has disappeared, as was to be expected since most of the molecules produced in V have no time to diffuse away. The fact that the variance is less than that of a Poisson distribution can be understood on the grounds that molecules are not independent, but are annihilated in pairs [1].

On the other hand, when V is small compared to κ^{-1} the diffusion is important, but its effect on the fluctuations depends on the precise shape of V. Yet an estimate of the integral Eq. 81 can be obtained

$$\langle n\rangle^{st}\left\{V - \frac{\kappa^2}{16\pi}\,\frac{V^2}{L}\right\} = \langle N_V\rangle^{st}\left\{1 - \frac{a}{4\pi L}\langle N_V\rangle^{st}\right\},$$

where L is of the order of the diameter of V. It is seen that when the size of V is reduced this tends to $\langle N_V\rangle^{st}$, in agreement with the Poisson distribution. In this situation the fluctuations are mainly determined by the diffusion, so that the interaction between the molecules becomes irrelevant.

This distinction between large and small V was pointed out by Kuramoto [31], albeit that he used D as a parameter to separate both cases. Also he expressed his results in the Fourier transform of the variance, for which we find from Eq. 80

$$\int\langle\langle n(\mathbf{r}_1)n(\mathbf{r}_1 + \mathbf{r})\rangle\rangle^{st}e^{i\mathbf{q}\cdot\mathbf{r}}d\mathbf{r} = \langle n\rangle^{st}\frac{3 + 4q^2/\kappa^2}{4 + 4q^2/\kappa^2}.$$

This is identical with equation (4) in reference [26].

X. A POPULATION PROBLEM

The Malthus-Verhulst equation for a population of N individuals is [37–39]

$$\frac{dN}{dt} = bN - aN - cN^2/\Omega. \qquad (83)$$

The parameter b is the natural birth rate per individual, a is the death rate, and cN/Ω the additional death rate due to overcrowding, which is taken to be propor-

tional to the density N/Ω of the population present, Ω being the size of the habitat. This is of course the macroscopic equation, which does not account for the fluctuations caused by the statistical nature of the individual birth and death events. They are described by the master equation for this problem, which can easily be constructed without any other assumptions than those used in arriving at Eq. 83.

So far the total population N was considered. We now want to include the effect of migration and therefore have to divide Ω in cells Δ with their own population numbers N_λ. The resulting multivariate master equation is easily seen to be

$$\frac{dP(\{N_\lambda\})}{dt} = b \sum_\lambda \left(\mathbf{E}_\lambda^{-1} - 1\right) N_\lambda P +$$

$$\sum_\lambda \left(\mathbf{E}_\lambda - 1\right) \left\{aN_\lambda + cN_\lambda^2/\Delta\right\} N_\lambda P. \quad (84)$$

We proceed to deduce the equations for the first and second moments, which will enable us to add the migration terms.

As Eq. 84 is nonlinear, the transformation Eq. 66 has to be carried out. The result is a master equation for $\Pi(\{\zeta_\lambda\}, t)$ similar to Eq. 67

$$\frac{\partial \Pi}{\partial t} - \Delta^{1/2} \sum_\lambda \frac{d\phi_\lambda}{dt} \frac{\partial \Pi}{\partial \zeta_\lambda} =$$

$$b\Delta \sum_\lambda \left\{-\Delta^{-\frac{1}{2}} \frac{\partial}{\partial \zeta_\lambda} + \frac{\Delta^{-1}}{2} \frac{\partial^2}{\partial \zeta_\lambda^2}\right\} \Pi$$

$$+\Delta \sum_\lambda \left\{\Delta^{-\frac{1}{2}} \frac{\partial}{\partial \zeta_\lambda} + \Delta^{-1} \frac{\partial^2}{\partial \zeta_\lambda^2}\right\} \times$$

$$\times \left\{a\phi_\lambda + c\phi_\lambda^2 + \Delta^{-\frac{1}{2}} \left(a + 2c\phi_\lambda\right) \zeta_\lambda\right\} \Pi.$$

The terms proportional to $\Delta^{\frac{1}{2}}$ are caused to cancel by choosing for the ϕ_λ solutions of

$$-\frac{d\phi_\lambda}{dt} = -b\phi_\lambda + a\phi_\lambda + c\phi_\lambda^2. \quad (85)$$

The terms proportional to Δ^0 give rise to a linear, multivariate, time-dependent Fokker-Planck equation for Π,

$$\frac{\partial \Pi}{\partial t} = \sum_\lambda \left(-b + a + 2c\phi_\lambda\right) \frac{\partial}{\partial \zeta_\lambda} \zeta_\lambda \Pi +$$

$$\frac{1}{2} \sum_\lambda \left(b\phi_\lambda + a\phi_\lambda + c\phi_\lambda^2\right) \frac{\partial^2 \Pi}{\partial \zeta_\lambda^2}. \quad (86)$$

The equations for the averages are

$$\frac{d\langle \zeta_\lambda \rangle}{dt} = (b - a - 2c\phi_\lambda) \langle \zeta_\lambda \rangle. \quad (87)$$

They are the variational equations associated with

Eq. 85. Together with Eq. 85 they give

$$\frac{d\langle N_\lambda \rangle}{dt} = (b - a)\langle N_\lambda \rangle - \frac{c}{\Delta} \langle N_\lambda \rangle^2.$$

This is simply the macroscopic Eq. 83 for each separate cell. In the continuous description they take the form

$$\frac{d\langle n(\mathbf{r}) \rangle}{dt} = (b - a)\langle n(\mathbf{r}) \rangle - c\langle n(\mathbf{r}) \rangle^2.$$

The equation for the second moments obtained from Eq. 86 is

$$\frac{d\langle \zeta_\alpha \zeta_\beta \rangle}{dt} = 2 \left(b - a - c\phi_\alpha - c\phi_\beta\right) \langle \zeta_\alpha \zeta_\beta \rangle +$$

$$+\delta_{\alpha,\beta} \left(b\phi_\alpha + a\phi_\alpha - c\phi_\alpha^2\right).$$

The same equation is obeyed by the covariance matrix $\langle\langle \zeta_\alpha \zeta_\beta \rangle\rangle$ and one subsequently finds

$$\frac{d\langle\langle N_\alpha N_\beta \rangle\rangle}{dt} = 2 \left(b - a - c\langle N_\alpha \rangle/\Omega - c\langle N_\beta \rangle/\Omega\right) \langle\langle N_\alpha N_\beta \rangle\rangle +$$

$$+\delta_{\alpha,\beta} \left(b\phi_\alpha + a\phi_\alpha - c\phi_\alpha^2\right).$$

In the continuous description

$$\frac{d\langle\langle n(\mathbf{r}_1) n(\mathbf{r}_2) \rangle\rangle}{dt} =$$

$$2 \left(b - a - c\langle n(\mathbf{r}_1) \rangle - c\langle n(\mathbf{r}_2) \rangle\right) \langle\langle n(\mathbf{r}_1) n(\mathbf{r}_2) \rangle\rangle +$$

$$+\delta(\mathbf{r}_1 - \mathbf{r}_2)\langle n(\mathbf{r}_1) \rangle \left(b + a - c\langle n(\mathbf{r}_1) \rangle\right). \quad (88)$$

We also need the equation for the factorial cumulants,

$$\frac{d[n(\mathbf{r}_1) n(\mathbf{r}_2)]}{dt} =$$

$$2 \left(b - a - c\langle n(\mathbf{r}_1) \rangle - c\langle n(\mathbf{r}_2) \rangle\right) [n(\mathbf{r}_1) n(\mathbf{r}_2)] +$$

$$+\delta(\mathbf{r}_1 - \mathbf{r}_2)\langle n(\mathbf{r}_1) \rangle \left(2b - 2c\langle n(\mathbf{r}_1) \rangle\right).$$

After this preliminary work, it is now possible to add the migration terms. We suppose that the migration occurs by diffusion and write accordingly for the total rate of change of the average

$$\frac{d\langle n(\mathbf{r}) \rangle}{dt} = (b - a)\langle n(\mathbf{r}) \rangle - c\langle n(\mathbf{r}) \rangle^2 + D\nabla^2 \langle n(\mathbf{r}_1) \rangle, \quad (89)$$

and the factorial cumulant

$$\frac{d[n(\mathbf{r}_1) n(\mathbf{r}_2)]}{dt} =$$

$$2 \left(b - a - c\langle n(\mathbf{r}_1) \rangle - c\langle n(\mathbf{r}_2) \rangle\right) [n(\mathbf{r}_1) n(\mathbf{r}_2)] +$$

$$+\delta(\mathbf{r}_1 - \mathbf{r}_2)\langle n(\mathbf{r}_1) \rangle \left(2b - 2c\langle n(\mathbf{r}_1) \rangle\right) +$$

$$+D \left(\nabla_1^2 + \nabla_2^2\right) [n(\mathbf{r}_1) n(\mathbf{r}_2)]. \quad (90)$$

The solution of these equations contains all information concerning the local average density of the population and the fluctuations about this average. Unfortunately they cannot be solved analytically and we therefore re-

strict ourselves to computing the fluctuations in the stationary state.

Suppose that the birth rate b is larger than the natural death rate a; then Eq. 89 has the stationary solution

$$\langle n(\mathbf{r})\rangle^{st} = \frac{b-a}{c}.$$

Substitution in Eq. 90 yields

$$\left\{D\left(\nabla_1^2 + \nabla_2^2\right) - 2(b-a)\right\}[n(\mathbf{r}_1)n(\mathbf{r}_2)]^{st} =$$
$$-2(b-a)(a/c)\delta(\mathbf{r}_1 - \mathbf{r}_2).$$

Observe that this equation has the same form as Eq. 78. Its solution is, in analogy with Eq. 79,

$$[n(\mathbf{r}_1)n(\mathbf{r}_2)]^{st} = \frac{a(b-a)}{4\pi cD}\,\frac{e^{-\kappa|\mathbf{r}_1 - \mathbf{r}_2|}}{|\mathbf{r}_1 - \mathbf{r}_2|},$$

where $\kappa^2 = (b-a)/D$. Consequently,

$$\langle\langle n(\mathbf{r}_1)n(\mathbf{r}_2)\rangle\rangle^{st} = \langle n\rangle^{st}\left\{\delta(\mathbf{r}_1 - \mathbf{r}_2) + \frac{a}{4\pi D}\,\frac{e^{-\kappa|\mathbf{r}_1 - \mathbf{r}_2|}}{|\mathbf{r}_1 - \mathbf{r}_2|}\right\}.$$

Note that the fluctuations are larger than for a Poisson distribution. Integration over a volume V with diameter large compared to κ^{-1} yields

$$\langle\langle N_V^2\rangle\rangle^{st} = \langle N_V\rangle^{st}\frac{b}{b-a} = \frac{b}{c}.$$

As another interesting example, Nicolis *et al.* [23] studied the Lotka-Volterra model for two interacting populations. They endowed each species with some internal "fitness parameter," which varies continuously. In their calculations, they made use of various approximations, and it would be of interest to know whether the present systematic method leads to the same results. Unfortunately, the calculations are too lengthy to be included in this article.

Appendix A: Factorial Cumulants

A probability distribution over the integers $N = 0, 1, 2, \ldots$ whose ordinary moments $\langle N^r\rangle$ exists, also has cumulants $\langle\langle N^r\rangle\rangle$ and factorial moments,

$$\langle N(N-1)(N-2)\cdots(N-r+1)\rangle, \qquad (91)$$

which are certain combinations of the moments. The *factorial cumulants* [40], $\Theta^{(r)} \equiv [N^r]$, are obtained by writing out Eq. 91 in the moments $\langle N^{r-s}\rangle$ and replacing each moment by the corresponding cumulant $\langle\langle N^{r-s}\rangle\rangle$. They are generated by the logrithm of the generating function $H(v)$ of the factorial moments,

$$\log H(v) = \log\langle(1+v)^N\rangle = \sum_{r=1}^{\infty}\frac{v^r}{r!}\Theta^{(r)}.$$

$H(v)$ is related to the characteristic function $g(t)$ by

$$H(e^{it} - 1) = G(t). \qquad (92)$$

The first factorial cumulant is the same as the first moment: $\Theta^{(1)} = \langle N\rangle$; the Poisson distribution is characterized by the vanishing of all higher $\Theta^{(r)}$.

The generalization to a multivariate distribution is trivial, but requires a more elaborate notation.

$$\log\Big\langle\prod_\lambda(1+v_\lambda)^{N_\lambda}\Big\rangle = \sideset{}{'}\sum_{\{r_\lambda\}}\frac{v_1^{r_1}v_2^{r_2}\cdots}{r_1!r_2!\cdots}\,\Theta^{(r_1, r_2, \ldots)}. \quad (93)$$

The prime indicates that the term with all r_λ equal to zero is absent. Alternatively, the right-hand side may be written

$$\sum_\lambda v_\lambda\Theta_\lambda + \frac{1}{2}\sum_{\lambda,\mu} v_\lambda v_\mu\Theta_{\lambda\mu} + \frac{1}{3!}\sum_{\lambda,\mu,\kappa} v_\lambda v_\mu v_\kappa\Theta_{\lambda\mu\kappa} + \cdots,$$

where, for instance, in agreement with Eq. 16,

$$\Theta_{\lambda\mu} \equiv [N_\lambda N_\mu] = \langle N_\lambda N_\mu\rangle - \langle N_\lambda\rangle\langle N_\mu\rangle - \delta_{\lambda,\mu}\langle N_\lambda\rangle.$$

Note that $\Theta_{\lambda\mu} = 0$ when N_λ and N_μ are uncorrelated, and in general all factorial cumulants involving groups of statistically independent variables vanish, just as ordinary cumulants. The factorial cumulant of the sum of two independent variables is the sum of their factorial cumulants; owing to Eq. 92 this follows immediately from the corresponding theorem for ordinary cumulants.

The relevance for the present work derives from the following property. The most general *linear* multivariate master equation contains four types of terms:

1. Particles λ are created in singles with probability b_λ, or in twins $b_{\lambda\mu}$, etc. The corresponding term is

$$\left\{\sum_\lambda b_\lambda(\mathbf{E}_\lambda^{-1} - 1) + \sum_{\lambda,\mu} b_{\lambda\mu}(\mathbf{E}_\lambda^{-1}\mathbf{E}_\mu^{-1} - 1) + \ldots\right\}P.$$

2. Particles are annihilated with probability a_λ per particle per unit time:

$$\sum_\lambda a_\lambda(\mathbf{E}_\lambda - 1)N_\lambda P.$$

3. Particles are transferred from μ to λ

$$\sum_{\lambda,\mu} w_{\lambda\mu}(\mathbf{E}_\lambda^{-1}\mathbf{E}_\mu - 1)N_\mu P.$$

4. Particles are created auto-catalytically (or born in litters):

$$\left\{\sum_{\lambda,\mu,\kappa} c_{\mu\kappa}^\lambda(\mathbf{E}_{\mu,\kappa}^{-1}\mathbf{E}_\lambda - 1)N_\lambda + \sum_{\lambda,\mu,\kappa,\rho} c_{\mu\kappa\rho}^\lambda(\mathbf{E}_{\mu,\kappa,\rho}^{-1}\mathbf{E}_\lambda - 1)N_\lambda\right\}P,$$

where $\mathbf{E}_{\mu,\kappa,\rho}^{-1} \equiv \mathbf{E}_\mu^{-1}\mathbf{E}_\kappa^{-1}\mathbf{E}_\rho^{-1}$ has been written as short-hand.

The solution of the master equation,

$$\frac{dP(\{N_\lambda\})}{dt} = \text{(i)} + \text{(ii)} + \text{(iii)} + \text{(iv)}, \qquad (94)$$

can be done exactly, but is facilitated by the use of the factorial cumulants.

From Eq. 94, an equation for the factorial moment generating function $H(\{v_\lambda\})$ may be derived. The term (i) yields,

$$\frac{dH}{dt} = \left\{ \sum_\lambda b_\lambda v_\lambda + \sum_{\lambda,\mu} b_{\lambda\mu} (v_\lambda + v_\mu) + \dots \right\} H.$$

Divide by H and expand the left-hand side in powers of the v. The result is a set of uncoupled equations for the several factorial cumulants, for instance

$$\frac{d\Theta_\alpha}{dt} =$$
$$b_\alpha + \sum_\mu b_{\alpha\mu} + \sum_\lambda b_{\lambda\alpha} + \sum_{\lambda,\mu} (b_{\alpha\lambda\mu} + b_{\lambda\alpha\mu} + b_{\lambda\mu\alpha}) + \cdots,$$
$$\frac{d\Theta_{\alpha\beta}}{dt} = b_{\alpha\beta} + b_{\beta\alpha} + \cdots$$

The birth term added in Section IV only involves a b_α, but no $b_{\alpha\beta}$; that is the reason why in that section $\Theta_{\alpha\beta}$ obeys the same equation as in Section II.

The term (ii) in Eq. 94 adds the contribution

$$\frac{dH}{dt} = \sum_\lambda a_\lambda \left(\frac{1}{1+v_\lambda} - 1 \right) (1 + v_\lambda) \frac{\partial H}{\partial v_\lambda}$$
$$= -\sum_\lambda a_\lambda v_\lambda \frac{\partial H}{\partial v_\lambda}.$$

It affects all factorial cumulants, but in a simple way:

$$\frac{d\Theta_{\alpha\beta\gamma\cdots}}{dt} = -(a_\alpha + a_\beta + a_\gamma + \cdots)\,\Theta_{\alpha\beta\gamma\cdots}.$$

This is the reason why in Section II, the factorial cumulants $\Theta_{\alpha\beta}$ obeyed a homogeneous equation. Similarly, term (iii) yields

$$\frac{dH}{dt} = \sum_{\lambda,\mu} w_{\lambda\mu} (v_\lambda - v_\mu) \frac{\partial H}{\partial v_\mu},$$

and couples the factorial cumulants of the same order with each other. This is the reason why in Section V, the factorial cumulant matrix $\boldsymbol{\Theta}$ obeyed the homogeneous Eq. 41.

Finally, a typical contribution of (iv) to dH/dt is

$$\sum c_{\mu\kappa\rho}^\lambda (v_\mu + v_\kappa + v_\rho) \frac{\partial H}{\partial v_\lambda}$$
$$+ \sum c_{\mu\kappa\rho}^\lambda (v_\mu v_\kappa + v_\kappa v_\rho + v_\rho v_\mu) \frac{\partial H}{\partial v_\lambda}$$
$$\sum c_{\mu\kappa\rho}^\lambda (v_\mu v_\kappa v_\rho - v_\lambda) \frac{\partial H}{\partial v_\lambda},$$

which couples each factorial cumulant to others of the same, and lower, orders. However, the same can be said about the second moments or the covariances, so that at this stage the factorial cumulant ceases to have a clear-cut advantage. As to nonlinear master equations, it is pointed out in Section VIII that for them, the use of factorial cumulants no longer gives a worthwhile simplification. Yet it is convenient to use for the purpose of adding the effect of diffusion, as in Sections IX and X.

Appendix B: The Addition of Flow Terms

Consider a collection of independent particles subject to a force field $\mathbf{F}(\mathbf{r})$. In the single particle phase space, they constitute a cloud with density $n(\mathbf{r}, \mathbf{v})$ obeying the equation,

$$\frac{\partial n}{\partial t} = -\mathbf{v} \cdot \frac{\partial n}{\partial \mathbf{r}} - \mathbf{F}(\mathbf{r}) \cdot \frac{\partial n}{\partial \mathbf{v}} \equiv \mathbf{L}n. \qquad (95)$$

Obviously the product $n(\mathbf{r}_1, \mathbf{v}_1)n(\mathbf{r}_2, \mathbf{v}_2)$ obeys

$$\frac{d}{dt}\{n(\mathbf{r}_1, \mathbf{v}_1)n(\mathbf{r}_2, \mathbf{v}_2)\} = (\mathbf{L}_1 + \mathbf{L}_2)\{n(\mathbf{r}_1, \mathbf{v}_1)n(\mathbf{r}_2, \mathbf{v}_2)\},$$
$$(96)$$

where $\mathbf{L}_1$ and $\mathbf{L}_2$ are the same differential operator as in Eq. 95, but acting on $\mathbf{r}_1, \mathbf{v}_1$ and $\mathbf{r}_2, \mathbf{v}_2$, respectively.

Next, consider an ensemble of such collections of particles. Obviously the ensemble average $\langle n(\mathbf{r}, \mathbf{v}) \rangle$ of the particle density obeys the same Eq. 95 and $\langle n(\mathbf{r}_1, \mathbf{v}_1)n(\mathbf{r}_2, \mathbf{v}_2) \rangle$ obeys Eq. 96. It is easy to derive from this that the cumulant $\langle\langle n(\mathbf{r}_1, \mathbf{v}_1)n(\mathbf{r}_2, \mathbf{v}_2) \rangle\rangle$ also obeys Eq. 96.

Subsequently, one obtains for the factorial cumulant

$$\frac{d[n(\mathbf{r}_1, \mathbf{v}_1)n(\mathbf{r}_2, \mathbf{v}_2)]}{dt} =$$
$$\frac{d\langle\langle n(\mathbf{r}_1, \mathbf{v}_1)n(\mathbf{r}_2, \mathbf{v}_2)\rangle\rangle}{dt} - \delta(\mathbf{r}_1 - \mathbf{r}_2)\delta(\mathbf{v}_1 - \mathbf{v}_2)\frac{d\langle n(\mathbf{r}_1, \mathbf{v}_1)\rangle}{dt}$$
$$= (\mathbf{L}_1 + \mathbf{L}_2)[n(\mathbf{r}_1, \mathbf{v}_1)n(\mathbf{r}_2, \mathbf{v}_2)]+$$
$$+ (\mathbf{L}_1 + \mathbf{L}_2)\{\delta(\mathbf{r}_1 - \mathbf{r}_2)\delta(\mathbf{v}_1 - \mathbf{v}_2)\langle n(\mathbf{r}_1, \mathbf{v}_1)\rangle\} -$$
$$- \delta(\mathbf{r}_1 - \mathbf{r}_2)\delta(\mathbf{v}_1 - \mathbf{v}_2)\mathbf{L}_1\langle n(\mathbf{r}_1, \mathbf{v}_1)\rangle$$
$$= (\mathbf{L}_1 + \mathbf{L}_2)[n(\mathbf{r}_1, \mathbf{v}_1)n(\mathbf{r}_2, \mathbf{v}_2)] + R$$

The remainder R is

$$(\mathbf{L}_1 + \mathbf{L}_2)\,\delta(\mathbf{r}_1 - \mathbf{r}_2)\delta(\mathbf{v}_1 - \mathbf{v}_2)\langle n(\mathbf{r}_1, \mathbf{v}_1)\rangle - \\ -\delta(\mathbf{r}_1 - \mathbf{r}_2)\delta(\mathbf{v}_1 - \mathbf{v}_2)\mathbf{L}_1\langle n(\mathbf{r}_1, \mathbf{v}_1)\rangle \\ = \langle n(\mathbf{r}_1, \mathbf{v}_1)\rangle\,(\mathbf{L}_1 + \mathbf{L}_2)\,\delta(\mathbf{r}_1 - \mathbf{r}_2)\delta(\mathbf{v}_1 - \mathbf{v}_2).$$

However, $\mathbf{L}_1 + \mathbf{L}_2$ acting on the delta functions equals

$$\left(\mathbf{v}_1 \cdot \frac{\partial}{\partial \mathbf{r}_1} - \mathbf{v}_2 \cdot \frac{\partial}{\partial \mathbf{r}_1} + \mathbf{F}(\mathbf{r}_1) \cdot \frac{\partial}{\partial \mathbf{v}_1} - \mathbf{F}(\mathbf{r}_2) \cdot \frac{\partial}{\partial \mathbf{v}_1} \right) \times \\ \delta(\mathbf{r}_1 - \mathbf{r}_2)\delta(\mathbf{v}_1 - \mathbf{v}_2) = 0.$$

Thus, *in the case of pure flow it makes no difference whether one adds the flow terms to the second moment, the covariance, or the factorial cumulant.* In the case of diffusion, it is shown in Section V that it does make a difference, and it is necessary to add them to the factorial cumulant, in order to account for the stochastic character of the diffusion process.

Appendix C: Connection with the Langevin Equation

Let $\mathbf{u}$ be a vector in a finite or infinite dimensional Banach space. Suppose it varies with time according to the linear equation

$$\frac{d\mathbf{u}}{dt} = \mathbf{A} \cdot \mathbf{u} + \boldsymbol{\varpi}(t). \tag{97}$$

$\mathbf{A}$ is a linear operator, which may depend upon time, and $\boldsymbol{\varpi}(t)$ is a stochastic vector with the statistical properties

$$\langle \boldsymbol{\varpi}(t)\rangle = 0, \quad \langle \boldsymbol{\varpi}(t_1)\widetilde{\boldsymbol{\varpi}}(t_2)\rangle = \delta(t_1 - t_2)\boldsymbol{\Gamma}(t_1), \tag{98}$$

where $\widetilde{\boldsymbol{\varpi}}$ is the transposed vector and $\boldsymbol{\Gamma}$ is a symmetric matrix.

The Langevin Eq. 97 can be solved in terms of the evolution operator (or 'propagator' or 'Green's function') $\mathbf{Y}(t|t')$ defined by

$$\frac{d}{dt}\mathbf{Y}(t|t') = \mathbf{A}(t) \cdot \mathbf{Y}(t|t'), \quad \mathbf{Y}(t|t') = \mathbf{1}. \tag{99}$$

The solution with given initial value $\mathbf{u}(0)$ is,

$$\mathbf{u}(t) = \mathbf{Y}(t|0) \cdot \mathbf{u}(0) + \int_0^t \mathbf{Y}(t|t') \cdot \boldsymbol{\varpi}(t')\,dt'.$$

It follows that the covariance matrix of $\mathbf{u}(t)$ is

$$\langle\langle \mathbf{u}(t)\widetilde{\mathbf{u}}(t')\rangle\rangle = \\ = \int_0^t dt' \int_0^t dt'' \mathbf{Y}(t|t') \cdot \langle \boldsymbol{\varpi}(t')\widetilde{\boldsymbol{\varpi}}(t'')\rangle \widetilde{\mathbf{Y}}(t|t'') \\ = \int_0^t dt' \mathbf{Y}(t|t') \cdot \boldsymbol{\Gamma}(t') \cdot \widetilde{\mathbf{Y}}(t|t').$$

From this expression one derives a differential equation for the covariance matrix by inspection:

$$\frac{d}{dt}\langle\langle \mathbf{u}(t)\widetilde{\mathbf{u}}(t)\rangle\rangle - \mathbf{A} \cdot \langle\langle \mathbf{u}(t)\widetilde{\mathbf{u}}(t)\rangle\rangle - \langle\langle \mathbf{u}(t)\widetilde{\mathbf{u}}(t)\rangle\rangle \cdot \widetilde{\mathbf{A}} = \boldsymbol{\Gamma}(t), \tag{100}$$

where use has been made of Eq. 99. It is easily seen that the equation remains true when the covariance matrix $\langle\langle \mathbf{u}(t)\widetilde{\mathbf{u}}(t)\rangle\rangle$ is replaced with the second moment matrix $\langle \mathbf{u}(t)\widetilde{\mathbf{u}}(t)\rangle$.

Our applications of the master equation also led in each case to an equation of type (100) for the covariance matrix. We now see the same result can be reproduced by a Langevin Eq. 97 with Eq. 98, provided $\boldsymbol{\Gamma}$ is chosen correctly.

For example, in Section V it was shown that the density of a diffusing substance obeys Eq. 46. It now follows that the same result can be obtained by supplementing the diffusion equation with a Langevin term

$$\frac{dn}{dt} = D\nabla^2 n + \varpi(\mathbf{r}, t),$$

where $\langle \varpi(\mathbf{r}, t)\rangle = 0$ and

$$\langle \varpi(\mathbf{r}_1, t_1)\varpi(\mathbf{r}_2, t_2)\rangle = \\ = 2D\delta(t_1 - t_2)\nabla_1 \cdot \nabla_2 \delta(\mathbf{r}_1 - \mathbf{r}_2)\langle n(\mathbf{r}, t)\rangle. \tag{101}$$

Thus we have justified the use of a Langevin equation and derived the correlation function for the Langevin force by showing that it leads to the correct result, which has been derived in Section V using no other assumptions than those by which the macroscopic diffusion equation is derived.

In summary, the conclusion from the calculation in this Appendix is that for linear or linearized equations, every result obtained for the covariance can be reproduced by a Langevin Eq. 97, combined with a suitable choice for the matrix $\boldsymbol{\Gamma}$. However, as appears in Eq. 101, the correct $\boldsymbol{\Gamma}$ may have to depend on time, and on the particular macroscopic solution for which one is investigating the fluctuations. It is therefore no easy matter to know *a priori* the correct form of $\boldsymbol{\Gamma}$; except in equilibrium where the fluctuation-dissipation theorem can be utilized. This is why authors who use the Langevin approach for computing fluctuations in other than equilibrium states have to resort to an *ad hoc* and often rather doubtful guess about the form of $\boldsymbol{\Gamma}$.

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[42] This is a purely formal device used as a poor man's substitute for functional integration. It is not the same as the "salami method" (due to Van der Ziel [28, 29], also used by Nicolis *et al.* [23]), since we make no *a priori* assumptions concerning the correlations between cells.

[43] The precise definition of a projection operator usually implies that it must be Hermitian; it can be seen that Eq. 56 meets this requirement when a suitable scalar product is defined.

[44] The solution of Eq. 69 is a Gaussian with mean and variance determined by Eqs. 70 and 71.