

Nonlinear Brownian motion

Yu L Klimontovich

Contents

1. Introduction	738
2. Two ways of describing Brownian motion	739
2.1 The Langevin equation; 2.2 The Fokker–Planck equation	
3. Brownian motion in a medium with nonlinear friction. Three forms of the Fokker–Planck equation	740
4. The Fokker–Planck equation for a Boltzmann gas	742
5. The Smoluchowski equation. The master equation	743
6. Two ways of transition from the master equation to the Fokker–Planck equation	744
6.1 The kinetic form of the Fokker–Planck equation; 6.2 Stationary solution of the Fokker–Planck equation	
7. The master equation for a system of atoms in an electromagnetic field	745
8. Brownian motion of quantum atom oscillators	746
8.1 The master equation; 8.2 The Fokker–Planck equation	
9. Master equations for one-step processes	747
9.1 Traditional definition of transition probability; 9.2 Nontraditional definition of transition probability	
10. Spatial diffusion. The Einstein–Smoluchowski equation	749
10.1 Spatial diffusion. The Langevin method; 10.2 Diffusion of a Brownian particle in an external field;	
10.3 Comparison of stationary distributions in ‘linear’ and ‘nonlinear’ thermostats	
11. Hydrodynamic description of Brownian motion	751
12. Evolution of free energy and entropy in Brownian motion. Lyapunov functionals A_F, A_S	752
12.1 The master equation. H-theorem; 12.2 The Fokker–Planck equation. H-theorem;	
12.3 The Einstein–Smoluchowski equation. H-theorem	
13. Brownian motion in self-oscillatory systems. The Van der Pol oscillator	754
14. The Van der Pol oscillator. Symmetrised nonlinearity	755
15. Combined action of natural and external noise	756
16. The symmetrised oscillator. Distribution of coordinates and velocities	757
17. H-theorem for the Van der Pol oscillator	757
18. Self-organisation in the Van der Pol oscillator. S-theorem	758
18.1 Shannon entropy and ‘S-information’	
19. Oscillator with inertial nonlinearity	759
20. Bifurcations of the energy of the limiting cycle. Oscillators with multistable stationary states	760
21. Oscillators in discrete time. Bifurcations of the energy of the limiting cycle and of the period of oscillations	762
22. Criterion of stability upon transition to discrete time, based on the H-theorem	762
23. Brownian motion in chemically reacting systems. Partially ionised plasma	764
24. The Malthus–Verhulst process	765
References	766

Yu L Klimontovich Department of Physics, M V Lomonosov State University, Vorob’evy gory, 119899 Moscow.
Tel. (7-095) 939-3825, Fax: (7-095) 143-8547
E-mail: ylklim@hklim.phys.msu.su

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Abstract. The theory of Brownian motion as described by nonlinear Langevin equations and the corresponding Fokker–Planck equations is discussed. The general problems of the theory of nonlinear Brownian motion considered are: Brownian motion in a medium with nonlinear friction; critical analysis of the three forms of the relevant Langevin and Fokker–Planck equations (Ito’s form, Stratonovich’s form, and the kinetic form); the Smoluchowski equations and master equations for different cases; two methods of transition from master equation to Fokker–Planck equation; master equations for one-step processes; traditional and nontraditional definition of transition probabilities, evolution of free energy and entropy at Brownian motion; Lyapunov functionals. The following particular examples are considered: Brownian motion in self-oscillatory systems; H-theorem for the Van der Pol oscillator; S-theorem; oscillator with inertial nonlinearity; bifurcation of energy of the limiting cycle; oscillator with multistable stationary states; oscillators in discrete time; bifurcations of energy of the limiting cycle and the period of oscillations; criterion of instability upon transition to discrete time, based on the H-theorem; Brownian motion of quantum atoms oscillators in the equilibrium electromagnetic field; Brownian motion in chemically reacting systems; partially ionised plasmas; the Malthus–Verhulst process.

1. Introduction

Brownian motion is a familiar classroom demonstration. This phenomenon was discovered as early as 1927 by a British botanist called Robert Brown, who was the first to report the incessant chaotic movement of pollen particles suspended in liquid.

The cause of Brownian motion was not understood until much later. The theoretical foundations were laid at the turn of this century in the classic papers of Albert Einstein, Marian Smoluchowski, and Paul Langevin. The jerky motion of a suspended particle struck at random by surrounding atoms is a manifestation of the atomic structure of a ‘continuous medium’. The main results of this theory were soon confirmed experimentally by Jean Perrin and Theodor Svedberg.

Today the term ‘Brownian motion’ has acquired a much broader meaning, and the theory of Brownian motion is one of the main chapters of the statistical theory of open systems. Let us explain this general statement.

In the statistical theory of open systems the atoms, considered as microscopic structural units, are used only at the first stage of construction of the theory, in connection with the choice of the initial model of the macroscopic system in question. The ‘working equations’ are the approximate dissipative nonlinear equations of the ‘mechanics of a continuous medium’. On the kinetic level, for example, they are given by the Boltzmann equations for the distribution function $f(r, \rho, t)$. Other examples are the Vlasov–Landau equations for a plasma, and the relevant equations in solid state physics. Here use is made of the concept of a ‘continuous medium’ in the six-dimensional space of coordinates and momenta.

On the hydrodynamic level the basis is provided by the dissipative nonlinear equations of a ‘continuous medium’ for local functions in three-dimensional space — for example, density $\rho(r, t)$, velocity $u(r, t)$, and temperature $T(r, t)$.

For an even coarser description one may use the equations of chemical kinetics in the case of concentrations of chemically reacting components. The latter characterise the motion averaged over the volume of the continuous medium; they satisfy a set of ordinary differential equations.

At all three levels of description — kinetic, hydrodynamic, chemical-kinetic — we are dealing with dissipative nonlinear equations of a continuous medium for deterministic (nonrandom) functions of varying degrees of complexity. Such equations may be referred to as the *dynamic* equations of the ‘continuous medium’, so as to distinguish them from the *stochastic* equations in random functions, such as the equations of the theory of turbulence in the pulsating (random) hydrodynamic functions.

At any level of description the approximate dynamic dissipative equations may be improved to better match the exact dynamic equations of the initial microscopic model of the system by taking into account the fluctuations which reflect the existence of ‘atomic structure’ of the ‘continuous medium’. The inclusion of fluctuations into the dissipative equations of a continuous medium is also necessitated by the fluctuation dissipation relations, which hold at all levels of description.

There are two general methods for calculating the fluctuations. The first is based on obtaining a solution of the set of equations in the moments or the correlation functions. This is the most general and consistent way of calculating both equilibrium and nonequilibrium fluctuations. The second method consists of solving the kinetic, hydrodynamic, or chemical-kinetic equations that include the appropriate random sources (Langevin sources), which reflect the structure of the ‘continuous medium’. Such sources were first introduced by Paul Langevin into the dissipative dynamic (but linear) equation of motion of a Brownian particle.

Dissipative dynamic equations are, as a rule, nonlinear. Because of this, one has to deal with the more general problem of defining the structure of Langevin sources for nonlinear systems both for equilibrium and for nonequilibrium processes. The intensities of such sources are, as a rule, nonlinear.

The solution of this problem paves the way for a more general description of processes in ‘continuous media’ on the bases of the appropriate Langevin equations — the equations of fluctuation kinetics and hydrodynamics, and fluctuation chemical kinetics. The role of ‘Brownian particles’ is then played by the distribution functions, hydrodynamic functions, or concentrations.

Fluctuations in the ‘continuous medium’ have to be taken into account in the descriptions of many fundamental phenomena. First of all, there is the ‘classical’ Brownian motion of small macroscopic particles in liquid. Its description calls for consideration of the fluctuations of hydrodynamic functions of the medium. This can be done by introducing Langevin sources into the equations of hydrodynamics. We see that Brownian motion itself is a consequence of the atomic structure of the medium.

Of course, the list of phenomena which cannot be explained without taking the fluctuations into account can be continued. There is the molecular light scattering, the equilibrium phase transitions which occur because the ‘former’ state is destroyed by fluctuations which gain strength as the critical point is approached. Finally, there

are innumerable nonequilibrium phase transitions, whose sequences form the processes of self-organisation.

Naturally, in many cases the fluctuations of macroscopic functions can be calculated in the linear approximation. This possibility is incorporated in the very concept of a ‘continuous medium’, which implies that a ‘point’ is defined as a physically infinitesimal volume which contains many particles. Owing to this fact, the fluctuations in many cases may be considered to be small, and the Gaussian approximation is efficient for the calculation of even the local fluctuations. The situation, however, becomes very different when the system approaches a critical point. The Gaussian approximation is then no longer adequate, and the nonlinear theory of fluctuations must be employed. Nonlinearity of fluctuations is even more important in the description of nonlinear phase transitions.

In the calculation of nonlinear fluctuation processes it is often more efficient to use the kinetic equations in the distribution functions of those macroscopic variables whose stochastic dynamics is determined by the corresponding Langevin equations. As a result, we come to the so-called master equations, which include the Fokker–Planck equations.

Master equations in the distribution functions of macroscopic variables are in many cases linear with respect to the distribution functions. The nonlinearity of the initial Langevin equations occurs because the relevant coefficients of diffusion and friction are themselves functions of the macroscopic variables.

An extensive body of literature is devoted to the theory of Brownian motion. The pioneering papers of Einstein and Smoluchowski are reprinted in Ref. [1]. This collection also includes two reviews by Yu A Krutkov and B I Davydov. Some textbooks on statistical physics and physical kinetics [2–9] include chapters devoted primarily to the linear theory of Brownian motion, in which the dissipative coefficients and the intensity of the Langevin source are constant. The mathematical basis for the equations of Brownian motion has been developed in the classic papers of A N Kolmogorov.

The nonlinear theory of Brownian motion [5–26] has been developing rapidly in recent years. Currently it constitutes one of the main chapters of the modern statistical theory of open systems. The applications of this theory are extremely diverse. For illustration the reader may refer to the reviews devoted to the theory of fluctuations in lasers [20, 21].

Despite considerable progress, however, there are a number of fundamental questions which have not yet received comprehensive treatment. This paper is intended to fill the remaining gaps; we have chosen only those issues which pertain to the foundations of the theory.

First of all, there is the intercomparison of different descriptions of nonlinear Brownian motion. Given the nonlinear dissipative coefficients, Brownian motion is still being described by Langevin equations and the corresponding Fokker–Planck equations which are different in structure and in physical content. One may distinguish three ways of writing the stochastic and the corresponding kinetic equations: Ito’s form (I-form), Stratonovich’s form (S-form), and the kinetic form (K-form) [17–19].

This ambiguity calls for additional physical analysis which would enable one to select the most physically

reasonable formulation of the equations of the nonlinear theory of Brownian motion.

A similar problem is encountered in case of the so-called master equations, which are a straightforward implication of the integral Smoluchowski equation. The choice of the most natural representation here is also based on additional physical arguments. The comparison between the alternative formulations is drawn for the exemplary case of one-step processes [19].

Issues of fundamental importance also include the transition from the Fokker–Planck equation for the distribution function $f(r, v, t)$ to the Einstein–Smoluchowski equation for the spatial distribution of Brownian particles $f(r, t)$. In particular, we discuss the analogy between this transition and the transition from the Boltzmann equation to the equations of gas dynamics. This analogy provides the basis for introducing the generalised kinetic equation [27, 28] in the theory of Brownian motion, which allows the use of the Kramers approximation [11, 12, 29, 30] without resorting to the methods of the perturbation theory.

The discussion is illustrated with examples of Brownian motion in passive and active nonlinear systems. In particular, we consider Brownian motion at equilibrium and non-equilibrium phase transitions. Some problems of nonlinear Brownian motion are also discussed.

For consistency, we start with a brief overview of the results of the linear theory of Brownian motion.

2. Two ways of describing Brownian motion

2.1 The Langevin equation

Let a Brownian particle be a sphere of radius a and mass M . By v we denote the velocity of a particle relative to the liquid. The sphere moving in the liquid is acted upon by the force of friction, which at constant velocity v is given by Stokes’ formula

$$F = -M\gamma v, \quad \gamma = \frac{6\pi a}{M}\eta, \quad \eta = \rho\nu, \quad (2.1)$$

where the coefficient of friction γ is proportional to the dynamic viscosity η .

The equation of motion with only the force of friction is not sufficient for describing Brownian motion (such an equation corresponds to the approximation of a continuous medium). To account for the atomic structure of the medium, Langevin introduced an additional force $F_L \equiv My(t)$

$$\frac{dr}{dt} = v, \quad \frac{dp}{dt} + \gamma p = F_0 + My(t), \quad F_0 = -\text{grad } U, \quad (2.2)$$

where F_0 is the external force (for instance, gravity).

This equation of motion includes three forces: the Stokes force (same as in the approximation of a continuous medium), the external force, and the Langevin force. The Langevin force is a random function of time, as it reflects the existence of atomic structure of the liquid.

Assume that the medium is at equilibrium, and the external force is zero. Then all directions of the random force are equivalent, and its mean value is therefore zero. Now what is the structure of the second moment $\langle y_i(t) y_j(t') \rangle$? We may assume that the characteristic time of correlation of the values of the Langevin force τ_{cor}^L is much less than the relaxation time due to viscous friction $\tau_{\text{rel}} = 1/\gamma$ — that is,

$$\tau_{\text{cor}}^L \ll \tau_{\text{rel}} = \frac{1}{\gamma}. \quad (2.3)$$

In the zero approximation in this parameter the correlation time is taken to be zero. This type of random source is said to be delta-correlated. It would be natural to further assume that there is no correlation between different components of the Langevin source, since there is no preferential direction. As a result, we obtain expressions for the two moments,

$$\langle y_i(t) \rangle = 0, \quad \langle y_i(t)y_j(t') \rangle = 2D\delta_{ij}\delta(t-t'), \quad (2.4)$$

where $2D$ is the intensity of the Langevin source (the mean intensity of random kicks from the side of atoms of the medium). A factor of two is introduced for convenience, so that D in the kinetic equation below could be regarded as a coefficient of diffusion.

Inequality (2.3) allows us to regard $y(t)$ as a Gaussian random process. Thus for the statistical description of the process the knowledge of the first two moments is sufficient.

Equations of motion with the Langevin source are not yet closed, because the intensity of noise D is unknown. It can be defined from the condition of statistical equilibrium between Brownian particles and the surrounding medium. This brings us to the so-called Einstein relation

$$D = \gamma \frac{kT}{M}, \quad (2.5)$$

which links the intensity of Langevin source with the dissipative factor γ and the temperature T . Historically, this formula is the first example of a fluctuation–dissipation relation.

2.2 The Fokker–Planck equation

The Langevin equation is a stochastic differential equation—that is, a differential equation which contains both deterministic and random forces. Any other parameter of the differential equation can also be random. The statistical characteristics of Brownian motion can be found by solving these equations.

There is, however, an alternative approach to the description of Brownian motion, based on the solution of the kinetic equation for the one-particle distribution $f(\mathbf{r}, \mathbf{v}, t)$ of Brownian particles in six-dimensional phase space \mathbf{r}, \mathbf{v} . This equation is similar to the Boltzmann kinetic equation for a rarefied gas. In the theory of Brownian motion the kinetic equation is usually referred to as the Kramers equation, or (more commonly) as the Fokker–Planck equation.

For the kinetic description of Brownian motion one has to define the ensemble of noninteracting Brownian particles (the appropriate Gibbs ensemble). Then, instead of following the movement of individual particles, we are dealing with their distribution in six-dimensional phase space or, in other words, with a ‘continuous medium’ of noninteracting Brownian particles. Drawing an analogy with hydrodynamics, the Langevin description corresponds to the method of Lagrange, and the kinetic description to the method of Euler.

The Fokker–Planck equation can be established in different ways, one of which is based on the Langevin equation. For example, the kinetic equation which corresponds to equations (2.2) has the following form:

$$\frac{\partial f}{\partial t} + \mathbf{v} \cdot \frac{\partial f}{\partial \mathbf{r}} - \frac{1}{M} \frac{\partial U}{\partial \mathbf{r}} \cdot \frac{\partial f}{\partial \mathbf{v}} = D \frac{\partial^2 f}{\partial \mathbf{v}^2} + \frac{\partial}{\partial \mathbf{v}} (\gamma \mathbf{v} f) = I_{F-P}. \quad (2.6)$$

Brownian particles move in the liquid which by itself is at equilibrium. If, by assumption, equilibrium can be established between the Brownian particles and the medium, then the equilibrium solution of the Fokker–Planck equation is the Maxwell–Boltzmann distribution

$$f(\mathbf{r}, \mathbf{v}) = C \exp \left[-\frac{(M|\mathbf{v}|^2/2) + U(\mathbf{r})}{kT} \right], \quad \int f d\mathbf{r} d\mathbf{v} = 1. \quad (2.7)$$

Substitution of this distribution into the Fokker–Planck equation results in the Einstein relation (2.5).

3. Brownian motion in a medium with nonlinear friction. Three forms of the Fokker–Planck equation

Consider now the case when the coefficient of friction is a function of velocity,

$$\gamma(\mathbf{v}) = \gamma(-\mathbf{v});$$

for example,

$$\gamma(\mathbf{v}) = \gamma(1 + b|\mathbf{v}|^3). \quad (3.1)$$

This will obviously complicate the Langevin equations. First of all, we should anticipate that the intensity of noise will also be a function of velocity $D = D(\mathbf{v})$, as an implication of the fluctuation dissipation relation in case of nonlinear friction. This gives rise to an additional ‘stochastic force’, proportional to the derivative of $D(\mathbf{v})$. Then it would be natural to represent the Langevin equations in the form

$$\frac{d\mathbf{r}}{dt} = \mathbf{v}, \quad \frac{d\mathbf{v}}{dt} + \gamma(\mathbf{v})\mathbf{v} + \frac{1}{M} \frac{\partial U}{\partial \mathbf{r}} + a \frac{\partial D}{\partial \mathbf{v}} = \sqrt{D(\mathbf{v})} \mathbf{y}(t). \quad (3.2)$$

Here we have already separated the intensity of the random source from the Langevin force, and so the moments of random function $\mathbf{y}(t)$ are now defined as

$$\langle \mathbf{y}(t) \rangle = 0, \quad \langle y_i(t)y_j(t') \rangle = 2\delta_{ij}\delta(t-t'). \quad (3.3)$$

Now we are worse off than before, because in place of one unknown constant $D(\mathbf{v})$ we have an unknown function $D(\mathbf{v})$ and an unknown coefficient a .

Assume as before that statistical equilibrium exists between the Brownian particles and the medium. Moreover, we assume that the structure of Einstein’s fluctuation–dissipation relation remains the same. Then Eqn (2.5) becomes

$$D(\mathbf{v}) = \gamma(\mathbf{v}) \frac{kT}{M}. \quad (3.4)$$

From the standpoint of the kinetic theory, the considerations which brought us to the generalised Einstein relation are so natural that the alternatives are not even being discussed [4, 9]. This relation follows naturally, in particular, from the Boltzmann equation for a mixture of heavy and light gases (see below), and from the kinetic Landau equation of the Balescu–Lenard equation for plasmas. Indeed, the Einstein relation of the form given by (3.4) is a direct implication of these equations for the state of equilibrium. Problems arise, however, when the initial equations for the description of nonlinear Brownian motion are the stochastic Langevin equations rather than the kinetic equations.

Then, depending on the mathematical treatment of the integrals containing delta-correlated Langevin sources, there are different ways of describing stochastic processes in systems with the same nonlinear friction. The most typical are three different presentations both of the Langevin equations and the corresponding Fokker–Planck equations: Ito’s (I-form), Stratonovich’s (S-form), and the K-form, which is a natural implication of the kinetic theory [11–15, 17–19].

To show the distinction between these three forms for the example of Brownian motion with nonlinear friction, let us accomplish the transition from Langevin equations (3.2) to the corresponding kinetic Fokker–Planck equation for the distribution function $f(r, v, t)$. We shall employ the procedure commonly used for deriving the kinetic equations for gases and plasmas [9].

Like we did in the kinetic theory of gases, we define the microscopic phase density in six-dimensional phase space r, v as

$$N(r, v, t) = \sum_{1 \leq i \leq N} \delta[r - r_i(t)] \delta[v - v_i(t)]. \quad (3.5)$$

This time, however, functions $r_i(t), v_i(t)$ satisfy the irreversible stochastic Langevin equations rather than the reversible Hamilton equations. Since the system of Brownian particles may be considered as an ‘ideal gas’, in place of phase density (3.5) we may use the one-particle dynamic distribution

$$f^{(d)}(r, v, t) = \delta[r - r(t)] \delta[v - v(t)],$$

$$\int f^{(d)} dr dv = 1. \quad (3.6)$$

Functions $r(t), v(t)$, satisfy the Langevin equations (3.2). Given this, the equation for the dynamic distribution can be written as the continuity equation

$$\frac{\partial f^{(d)}}{\partial t} + v \cdot \frac{\partial f^{(d)}}{\partial r} - \frac{1}{M} \frac{\partial U}{\partial r} \cdot \frac{\partial f^{(d)}}{\partial v}$$

$$= \frac{\partial}{\partial v} \left[\left(\gamma(v)v + a \frac{\partial D}{\partial v} \right) f^{(d)} \right] - \frac{\partial}{\partial v} \left(\sqrt{D(v)} \mathbf{y}(t) f^{(d)} \right).$$

(3.7)

The sought-for statistical distribution is the first moment:

$$f(r, v, t) = \langle f^{(d)} \rangle \text{ and the fluctuation } \delta f = f^{(d)} - f. \quad (3.8)$$

The equation for the distribution function f follows from Eqn (3.7), which after averaging over the Gibbs ensemble takes on the form

$$\frac{\partial f}{\partial t} + v \cdot \frac{\partial f}{\partial r} - \frac{1}{M} \frac{\partial U}{\partial r} \cdot \frac{\partial f}{\partial v}$$

$$= \frac{\partial}{\partial v} \left[\left(\gamma(v)v + a \frac{\partial D}{\partial v} \right) f \right] - \frac{\partial}{\partial v} \left(\sqrt{D(v)} \langle \mathbf{y}(t) \delta f \rangle \right).$$

(3.9)

This equation is not closed because it includes, along with the distribution function f , the correlator $\langle \mathbf{y}(t) \delta f \rangle$ (we have noted that the mean value of $\langle \mathbf{y}(t) \rangle$ is zero).

To obtain a closed equation, one must express the unknown correlator in terms of the distribution function f . This task is now simpler than it was in the case of Boltzmann’s kinetic equation, since we start with the

dissipative Langevin equations rather than with the reversible Hamilton equations. We proceed as follows [9, 10, 17].

Using equations for functions $f^{(d)}$ and f we construct an equation for the fluctuation δf . For calculating the correlator $\langle \mathbf{y}(t) \delta f \rangle$ it is sufficient to know the solution for δf on small time intervals of the order of τ^L . This allows us to keep only the term with the delta-correlated source $\delta \dot{f}$ in the equation for $\mathbf{y}(t)$. As a result, the equation for fluctuation of the distribution function assumes the form

$$\frac{\partial \delta f}{\partial t} = - \frac{\partial}{\partial v} \left[\sqrt{D(v)} \mathbf{y}(t) f(r, v, t) \right], \quad (3.10)$$

whence follows the desired solution

$$\delta f(r, v, t) = - \frac{\partial}{\partial v} \left[\sqrt{D(v)} \int_0^\infty \mathbf{y}(t - \tau) f(r, v, t - \tau) d\tau \right]. \quad (3.11)$$

Substituting this solution into the last term of Eqn (3.9) and taking into account the structure of correlator $\langle y_i(t) y_j(t') \rangle$, we get

$$- \frac{\partial}{\partial v} \left[\sqrt{D(v)} \langle \mathbf{y} \delta f \rangle \right] = \frac{\partial}{\partial v} \sqrt{D(v)} \frac{\partial}{\partial v} \left[\sqrt{D(v)} f \right]$$

$$= \frac{\partial}{\partial v} \left[D(v) \frac{\partial f}{\partial v} \right] + \frac{\partial}{\partial v} \left(\frac{1}{2} \frac{\partial D}{\partial v} f \right).$$

(3.12)

Substituting this into the right-hand side of Eqn (3.9), we obtain the kinetic Fokker–Planck equation:

$$\frac{\partial f}{\partial t} + v \cdot \frac{\partial f}{\partial r} - \frac{1}{M} \frac{\partial U}{\partial r} \cdot \frac{\partial f}{\partial v}$$

$$= \frac{\partial}{\partial v} \left[D(v) \frac{\partial f}{\partial v} \right] + \frac{\partial}{\partial v} \left\{ \left[\gamma(v)v + \left(a + \frac{1}{2} \right) \frac{\partial D}{\partial v} \right] f \right\}.$$

(3.13)

Like the initial Langevin equations, the Fokker–Planck equation (3.13) is not yet completely closed because it still contains an unknown function $D(v)$ and an unknown coefficient a . To make further progress, we take advantage of the statistical equilibrium between the Brownian particles and the surrounding medium.

Under this condition, the equilibrium solution of the Fokker–Planck equation must have the form of the Maxwell–Boltzmann distribution (2.7). Substituting this distribution into Eqn (3.13) for the state of equilibrium we come to the following equation:

$$D(v) = \left[\left(a + \frac{1}{2} \right) \frac{v}{v^2} \cdot \frac{\partial D(v)}{\partial v} + \gamma(v) \right] \frac{kT}{M}, \quad (3.14)$$

which links the source intensity $D(v)$ with the nonlinear dissipative coefficient $\gamma(v)$, but the unknown coefficient a is still there.

If the coefficient of nonlinear friction is known, this equation may be regarded as a differential equation with respect to the intensity of random source $D(v)$ —that is, as a differential fluctuation–dissipation relation. From statistical theory, however, it follows that such a linkage is not differential (on the contrary, the fluctuation factor is proportional to the dissipation factor). This implies that for the case in question the fluctuation–dissipation relation must reduce to the generalised Einstein relation (3.4), and hence the coefficient a in the equations of Langevin and Fokker–Planck is $a = -1/2$. On the strength of these arguments we come to the so-called ‘kinetic form’ of the

Fokker–Planck equation for Brownian motion in a medium with nonlinear friction [17–19]

$$\frac{\partial f}{\partial t} + v \cdot \frac{\partial f}{\partial \mathbf{r}} - \frac{1}{M} \frac{\partial U}{\partial \mathbf{r}} \cdot \frac{\partial f}{\partial \mathbf{v}} = \frac{\partial}{\partial v} \left[D(v) \frac{\partial f}{\partial v} \right] + \frac{\partial}{\partial v} \left[\gamma(v) v f \right]. \quad (3.15)$$

The corresponding Langevin equations are:

$$\frac{d\mathbf{r}}{dt} = v, \quad \frac{dv}{dt} + \gamma(v)v + \frac{1}{M} \frac{\partial U}{\partial \mathbf{r}} - \frac{1}{2} \frac{\partial D}{\partial v} = \sqrt{D(v)} \mathbf{y}(t). \quad (3.16)$$

Now we can summarise the results.

Although the Langevin equations are nonlinear with respect to dissipation, the kinetic Fokker–Planck equation is linear with respect to the distribution function. The nonlinearity of the medium is taken into account by regarding the coefficients $D(v)$, $\gamma(v)$ as functions of velocity.

Along with the ‘kinetic’ Fokker–Planck equation (3.15) (K-form), other representations of Langevin equations and the corresponding Fokker–Planck equations can be found in the literature. The most important of these are Ito’s (I-form) and Stratonovich’s (S-form) representations.

The difference between Ito’s and Stratonovich’s approaches consists mainly in the different treatment of stochastic integrals which come up in the solutions of nonlinear stochastic Langevin equations [11–15, 17–19]. Even though these representations are based on similar stochastic equations [such as Eqn (3.2) with $a = 0$], they result in different Fokker–Planck equations:

$$\frac{\partial f}{\partial t} + v \cdot \frac{\partial f}{\partial \mathbf{r}} - \frac{1}{M} \frac{\partial U}{\partial \mathbf{r}} \cdot \frac{\partial f}{\partial \mathbf{v}} = \frac{\partial^2}{\partial v^2} \left[D(v) f \right] + \frac{\partial}{\partial v} \left[\gamma(v) v f \right] \quad (3.17)$$

(I-form), and

$$\begin{aligned} \frac{\partial f}{\partial t} + v \cdot \frac{\partial f}{\partial \mathbf{r}} - \frac{1}{M} \frac{\partial U}{\partial \mathbf{r}} \cdot \frac{\partial f}{\partial \mathbf{v}} &= \frac{\partial}{\partial v} \left[\sqrt{D(v)} \frac{\partial}{\partial v} \left(\sqrt{D(v)} f \right) \right] \\ &+ \frac{\partial}{\partial v} \left[\gamma(v) v f \right] \end{aligned} \quad (3.18)$$

(S-form).

Let us compare the above three forms of the Fokker–Planck equation with Eqn (3.13) where parameter a has an unspecified value. We see that the I-form corresponds to $a = 1/2$, the S-form to $a = 0$, and the K-form to $a = -1/2$, and it is only in the last case that Eqn (3.14) coincides with the generalised Einstein relation.

Observe that Eqn (3.14) can be formally reduced to the Einstein relation (3.4) by introducing the effective coefficient of friction as

$$D(v) = \gamma_{\text{eff}} \frac{kT}{M}, \quad \gamma_{\text{eff}} = \gamma(v) + \left(a + \frac{1}{2} \right) \frac{v}{|v|^2} \cdot \frac{\partial D(v)}{\partial v}. \quad (3.19)$$

Using this definition, one can also reduce the general Fokker–Planck equation (3.13) to the ‘kinetic’ form.

This, however, is just a formal trick. The question is why are there three different Fokker–Planck equations which correspond to the same nonlinear dynamic system with a given dissipative coefficient.

Three different Fokker–Planck equations give rise to three different forms of stationary distributions:

$$f(v) = \frac{C}{|D(v)|^v} \exp \left[- \int_0^v \frac{\gamma(v')}{D(v')} d \frac{v'^2}{2} \right], \quad \int f(v) dv = 1, \quad (3.20)$$

where $v = 1$ for the I-form, $v = 1/2$ for the S-form, and $v = 0$ for the K-form. Only in the last case does the stationary solution depend on the ratio of fluctuation and dissipation factors, in agreement with the general structure of fluctuation–dissipation relations.

Let us return to the Langevin equation (3.2) with an arbitrary a . The Langevin force depends on v , and therefore its mean value is nonzero. The correlator is defined as above. As a result we obtain the expression

$$\left\langle \sqrt{D(v)} \mathbf{y}(t) \right\rangle = \frac{1}{2} \left\langle \frac{\partial D(v)}{\partial v} \right\rangle, \quad (3.21)$$

which does not depend on a .

In the next section we are going to give physical examples which support the choice of the Fokker–Planck equation in the ‘kinetic’ form (3.15). Later we shall also discuss the inverse transition from the Fokker–Planck equation to the Langevin equation [5]. This is important because in the context of statistical theory it is the kinetic equations that are more justified, or, so to speak, ‘primary’.

4. The Fokker–Planck equation for a Boltzmann gas

Consider an admixture of a heavier gas and a lighter gas. The heavier atoms act as Brownian particles in the medium represented by the main gas. We denote the distribution function of impurity atoms by $f_{\text{im}}(\mathbf{r}, \mathbf{p}, t)$, and proceed to find the appropriate kinetic equation.

We start with the set of Boltzmann equations for distribution functions of light and heavy atoms. Assume that the concentration of heavy atoms is small enough to make the collisions between them entirely unimportant. In the collision integral of heavy and light atoms we carry out expansion in a small parameter $|\mathbf{p} - \mathbf{p}'|/Mv_T$ (the ratio of the change in momentum of light atom to the momentum of impurity atom). As a result, we come to the Fokker–Planck equation

$$\frac{\partial f_{\text{im}}}{\partial t} + v \cdot \frac{\partial f_{\text{im}}}{\partial \mathbf{r}} + \mathbf{F} \cdot \frac{\partial f_{\text{im}}}{\partial \mathbf{p}} = \frac{\partial}{\partial p_i} \left[D_{ij}(\mathbf{p}) \frac{\partial f_{\text{im}}}{\partial p_j} \right] + \frac{\partial}{\partial p_i} \left[A_i(\mathbf{p}) f_{\text{im}} \right]. \quad (4.1)$$

In structure, this equation corresponds to Eqn (3.15); the difference is that now the diffusion is characterised by a tensor $D_{ij}(\mathbf{p})$, and the dissipation by a vector $A_i(\mathbf{p})$.

There are two possible ways of expressing these functions:

(1) The tensor of diffusion and the vector of dissipation are expressed directly via the distribution function of the gas of light particles, which satisfies the relevant Boltzmann equation.

(2) Using the fluctuation representation of the Boltzmann collision integral [31, 9, 17], one can express the tensor of diffusion in terms of spectral density of fluctuations of the ‘potential of scattering’ δU , and the vector of dissipation in terms of the imaginary part of the relevant susceptibility α :

$$D_{ij}(\mathbf{p}) = \frac{1}{16\pi^3} \int \delta(\omega - \mathbf{k} \cdot \mathbf{v}) (\delta U \delta U)_{\omega, \mathbf{k}, p} k_i k_j d\omega d\mathbf{k}, \quad (4.2)$$

$$A_i(\mathbf{p}) = \frac{1}{8\pi^3} \int \delta(\omega - \mathbf{k} \cdot \mathbf{v}) k_i \text{Im} \alpha(\omega, \mathbf{k}, p) d\omega d\mathbf{k}. \quad (4.3)$$

Both functions in the integrands are in their turn expressed via the distribution function of the light particles which satisfies the Boltzmann equation. At equilibrium, when the solution of the Boltzmann equation is the Maxwell distribution, these functions are linked by the fluctuation–dissipation relation

$$(\delta U \delta U)_{\omega, k, p} = \frac{2}{\omega} \text{Im} \alpha(\omega, \mathbf{k}, \mathbf{p}) k T . \quad (4.4)$$

This relation ensures that functions $D_{ij}(\mathbf{p})$, $A_i(\mathbf{p})$ satisfy Einstein’s formula

$$\frac{v_i D_{ij} v_j}{v^2} = D(v) = \gamma(v) k T ,$$

$$\gamma(v) = \frac{1}{(2\pi)^3} \int \delta(\omega - \mathbf{k} \cdot \mathbf{v}) \frac{\mathbf{k} \cdot \mathbf{v}}{|\mathbf{v}|^2} \text{Im} \alpha(\omega, \mathbf{k}, \mathbf{p}) d\omega d\mathbf{k} . \quad (4.5)$$

The concrete dependence of the coefficient of friction on velocity via the function $\alpha(\omega, k, p)$ is defined by the potential of interaction of gas particles.

Observe once again that the coefficient of diffusion and the intensity of the Langevin force reflect the existence of the atomic structure of the medium. Atomic structure is the source of intrinsic ‘natural’ noise. Of course, the intensity of external noise from various sources may be much greater. Nevertheless, the role of even the low-intensity natural sources is quite important, as will be illustrated with numerous examples. We shall also see that the intensity of natural noise increases dramatically when the system approaches all kinds of critical points (points of equilibrium or nonequilibrium phase transitions). As a rule, the growth of fluctuations is a harbinger of forthcoming structural change.

5. The Smoluchowski equation. The master equation

Other forms of kinetic equations are also extensively used in the theory of Brownian motion. First of all, there is the Smolukhovski equation (also known as Chapman–Kolmogorov equation). This equation may be interpreted as the condition of consistency of distribution functions of different orders.

Let us denote by x an arbitrary set of variables, and by $f(x, t)$, and $f(x, t, x', t')$ the distribution functions for, respectively, the given time t , and the two consecutive instants t, t' . We use two identities,

$$f(x, t) = \int f(x, t, x', t') dx' \quad (5.1)$$

$$\equiv \int f(x, t|x', t') f(x', t') dx' , \quad \int f(x, t) dx = 1 .$$

The first of these is the condition of consistency, and the second gives the definition of the conditional distribution function referring to two different time instants. This distribution function is called the probability of transition. We denote it as

$$f(x, t|x', t') \equiv p(x, t, x', t') ,$$

and use the normalisation condition

$$\int f(x, t|x', t') dx = \int p(x, t, x', t') dx = 1 .$$

Then Eqn (5.1) can be rewritten as

$$f(x, t) = \int p(x, t, x', t') f(x', t') dx' , \quad p(x, t, x', t') > 0 . \quad (5.2)$$

Substituting the value of $f(x', t')$ expressed via the distribution $f(x_0, t_0)$ at an earlier time into the right-hand side, we obtain the integral relation which includes an intermediate point x' :

$$f(x, t) = \int p(x, t, x', t') p(x', t', x_0, t_0) f(x_0, t_0) dx' dx_0 . \quad (5.3)$$

This can be used to obtain a closed equation for transition probabilities. Into the left-hand side of Eqn (5.3) we substitute Eqn (5.2) with $x', t' \rightarrow x_0, t_0$. Since the equation obtained in this way holds for the arbitrary distribution $f(x_0, t_0)$, we may equate the integrands. As a result, we arrive at the Smoluchowski equation

$$p(x, t, x_0, t_0) = \int p(x, t, x', t') p(x', t', x_0, t_0) dx' . \quad (5.4)$$

In order to return to Eqn (5.3), we have to multiply both sides by $f(x_0, t_0)$ and carry out integration with respect to x_0 . It is also possible to go back from Eqn (5.3) to Eqns (5.2) and (5.1).

It follows that the above relations are exact, because our transitions between equations did not involve any simplifying assumptions. In particular, the integral relation (5.2) links the distribution functions $f(x, t)$, $f(x', t')$ via the probability of transition. However, this is not yet a kinetic equation, since the transition probability is not known. We may only argue that it satisfies the Smoluchowski equation (5.4) and the normalisation condition. To obtain a closed kinetic equation we need additional information about the system. How do we proceed now?

First we shall go over from the exact relation (5.2) to the so-called master equation which is simpler. For this we assume that there are two characteristic time scales, the ‘fast’ and the ‘slow’. Recall that we have used a similar assumption in the derivation of the Langevin equations.

Assume that the distribution function $f(x, t)$ changes slowly with time. As before, the characteristic relaxation time we denote by τ_{rel} ; the characteristic correlation time for the ‘fast’ process is τ_{cor} . The probability of transition depends explicitly only on the fast time, since in the zero approximation with respect to τ_{cor}/τ_{rel} the process may be regarded as stationary.

Given this, Eqn (5.2) can be rewritten as (with the replacement t, t' , where $t + \Delta t, t(\Delta t = t - t')$)

$$f(x, t + \Delta t) = \int p(x, x', \Delta t) f(x', t) dx' . \quad (5.5)$$

Now we expand the left-hand side in Δt and retain the first two terms. Assume that there exists the limit

$$\lim_{\Delta t \rightarrow 0} \frac{1}{\Delta t} p(x, x', \Delta t) = W(x, x') , \quad (5.6)$$

which determines the rate of change of the transition probability. We also note that the probability of transition has the following properties:

$$\int p(x', t', x, t) dx' = 1 , \quad p(x', x, \Delta t) = \Delta t W(x', x) . \quad (5.7)$$

The first of these follows from the normalisation condition of the distribution functions $f(x, t), f(x', t')$ in Eqn (5.1). In the second we use the limit (5.6) and retain only the main term in the expansion in Δt . Taking advantage of these properties, we transform the first term in the expansion in Δt on the left-hand side of Eqn (5.5):

$$f(x, t) = \int p(x', x, \Delta t) dx' f(x', t) = \Delta t \int W(x', x) dx' f(x', t) . \quad (5.8)$$

Now we use Eqns (5.6) and (5.8) to convert Eqn (5.5) to the desired 'master equation'

$$\frac{\partial f(x, t)}{\partial t} = \int [W(x, x')f(x', t) - W(x', x)f(x, t)] dx' , \quad (5.9)$$

$$W(x, x') > 0 .$$

This equation is not yet closed, since the structure of function W is not defined. Observe that many of the known kinetic equations of the statistical theory of nonequilibrium processes, including the kinetic Boltzmann equation, can be reduced to this form. The master equation, however, is nonlinear because the probabilities W themselves depend on the distribution function $f(x, t)$. For a spatially homogeneous Boltzmann gas the variable $x = p$ is a vector.

So, we have made the first step towards particularising Eqn (5.2). This has been made possible by the simplifying assumption that there exist two different time scales, and that the process is stationary on small time intervals. Then the probability of transition $p(x, t, x', t')$ is replaced by the simpler functions $W(x, x'), W(x', x)$, which do not explicitly depend on time. The resulting kinetic equation is irreversible because small-scale correlations are eliminated. This will be confirmed by the analysis of the relevant equation of entropy balance.

6. Two ways of transition from the master equation to the Fokker-Planck equation

For future discussion it will be convenient to use a different form of master equation in place of Eqn (5.9). We represent the probability of transition as a sum of symmetrical and antisymmetrical parts:

$$W(x', x) = W^s(x, x') + W^a(x, x') , \quad (6.1)$$

$$W^s(x, x') = W^s(x', x) , \quad W^a(x, x') = -W^a(x', x) .$$

Then Eqn (5.9) can be rewritten in the form

$$\frac{\partial f(x, t)}{\partial t} = \int \left\{ W^s(x, x')[f(x', t) - f(x, t)] - W^a(x, x')[f(x', t) + f(x, t)] \right\} dx' . \quad (6.2)$$

It is interesting that all major kinetic equations (including the kinetic Boltzmann equation) can be reduced to this form. Reduced to the same form can also be the quantum kinetic equations for a plasma and for a system of atoms interacting with an electromagnetic field. In such cases, however, the master equations are nonlinear because the probabilities of transition themselves depend on the distribution functions. The transition to linear equations is only possible for Brownian motion, where the statistical properties of the medium are known and the interaction between Brownian particles can be neglected.

As follows from Eqn (6.2), the transition probabilities W^s, W^a in the stationary state are linked by

$$W^s(x, x') = W^a(x, x') \frac{f^{(st)}(x') + f^{(st)}(x)}{f^{(st)}(x') - f^{(st)}(x)} , \quad (6.3)$$

which is a fluctuation-dissipation relation. Here the symmetrical function $W^s(x, x')$ defines the coefficient of diffusion in the corresponding Fokker-Planck equation, and the antisymmetrical function $W^a(x, x')$ defines the coefficient of friction.

Now let us go over from the master equation (6.2) to the Fokker-Planck equation. We shall see that this transition is not unambiguous, and may lead to different forms of the desired equation.

6.1 The kinetic form of the Fokker-Planck equation

In place of x, x' we introduce new variables $\Delta x, x$:

$$W^{s,a}(x, x') = W^{s,a}\left(x - x', \frac{x + x'}{2}\right) = W^{s,a}\left(\Delta x, x - \frac{\Delta x}{2}\right) , \quad (6.4)$$

$$\Delta x = x - x'$$

and rewrite the master equation as

$$\frac{\partial f(x, t)}{\partial t} = \int \left\{ W_{\Delta x, x - (\Delta x/2)}^s [f(x - \Delta x, t) - f(x, t)] - W_{\Delta x, x - (\Delta x/2)}^a [f(x - \Delta x, t) + f(x, t)] \right\} d\Delta x . \quad (6.5)$$

Assume (!) that $\Delta x \ll x$, but the dependence of functions $W^{s,a}$ on the first argument Δx is not weak. For the sake of simplicity we consider a one-dimensional case when x is a scalar, and carry out the expansion in a small variation $\Delta x \partial/\partial x$. Making use of equations

$$\int W_{\Delta x, x}^s \Delta x d\Delta x = 0 , \quad \int W_{\Delta x, x}^a d\Delta x = 0 \quad (6.6)$$

we transform the first and the second terms in Eqn (6.5):

$$\frac{1}{2} \int (\Delta x)^2 \frac{\partial}{\partial x} \left(W_{\Delta x, x}^s \frac{\partial f}{\partial x} \right) d\Delta x = \frac{\partial}{\partial x} \left[D(x) \frac{\partial f}{\partial x} \right] ,$$

$$\int \Delta x \frac{\partial}{\partial x} \left(W_{\Delta x, x}^a f \right) d\Delta x = \frac{\partial}{\partial x} [A(x)f] , \quad (6.7)$$

where the coefficients of diffusion and friction are

$$D(x) = \frac{1}{2} \int (\Delta x)^2 W_{\Delta x, x}^s d\Delta x , \quad A(x) = \int \Delta x W_{\Delta x, x}^a d\Delta x . \quad (6.8)$$

As a result, we come to the kinetic form of the Fokker-Planck equation:

$$\frac{\partial f(x, t)}{\partial t} = \frac{\partial}{\partial x} \left[D(x) \frac{\partial f}{\partial x} \right] + \frac{\partial}{\partial x} [A(x)f] . \quad (6.9)$$

In order to obtain the Fokker-Planck equation in the forms of Ito and Stratonovich, similar to Eqns (3.17) and (3.18), we must make the following replacement in Eqn (6.9):

$$A(x) \rightarrow A(x) + \left(a + \frac{1}{2} \right) \frac{dD(x)}{dx} , \quad (6.10)$$

which implies redefinition of either the coefficient of diffusion or the coefficient of friction. Here $a = 1/2$ for Ito's representation, and $a = 0$ for Stratonovich's representation.

We see that the coefficients of diffusion and friction are completely defined in terms of, respectively, the even and the odd parts of the transition probability only for the kinetic representation of the Fokker–Planck equation. This is yet another argument in favour of the K-form of the kinetic equation.

How do we obtain different forms of the Fokker–Planck equation from one and the same master equation? Return to the initial equation (5.9) and introduce new variables in a fashion less symmetrical than in Eqn (6.4) [11]: $x, x' \rightarrow x' - x, x' = \Delta x, x + \Delta x$, where $(\Delta x = x' - x)$. Accordingly, the first argument is now the difference in the former second and first arguments. The second argument remains the same. The transition probabilities are then redefined as follows:

$$\begin{aligned} W(x, x') &\rightarrow W(x' - x, x') = W(\Delta x, x + \Delta x), \\ W(x', x) &\rightarrow W(-\Delta x, x). \end{aligned} \tag{6.11}$$

As a result, the master equation (5.9) becomes

$$\frac{\partial f(x, t)}{\partial t} = \int [W(\Delta x, x + \Delta x)f(x + \Delta x, t) - W(-\Delta x, x)f(x, t)] d\Delta x. \tag{6.12}$$

The terms of zero order cancel out because

$$\int W(\Delta x, x) d\Delta x = \int W(-\Delta x, x) d\Delta x. \tag{6.13}$$

[This can be easily proved using definition (6.1) of the transition probability.]

As a result, we come to the Fokker–Planck equation in I-form:

$$\frac{\partial f(x, t)}{\partial t} = \frac{\partial^2}{\partial x^2} [D(x) f(x, t)] + \frac{\partial}{\partial x} [A(x) f], \tag{6.14}$$

where the coefficients of diffusion and friction are defined by

$$\begin{aligned} D(x) &= \frac{1}{2} \int (\Delta x)^2 W(\Delta x, x) d\Delta x, \\ A(x) &= \int \Delta x W(\Delta x, x) d\Delta x, \end{aligned} \tag{6.15}$$

which are close in form to Eqn (6.8).

So we see that different expansions in Δx lead to different forms of the Fokker–Planck equation. As we have already noted in Section 3, the choice between them has to be based on additional physical considerations. Now we are going to continue discussing this point.

6.2 Stationary solution of the Fokker–Planck equation

As indicated above, the replacement (6.10) allows one to obtain all three forms of the Fokker–Planck equation from Eqn (6.9). For the one-dimensional case the general stationary solution of these equations can be written in the form

$$f(x) = \frac{C}{[D(x)]^{\nu/2}} \exp \left[- \int_0^x \frac{A(x')}{D(x')} dx' \right], \quad \int f dx = 1 \tag{6.16}$$

where $\nu = 2$ for the I-form, $\nu = 1$ for the S-form, and $\nu = 0$ for the K-form of the Fokker–Planck equation. Only in the last case is the structure of the stationary solution

simple and is completely defined by the ratio of the fluctuation factor $D(x)$ and the dissipation factor $A(x)$.

Elsewhere we have considered the stationary solution of the Fokker–Planck equation for Brownian motion in a medium with nonlinear friction [see Eqn (3.20)]. At equilibrium, the coefficients $D(v), \gamma(v)$ satisfy the Einstein relation (3.4), and we come to the Maxwell distribution. When the master equation (5.9) [or (6.2)] is used, the situation is in general more complicated.

Master equations are used not only for systems in the thermostat, when the motion of Brownian particles occurs in the medium which is at equilibrium. They also describe relaxation in media which are in a stationary but not equilibrium state. In such a situation one might question the validity of the Einstein relation. We shall see, however, that it is both possible and necessary to use this relation.

Assume once again that the generalised coordinate x in the master equation can be interpreted as the velocity of the ‘Brownian particle’. Expressed in terms of this velocity can be, for instance, the electric current in a self-oscillatory system (Van der Pol oscillator). Then the coefficients of diffusion $D(v)$ and friction $A(v)$ will depend also on the coefficient of feedback a_f . Owing to the presence of feedback, the stationary state will be other than the state of equilibrium. From Eqn (6.16) we find that

$$f(v, a_f) = \frac{C}{[D(v, a_f)]^{\nu/2}} \exp \left[- \int_0^v \frac{A(v', a_f)}{D(v', a_f)} dv' \right], \quad \int f dv = 1, \tag{6.17}$$

with the above values of ν for the three forms of the Fokker–Planck equation.

It would be natural to assume that in the absence of feedback ($a_f = 0$) the stationary state coincides with the state of equilibrium. Then the coefficients of diffusion and friction

$$D(v, a_f = 0) = D(v), \quad A(v, a_f = 0) = A(v) \equiv \gamma(v)v \tag{6.18}$$

satisfy the Einstein relation (3.4), and the distribution $f(v)$ coincides with the Maxwell distribution. The latter only occurs for the K-form of the Fokker–Planck equation — that is, when $\nu = 0$.

This again brings us to the conclusion that the kinetic form of the Fokker–Planck equation is preferable from the standpoint of statistical theory. This conclusion will be corroborated with numerous concrete examples in the sections to follow.

Now we are going to study the master equation for a system of atoms interacting with an electromagnetic field. Here the atoms act as Brownian particles, and the fluctuating electromagnetic field as the medium.

7. The master equation for a system of atoms in a electromagnetic field

The kinetic theory of atoms and fields has advanced greatly in recent years to match the progress in quantum electronics [9, 20–22, 31–34]. This theory stems from Einstein’s classic paper of 1916 [35], in which he formulated the first equation of balance for atoms at rest and equilibrium in a field, and introduced the coefficients of induced and spontaneous emission (Einstein’s coefficients).

Consider the most simple model when the atoms are at rest and are homogeneously distributed in space. Then the state of the atoms is characterised by the distribution

function f_n for atoms of internal energy E_n . In this way, the state of the atoms is defined by a discrete set of variables n . For the case in question, the kinetic equation can be written in the form [9, 33,]

$$\frac{\partial f_n}{\partial t} = \sum_m \left[B_m^n \frac{(\delta E \cdot \delta E)_{\omega_{nm}}}{4\pi^2} (f_m - f_n) - \frac{1}{2} A_m^n (f_m + f_n) \right] = I_n, \quad (7.1)$$

where

$$B_m^n = \frac{4\pi^2 |d_{nm}|^2}{3\hbar^2}, \quad A_m^n = \frac{4|d_{nm}|^2}{3\hbar c^3} \omega_{nm}^3 \equiv \gamma_m^n, \quad (7.2)$$

are Einstein's coefficients which are found in the course of derivation of the kinetic equation (7.1). For the system of atoms in an equilibrium electromagnetic field the spectral density of fluctuations of the electric field is

$$\frac{1}{4\pi^2} (\delta E \cdot \delta E)_\omega \equiv \rho_\omega = \frac{\omega^2}{\pi^2 c^3} \frac{1}{2} \hbar \omega \coth \frac{\hbar \omega}{2kT} \\ \equiv \frac{\omega^2}{\pi^2 \omega^3} kT \omega, \quad \omega = \omega_{nm}, \quad (7.3)$$

where ρ_ω is Planck's distribution for the mean energy of equilibrium electromagnetic radiation including the zero-point energy.

At equilibrium, the solution of the kinetic equation (7.1) is the Gibbs–Boltzmann distribution

$$f_n = \frac{1}{Z} \exp\left(-\frac{E_n}{kT}\right), \quad Z = \sum_n \exp\left(-\frac{E_n}{kT}\right). \quad (7.4)$$

It is important that the ‘collision integral’ includes only the spectral density at the transition frequencies ω_{nm} . This corresponds to the approximation of infinitesimally narrow resonances, when the spectral density of fluctuations is represented by a set of infinitesimally narrow spectral lines. The essence of this approximation is discussed in detail in Refs [17, 36]; it corresponds to the so-called ‘collisionless approximation’ in the calculation of small-scale fluctuations, the exclusion of which leads to irreversible kinetic equations.

Let us compare the kinetic equation (7.1) with master equation (6.2). This will allow us to particularise the expressions for the transition probabilities $W^{s,a}$ for the case in question:

$$W_{nm}^s = B_m^n \frac{(\delta E \cdot \delta E)_{\omega_{nm}}}{4\pi^2}, \quad W_{nm}^a = \frac{1}{2} A_m^n \equiv \gamma_m^n. \quad (7.5)$$

Using Eqns (7.2), (7.3), and (7.5), we find the connection between the transition probabilities W^s, W^a :

$$W_{nm}^s = W_{nm}^a \coth \frac{\hbar \omega_{nm}}{2kT}. \quad (7.6)$$

In case of the system of atoms in an equilibrium electromagnetic field, this equation links the fluctuation characteristic W_{nm}^s to the dissipation characteristic W_{nm}^a , and particularises the general fluctuation–dissipation relation (FDR) (6.3) which also holds good for nonequilibrium stationary states.

Observe that the argument of \coth is not the current frequency of the spectrum, but the transition frequency ω_{nm} . This structure of the FDR is typical of quantum systems [17, 36].

Let us quote the expressions for the coefficients of diffusion and friction [in the general case they are defined by Eqn (6.8)]. We go over from continuous variables x, x'

to the corresponding discrete variables n, m and make substitutions similar to Eqn (6.4). Then the expressions (7.5) for the probabilities of transition take on the form

$$W_{n,m}^{s,a} = W_{n-m, (n+m)/2}^{s,a} \equiv W_{\Delta_{nm}, n-\Delta_{nm}/2}^{s,a}, \quad \Delta_{nm} = n - m. \quad (7.7)$$

We assume, like we did in case of continuous variables, that the dependence on Δ_{nm} is strong, but at the same time it is possible to expand the argument $n - \Delta_{nm}/2$ in Δ_{nm} . As a result, we come to the following expressions for the local (n -dependent) coefficients of diffusion and friction:

$$D_n = \frac{1}{2} \sum_{\Delta_{nm}} (\Delta_{nm})^2 W_{\Delta_{nm}, n}^s, \quad A_n = \sum_{\Delta_{nm}} \Delta_{nm} W_{\Delta_{nm}, n}^a, \quad (7.8)$$

which are similar to Eqn (6.8) above. In the next section these results will be particularised for the case of the quantum atom oscillator.

To end this section, we quote the equation of balance of the mean energy, which follows from the kinetic equation (7.1):

$$\langle E \rangle = \sum_n E_n f_n(t), \\ \frac{d\langle E \rangle}{dt} = \sum_{nm} \gamma_m^n (f_m - f_n) \left(kT \omega_{nm} - \frac{1}{2} \hbar \omega_{nm} \frac{f_m + f_n}{f_m - f_n} \right). \quad (7.9)$$

At equilibrium the right-hand side of the equation of balance is zero, and the mean energy is defined by Planck's formula.

So we have succeeded in particularising the expressions for the transition probabilities $W^{s,a}$ for the system of atoms and field. As follows from Eqns (7.3)–(7.6), they are defined in terms of atomic characteristics: the matrix element of the dipole moment d_{nm} , the transition frequency ω_{nm} , and the field temperature—that is, the temperature of the medium where the Brownian motion of atoms takes place. To refine our results even further, we must consider a particular model of the atom.

8. Brownian motion of quantum atom oscillators

8.1 The master equation

Let us return to the kinetic equation (7.1), and consider the atom as a one-dimensional quantum oscillator. Such an ‘atom oscillator’ (that is, Brownian particle) can be visualised as a small but macroscopic electric circuit. By ω_0 we denote the eigenfrequency of the oscillator; then the square of the matrix element can be represented as

$$|d_{nm}|^2 \rightarrow |x_{nm}|^2 = \frac{\hbar}{m\omega_0} \left(\frac{m}{2} \delta_{m-1,n} + \frac{m+1}{2} \delta_{m+1,n} \right), \quad (8.1)$$

and we come to the following expression for the ‘collision integral’ I_n in the kinetic equation (7.1):

$$I_n = \gamma(\omega_0) \left\{ \coth \frac{\hbar \omega_0}{2kT} \left[\frac{n+1}{2} (f_{n+1} - f_n) \right. \right. \\ \left. \left. + \frac{n}{2} (f_{n-1} - f_n) \right] - \left[-\frac{n+1}{2} (f_{n+1} + f_n) \right. \right. \\ \left. \left. + \frac{n}{2} (f_{n-1} + f_n) \right] \right\}, \quad \omega_0 = \omega_{n+1,n}, \quad (8.2)$$

where

$$\gamma(\omega_0) = \frac{2e^2\omega_0^2}{3mc^3}, \quad (8.3)$$

is the coefficient of radiation friction.

Now we can write the equation of balance of the mean energy:

$$\langle E \rangle = \sum_n \left(n + \frac{1}{2} \right) \hbar\omega_0 f_n. \quad (8.4)$$

Two forms of the equation in the mean energy $\langle E \rangle$ are useful,

$$\frac{d\langle E \rangle}{dt} = \gamma(\omega_0)(kT_{\omega_0} - \langle E \rangle) \equiv D_{(E)} - \gamma(\omega_0)\langle E \rangle, \quad (8.5)$$

where we have used the definition of the coefficient of diffusion

$$D_{(E)} = \gamma(\omega_0)kT_{\omega_0}, \quad kT_{\omega_0} = \frac{1}{2}\hbar\omega_0 \coth \frac{\hbar\omega_0}{2kT}, \quad (8.6)$$

subscript (E) indicates that $D_{(E)}$ is the coefficient of diffusion with respect to the energy values. This formula may be regarded as the quantum generalisation of the classic Einstein relation (2.5).

Now let us look at expressions (7.8) which define the local coefficients of diffusion and friction in the zero approximation with respect to Δ_{nm}/n . For a quantum atom oscillator they can be rewritten as

$$D_n = \gamma(\omega_0) \frac{1}{2} \coth \frac{\hbar\omega_0}{2kT} n \equiv \frac{D_{(E)}}{\hbar\omega_0} n, \quad A_n = \gamma(\omega_0)n, \quad (8.7)$$

where use has been made of definitions (8.3) and (8.4) for the coefficients of radiation friction and diffusion, respectively. We see that the local coefficients of diffusion and friction are linear functions of n . Making use of Eqn (8.7), we can reduce the quantum ‘collision integral’ (8.2) to a more convenient form:

$$I_n(t) = [D_{n+1}(f_{n+1} - f_n) - D_n(f_n - f_{n-1})] + \frac{1}{2}[A_{n+1}(f_{n+1} + f_n) - A_n(f_n + f_{n-1})]. \quad (8.8)$$

We see that the right-hand side contains two induced contributions, which are proportional to the relevant coefficients of diffusion. Their signs are determined by the relative population of the adjacent levels. The last two terms are proportional to the relevant coefficients of friction. The first of these is positive, since it corresponds to the increase in population at the expense of the higher level. The second is negative and corresponds to the escape to the lower level.

At equilibrium, the diffusion and the dissipation terms cancel out in pairs by virtue of the fluctuation–dissipation relation (8.6).

Observe that for the quantum atom oscillator the matrix elements are defined by Eqn (8.1), which implies that the transition probabilities in variable n can only change by ± 1 in the course of time evolution, as described by the master equation (7.1). Processes of this kind are commonly referred to as *one-step processes*. Accordingly, the quantum kinetic equation with ‘collision integral’ (8.2) or (8.8) is an example of the master equation for a *one-step process*. Since the local coefficients of diffusion and friction given by (8.7) are linear functions of n , we are dealing here with a linear *one-step process*.

The general structure of master equations for one-step processes will be discussed in the next section. We shall once again see the ambiguity of such equations. Prior to that, however, we shall prove that our current master equation corresponds to the canonical form of the Fokker–Planck equation.

8.2 The Fokker–Planck equation

Transition to the Fokker–Planck equation is based on the expansion in the inverse quantum number $1/n$, which implies that from the discrete spectrum of oscillator energy values we go over to the continuous spectrum. Since for large n , $E = n\hbar\omega_0$, we come to the following equation for the distribution function of energy:

$$\frac{\partial f(E, t)}{\partial t} = \frac{\partial}{\partial E} \left(D_{(E)} E \frac{\partial f}{\partial E} \right) + \frac{\partial}{\partial E} (\gamma E f), \quad \int_0^\infty f dE = 1. \quad (8.9)$$

The equilibrium solution of the Fokker–Planck equation (8.9) is the Boltzmann distribution with the quantum temperature given by Eqn (8.6):

$$f(E) = \frac{1}{kT_{\omega_0}} \exp\left(-\frac{E}{kT_{\omega_0}}\right), \quad \langle E \rangle = kT_{\omega_0}. \quad (8.10)$$

Examples of such equations for an electric circuit can be found in Refs [17, 36]; the emf is then given by

$$\langle E^2 \rangle_\omega = 2RkT_{\omega_0}, \quad (8.11)$$

which in the classical approximation coincides with the well-known Nyquist formula.

9. Master equations for one-step processes

9.1 Traditional definition of transition probability

For description of one-step processes we return to the master equation (5.9) and introduce discrete variable x in place of continuous variable n . The relevant equation is

$$\frac{\partial f_n}{\partial t} = \sum_{n'} [W_{nn'} f_{n'}(t) - W_{n'n} f_n(t)], \quad \sum_{n=0}^\infty f_n(t) = 1. \quad (9.1)$$

The following expression is traditionally used for the transition probability [11, 14]

$$W_{nn'} = g_{n'} \delta_{n, n'+1} + r_{n'} \delta_{n, n'-1}. \quad (9.2)$$

It is assumed therefore that one event corresponds either to *emergence (birth)* at state n , or to *disappearance* from state n (*death*). This parlance is used in the theory of populations. In semiconductor theory one refers to g_n as the coefficient of *generation*, and to r_n as the coefficient of *recombination*.

Substitution of Eqn (9.2) into Eqn (9.1) results in the following master equation:

$$\frac{\partial f_n}{\partial t} = g_{n-1} f_{n-1} + r_{n+1} f_{n+1} - (g_n + r_n) f_n, \quad \sum_{n=0}^\infty f_n = 1. \quad (9.3)$$

(we assume that n varies from zero to infinity, $0 \leq n \leq \infty$). Equation (9.3) must be supplemented by ‘boundary conditions’

$$r_0 = 0, \quad g_{-1} = 0$$

and therefore

$$\frac{\partial f_0}{\partial t} = r_1 f_1 - g_0 f_0. \quad (9.4)$$

These conditions forbid escape (recombination) from the lowest state and generation from states with negative numbers n . The local coefficients of diffusion and friction are defined by combinations of r_n , g_n :

$$D_n = \frac{1}{2}(r_n + g_n), \quad A_n = r_n - g_n. \quad (9.5)$$

The master equation (9.3) is widely used for describing many diverse processes [11, 14]: radioactive decay, shot noise, processes of chemical kinetics, and various ‘predator–prey’ (Volterra) systems. This equation works well whenever the main feature of the phenomenon in question is the competition between *birth* and *death*, or *ionisation* and *recombination*. As a rule, such cases are dominated by nonequilibrium (although perhaps stationary) states, while the stationary state becomes less significant. In such situations the use of functions g_n , r_n as the main characteristics is perfectly justified.

Equilibrium, however, remains a fundamental concept. It is this state that is stable when the controlling factors are switched off, and which corresponds to the highest degree of chaos. It would be natural to demand therefore that master equations should describe, in particular, the processes of evolution towards equilibrium. Such equations may be more conveniently formulated not in terms of functions g_n , r_n but in terms of the local coefficients of diffusion and friction, which are related to the former by Eqn (9.5). We have seen this in case of Brownian motion of quantum atom oscillators in an equilibrium electromagnetic field.

To confirm this point, let us consider some implications of the master equation (9.3).

The equation for the first moment of the distribution f_n is obtained with the aid of the master equation (9.5) and has the form

$$\frac{d\langle n \rangle}{dt} = -(\langle r_n \rangle - \langle g_n \rangle) = -\langle A_n \rangle. \quad (9.6)$$

Accordingly, the relaxation of the first moment is determined by the mean value of the local coefficient of friction. In the case of the linear one-step process, when

$$D_n = Dn, \quad A_n = (r - g)n \equiv \gamma n, \quad (9.7)$$

equation (9.6) becomes

$$\frac{d\langle n \rangle}{dt} = -\gamma \langle n \rangle. \quad (9.8)$$

We see that the value of $\langle n \rangle$ relaxes towards zero. If, by way of example, we use formulas (9.8) for a quantum atom oscillator, the equation for the mean energy becomes

$$\frac{d\langle E \rangle}{dt} = -\gamma(\omega_0)\langle E \rangle, \quad \gamma(\omega_0) = \frac{2e^2\omega_0^2}{3mc^3}. \quad (9.9)$$

As opposed to Eqn (8.5), which follows from the master equation with collision integral (8.2) or (8.8), Eqn (9.9) does not describe relaxation towards equilibrium with the thermostat.

This disadvantage of the master equation (9.3) also manifests itself in the structure of the corresponding

Fokker–Planck equation. Indeed, expansion in $1/n$ results in the Fokker–Planck equation in Ito’s form

$$\frac{\partial f(n, t)}{\partial t} = \frac{\partial^2}{\partial n^2} [D_n f(n, t)] + \frac{\partial}{\partial n} [A_n f(n, t)]. \quad (9.10)$$

From this one can also obtain Eqn (9.8). The stationary solution of this equation has the structure (6.16) with $\nu = 2$, and does not agree with the Einstein formula.

We see that the traditional definition of transition probability (9.2) for one-step processes leads to a number of results which contradict the main assumptions of the statistical theory. Our immediate task will therefore be to try and define the probability of transition in such a way as to overcome these difficulties.

9.2 Nontraditional definition of transition probability

We use the general form of the master equation (6.2) and define the symmetrical and antisymmetrical parts of transition probability as [19]

$$W_{nn'}^s = D_{n+1}\delta_{n+1, n'} + D_n\delta_{n-1, n'} = W_{n'n}^s, \\ W_{nn'}^a = -\frac{1}{2}[A_{n+1}\delta_{n+1, n'} - A_n\delta_{n-1, n'}] = -W_{n'n}^a. \quad (9.11)$$

Substituting these expressions into Eqn (6.2), we get the master equation for one-step processes which is different from Eqn (9.3). The ‘collision integral’ is now defined by Eqn (8.8), and the local coefficients of diffusion and friction are, as before, linked with the coefficients of generation and recombination by Eqn (9.5).

Recall that for the linear one-step process, when the coefficients of diffusion and friction are defined by Eqn (8.7), we come to the kinetic equation for quantum atom oscillators in an equilibrium electromagnetic field.

Expression (8.8) for the collision integral has a clear-cut physical meaning. The diffusion is responsible for the ‘induced’ transitions, and the signs of the respective terms depend on the relative population of the adjacent states.

The traditional definition of transition probability allows one to separate the symmetrical and antisymmetrical parts $W_{nn'}^{s,a}$ which are expressed via the coefficients of diffusion and friction. The resulting expressions, however, defy straightforward physical interpretation.

Let us discuss some properties of the nontraditional master equation for one-step processes. For the state of equilibrium, from Eqn (8.8) follows the relationship between the local coefficients of diffusion and friction

$$D_n = \frac{1}{2} A_n \frac{f_n + f_{n+1}}{f_n - f_{n+1}}, \quad (9.12)$$

which is a fluctuation–dissipation relation. Since the coefficients of diffusion and friction are positive, the higher level at equilibrium is less populated. In general, the distribution function cannot be expressed in terms of the coefficients of diffusion and friction with the aid of Eqn (9.12). For the linear one-step process, when the formulae in Eqn (8.7) hold good, the Boltzmann distribution

$$f_n = \frac{1}{Z} \exp\left(-\frac{E_n}{kT\omega_0}\right), \quad \sum_n f_n = 1, \quad (9.13)$$

follows from Eqn (9.12). The distribution with respect to n is, therefore, exponential.

There are other implications of the master equation obtained in this section. The equation for the mean value of $\langle n \rangle$ differs from Eqn (9.6) and has the form

$$\frac{d\langle n \rangle}{dt} = \frac{1}{2} \left[(g_{n+1} - r_{n-1}) - (r_n - g_n) \right]. \quad (9.14)$$

To understand the physical meaning of this difference, let us consider the approximation of large n , when we can go over to the continuous variable. Retaining only the main terms, we arrive at the following equation:

$$\frac{d\langle n \rangle}{dt} = \left\langle \frac{dD_n}{dn} \right\rangle - (\langle r_n \rangle - \langle g_n \rangle) \equiv \left\langle \frac{dD_n}{dn} \right\rangle - \langle A_n \rangle. \quad (9.15)$$

This equation differs from Eqn (9.6) in that it contains an additional term which is determined by the diffusion. For the linear one-step process—when the coefficients of diffusion and friction are defined by Eqn (8.7)—from Eqn (9.15) follows the equation of balance of mean energy of quantum atom oscillators in an equilibrium electromagnetic field,

$$\frac{d\langle E \rangle}{dt} = \gamma(\omega_0)(kT_{\omega_0} - \langle E \rangle), \quad (9.16)$$

which, in contrast to Eqn (9.9), does describe the process of relaxation towards the equilibrium value of mean energy.

Finally let us consider the corresponding Fokker–Planck equation. Unlike Eqn (9.10), it now has the canonical form

$$\frac{\partial f_n}{\partial t} = \frac{\partial}{\partial n} \left(D_n \frac{\partial f_n}{\partial n} \right) + \frac{\partial}{\partial n} \left(A_n f_n \right). \quad (9.17)$$

Because of this, the equilibrium solution is completely defined by the ratio of fluctuation and dissipation factors,

$$f_n = C \exp \left(- \int_0^n \frac{A_{n'}}{D_{n'}} dn' \right). \quad (9.18)$$

For the linear one-step process—when the local coefficients of diffusion and friction are defined by Eqn (8.7)—from Eqn (9.17) follows the Fokker–Planck equation (8.9) for the distribution function of energy values, and the Boltzmann distribution (8.10) follows from Eqn (9.18).

In the sections to follow we shall illustrate the difference between alternative descriptions of stochastic processes with concrete examples. First, however, we are going to explore the possibility of transition from the Fokker–Planck equation for the distribution $f(r, v, t)$ in the phase space of coordinates and velocities to the Einstein–Smoluchowski equation for the simpler distribution $f(r, t)$. We shall see that this task is very similar to the problem of transition from the kinetic equation to equations of gas dynamics, and is associated with similar difficulties [27, 28]. These difficulties can and will be overcome in the theory of Brownian motion by going over to the description of nonequilibrium processes based on the generalised kinetic equations. We shall see that these equations work especially well in the case of Brownian motion in nonlinear active media.

In particular, the generalised kinetic equations will help us to draw the limits of applicability of reaction–diffusion equations, such as the famous Ginzburg–Landau equation. It will be possible to go beyond the limitations of these equations so as to obtain information concerning the higher moments (which is important in the neighbourhood of

critical points), and to construct a more consistent theory of nonequilibrium large-scale (also known as coarse-grained) fluctuations (kinetic, hydrodynamic, reaction–diffusion fluctuations).

10. Spatial diffusion. The Einstein–Smoluchowski equation

So far, in the Langevin equations and in the kinetic equations, we have assumed that the medium which hosts Brownian motion is practically unlimited. Now, in addition to the internal parameters D, γ we introduce the external parameter, the characteristic size of the system L . This gives rise to a new parameter of time, the diffusion time

$$\tau_D = \frac{L^2}{D_r}, \quad (10.1)$$

where D_r is the coefficient of spatial diffusion. Earlier for the description of Brownian motion we used the coefficient of diffusion in the space of velocities $D \equiv D_v$. Now the meaning of coefficient D in equations (6.9), (6.14) depends on the interpretation of the generalised coordinates x .

When the Langevin equations (2.2) are used, the correlation time of the source is $\tau_{cor} = 0$. Nonzero are two time-like parameters, $\tau_{rel} = 1/\gamma$ and τ_D . If the diffusion time is much greater than the relaxation time, it would be natural to anticipate the feasibility of transition from the Fokker–Planck equation for the distribution $f(r, v, t)$ to the Einstein–Smoluchowski equation for the simpler distribution $f(r, t)$. There are two ways of doing this but only at constant γ !

10.1 Spatial diffusion. The Langevin method

We return to the Langevin equations (2.2) for a Brownian particle, and assume that the external field is absent ($U = 0$), and that the diffusion is slow, $\tau_D \gg \tau_{rel}$. It would then be natural to neglect the velocity derivative dv/dt in the Langevin equations as small compared to γv . Eliminating the velocity, we come to the Langevin equation in the coordinate

$$\frac{dr}{dt} = \frac{y(t)}{\gamma} \equiv y_r(t), \quad (10.2)$$

where $y_r(t)$ is the Langevin source which determines the displacement of the Brownian particle kicked about by the atoms of the medium. The moments of this source are

$$\langle y_r(t) \rangle = 0, \quad \langle y_r(t) y_r(t') \rangle = 3[2D_r \delta(t - t')],$$

$$D_{(r)} = \frac{D}{\gamma^2} = \frac{kT}{M\gamma}. \quad (10.3)$$

The last of these defines the linkage between the coefficients of diffusion in conventional space and in velocity space.

From the Langevin equation (10.2) one can go over to the relevant equation for the distribution function $f(r, t)$. Following the guidelines set forth in Section 3, we come to the Einstein–Smoluchowski equation in $f(r, t)$:

$$\frac{\partial f}{\partial t} = D_{(r)} \Delta_r f, \quad \int f(r, t) \frac{dr}{V} = 1, \quad n(r, t) = Nf(r, t). \quad (10.4)$$

It is natural that this equation coincides with the familiar equation of diffusion. We have also defined the density of Brownian particles. The analytical solution of this equation

is well known. We shall only quote expressions for the moments of displacement $r - r_0$ of the Brownian particle:

$$\langle r - r_0 \rangle = \mathbf{0}, \quad \langle (r - r_0)^2 \rangle = 3[2D_{(r)}(t - t_0)]. \quad (10.5)$$

The second equation states that the mean square displacement of the Brownian particle is proportional to time (Einstein's formula).

10.2 Diffusion of a Brownian particle in an external field

When the external force $\#$ is taken into account in the description of slow processes, Eqn (10.2) includes a new term and becomes

$$\frac{d\mathbf{r}}{dt} = -\frac{1}{M\gamma} \frac{\partial U}{\partial \mathbf{r}} + \mathbf{y}_r(t). \quad (10.6)$$

The moments of the Langevin source are, as before, given by Eqns (10.3). Transition to the kinetic equation follows the familiar scheme. As a result, we come to a more general Einstein–Smoluchowski equation

$$\begin{aligned} \frac{\partial f(\mathbf{r}, t)}{\partial t} &= D_{(r)} \frac{\partial^2 f}{\partial |\mathbf{r}|^2} - \frac{\partial}{\partial \mathbf{r}} \left[\frac{\mathbf{F}(\mathbf{r})}{M\gamma} f \right], \\ D_{(r)} &= \frac{kT}{M\gamma}, \quad \mathbf{F}(\mathbf{r}) = -\frac{\partial U}{\partial \mathbf{r}}, \end{aligned} \quad (10.7)$$

which is also known as the Kramers equation. Its equilibrium solution is the Boltzmann distribution

$$f(\mathbf{r}, t) = \exp \left[-\frac{U(\mathbf{r})}{kT} \right] \left[\int \exp \left(-\frac{U(\mathbf{r}')}{kT} \right) d\mathbf{r}' \right]^{-1}. \quad (10.8)$$

So, we have obtained the equation for the distribution function $f(\mathbf{r}, t)$ for two cases: (1) free Brownian particles, and (2) particles in an external field. In both cases we started with the Langevin equation under the condition of slow motion $d\mathbf{v}/dt \ll \gamma\mathbf{v}$. The meaning of this condition may be quite different, however, depending on the form of the potential $U(\mathbf{r})$. Let us look at this matter more closely.

Recall that we considered the free motion of particles under the condition

$$\tau_D = \frac{L^2}{D_{(r)}} \gg \tau_{\text{rel}} = \frac{1}{\gamma}, \quad (10.9)$$

which contains the squared size of the system, and therefore holds as long as the system is large enough.

In the presence of an external field the situation becomes very different if the field restricts the movement of particles. By $U(\mathbf{r})$ we denote the characteristic size of the region, and consider two examples which are important for our subsequent discussion.

1. *Harmonic oscillator*: $F = -M\omega_0^2 r$ is the elastic force. Then the equilibrium solution (10.8) coincides with the Gaussian distribution

$$U(\mathbf{r}) = \frac{M\omega_0^2 |\mathbf{r}|^2}{2}, \quad |\mathbf{r}|_0^2 \sim \langle |\mathbf{r}|^2 \rangle = \frac{kT}{M\omega_0^2}. \quad (10.10)$$

Now L acts as r_0 , and condition (10.9) becomes

$$\tau_D = \frac{\gamma}{\omega_0^2} \gg \frac{1}{\gamma} \quad \text{and therefore,} \quad \gamma \gg \omega_0. \quad (10.11)$$

We see that the Kramers equation (10.7) holds for a bounded Brownian particle (harmonic oscillator) only when the damping is strong (overdamped oscillator). This situation is of certain practical interest—for instance, in

connection with the Brownian motion of fragments of polymer molecules. No less interesting, however, is the opposite extreme, when the damping is weak.

2. *Brownian particle as a bistable element*: the elastic force is nonlinear, and the potential is given by

$$U(\mathbf{r}) = \frac{M\omega_0^2 |\mathbf{r}|^2}{2} \left(-a + \frac{b}{2} r^2 \right), \quad a = a_f - 1, \quad b > 0. \quad (10.12)$$

The coefficient a_f characterises the action of an effective field—for instance, the Lorentz field in a dielectric [33, 37]. When this coefficient is large enough, so that $a > 0$, the coefficient of elasticity becomes negative, and the system becomes bistable. As in the case of self-oscillatory systems (open active systems), the coefficient a_f can be referred to as the coefficient of feedback.

As a rule, feedback is the property of the medium in which Brownian motion takes place. The coefficient of nonlinearity b may have a different nature; we shall distinguish two possible cases.

(a) Nonlinearity is the property of the individual element of the system (Brownian particle), rather than the property of the medium. The equilibrium solution of Eqn (10.7) is then represented by the Boltzmann distribution (10.8) with the potential given by Eqn (10.12). If $a < 0$, the Boltzmann distribution has one maximum at $x = 0$. The behaviour at large x is determined by the term with coefficient b . If $a > 0$, the Boltzmann distribution has two maxima, and the system is bistable.

(b) Both the coefficient of feedback and the coefficient b depend on the characteristics of the medium. In the absence of feedback ($a_f = 0$) we come to the Boltzmann distribution for a harmonic oscillator. This implies that at $a_f = 0$ the system has the highest possible symmetry—in other words, the system is in the most chaotic state.

From the condition of existence of the Boltzmann distribution in the medium with nonlinear elasticity, one can define the coefficient of diffusion as a function of coordinates,

$$D_{(r)}(\mathbf{r}) = D_{(r)}(1 + b|\mathbf{r}|^2), \quad D_{(r)} = \frac{kT}{M\gamma}. \quad (10.13)$$

Given this, the equilibrium distribution (for any value of a_f) can be represented as

$$\begin{aligned} f(\mathbf{r}) &= C \exp \left[-\frac{U_{\text{eff}}(\mathbf{r})}{kT} \right], \\ U_{\text{eff}}(\mathbf{r}) &= \frac{M\omega_0^2}{2} \left[|\mathbf{r}|^2 - \frac{a_f}{b} \ln(1 + b|\mathbf{r}|^2) \right], \end{aligned} \quad (10.14)$$

where we have introduced the effective potential which takes into account the lowering of thermostat symmetry as the coefficient a_f increases. The change may be caused, for instance, by the change in temperature or density.

Eventually we come to the Einstein–Smoluchowski equation with the variable coefficient of spatial diffusion:

$$\begin{aligned} \frac{\partial f(\mathbf{r}, t)}{\partial t} &= \frac{\partial}{\partial \mathbf{r}} \left[D_{(r)}(\mathbf{r}) \frac{\partial f}{\partial \mathbf{r}} \right] - \frac{\partial}{\partial \mathbf{r}} \left[\frac{\mathbf{F}(\mathbf{r})}{M\gamma} f \right], \\ F(\mathbf{r}) &= -M\omega_0^2 r(1 - a_f + br^2). \end{aligned} \quad (10.15)$$

For $a_f = 0$ (the highest possible symmetry) we obtain the Einstein–Smoluchowski equation for a linear oscillator.

Presently we shall illustrate the use of the more general equation (10.15).

The change in the symmetry of the thermostat can be taken into account in a different way. Namely, the coefficient of diffusion remains the same, whereas the force $F(r)$ is replaced by the relevant effective force

$$F(r) \rightarrow F_{\text{eff}}(r) = -\frac{\partial U_{\text{eff}}}{\partial r} = -M\omega_0^2 r \left(1 - \frac{a_f}{1 + b|r|^2} \right). \quad (10.16)$$

As a result, we get the following Einstein–Smoluchowski equation:

$$\frac{\partial f(r, t)}{\partial t} = D_{(r)} \frac{\partial^2 f}{\partial |r|^2} - \frac{\partial}{\partial |r|^2} \left[\frac{F_{\text{eff}}}{M\gamma} f \right], \quad D_{(r)} = \frac{kT}{M\gamma}, \quad (10.17)$$

the equilibrium solution of which is, as before, the distribution given by Eqn (10.14).

We see that in the presence of a nonlinear potential (10.12) the thermostat (the surrounding medium) may act on the Brownian particles in different ways. In the first case one may speak of the *linear thermostat*, since the nonlinearity is the property of each individual Brownian particle. The surrounding medium only affects the characteristic frequency: $\omega_0^2 \rightarrow \omega_0^2(1 - a_f)$. The value of $a_f = 1$ corresponds to the bifurcation point (the appearance of a ‘soft mode’). In the second case one may refer to a *nonlinear thermostat*, since the nonlinearity of the force acting on the Brownian particle is due to the surrounding medium.

10.3 Comparison of stationary distributions in ‘linear’ and ‘nonlinear’ thermostats

The above two cases correspond to different Einstein–Smoluchowski equations and, as a consequence, to different stationary solutions: the Boltzmann distribution with the potential given by Eqn (10.12), and the distribution given by Eqn (10.14). It is only the latter that at $a_f = 0$ coincides with the Boltzmann distribution for Brownian motion of harmonic oscillators. The distinction between these two distributions manifests itself, in particular, in the different behaviour (at large values of r) at the ‘tails’ of the distributions.

For that matter, for the linear thermostat the fall-off of the distribution at large values of r is controlled by the nonlinear factor $\exp(-br^4)$ and is therefore much faster than in the case of the Boltzmann distribution with the harmonic oscillator potential given by Eqn (10.10). By contrast, the distribution given by (10.14), as a_f increases (that is, as the symmetry becomes lower), falls off at large values of r much slower than the Boltzmann distribution for a linear oscillator.

Let us consider some other characteristics using the example of one-dimensional motion.

(1) The locations of maxima coincide:

$$x_{\text{max}} = 0 \quad \text{if} \quad a_f < 1, \\ x_{\text{max}} = \pm \left(\frac{a_f - 1}{b} \right)^{1/2} \quad \text{if} \quad a_f > 1. \quad (10.18)$$

(2) The ratios of distribution functions at $x = x_{\text{max}}$ and $x = 0$ indicate that the relative depth of pit for the symmetrical bistable potential is smaller in the case of the nonlinear thermostat. Because of this, the barrier is surmounted more easily in case of collective nonlinearity.

For instance, in the domain of Gaussian approximation the ratio of dispersions is given by

$$\langle (\delta x)^2 \rangle_{NL} [\langle (\delta x)^2 \rangle_L]^{-1} = a_f, \quad a_f > 1. \quad (10.19)$$

We see that the form of the Einstein–Smoluchowski equation depends on the nature of interaction between the particle and the medium.

The Einstein–Smoluchowski equation has been obtained on the basis of the Langevin equation (10.6), which follows in its turn from the more general set of Langevin equations (2.2) for position and velocity of a Brownian particle. As simplifying assumptions we used either the condition of slow spatial diffusion of free particle ($\tau_D \gg \gamma^{-1}$), or the condition of strong damping ($\gamma \gg \omega_0$). The first condition is always satisfied for large systems, since the diffusion time is proportional to the square of the characteristic size of the system L . The second condition is based on the internal parameters, and is far from universal. What can be done if inequality $\gamma \gg \omega_0$ does not hold?

There are also other important questions. How do we describe, for instance, spatial diffusion when the coefficient of friction depends on velocity and we have to deal with dissipative nonlinearity? All these questions are part of the general problem concerning the relationship between the kinetic and the hydrodynamic description of Brownian motion. To begin with, let us consider the possibility of hydrodynamic description for the simplest model of Brownian motion. This will help us later to find solutions for the more complicated cases.

11. Hydrodynamic description of Brownian motion

Let us go back to the Fokker–Planck equation (2.6) for the distribution function $f(r, v, t)$. The temperature of the thermostat enters this equation via the coefficient of diffusion $D_{(v)}$. We are going to use the known scheme of transition from the kinetic Boltzmann equation to the equations of gas dynamics.

In the kinetic description of Brownian motion we are actually dealing with a two-component continuous medium. One of the components is the medium which represents the thermostat. This medium may be at nonequilibrium. The second component of our exemplary continuous medium — when Euler’s method is used — consists of noninteracting Brownian particles. Naturally a more general case is also possible when the interaction of Brownian particles is taken into account [3].

So, we are considering a two-component continuous medium comprised of the thermostat with the temperature T and the ‘continuous medium’ of noninteracting particles. The thermostat is assumed to be linear in the above sense. Then the Brownian motion is described by the kinetic equation (2.6).

The hydrodynamic functions for Brownian particles are $\rho_B(r, t)$, $u_B(r, t)$, $T_B(r, t)$; we shall retain subscript ‘B’ only for the temperature to distinguish it from the temperature of the thermostat. The continuity equation for the density of particles ρ is

$$\frac{\partial \rho}{\partial t} + \frac{\partial \rho u}{\partial r} = 0. \quad (11.1)$$

The equation for the density of momentum of Brownian particles is

$$\frac{\partial \rho u_i}{\partial t} + \frac{\partial \rho u_i u_j}{\partial r_j} = -\frac{\partial p}{\partial r_i} - \frac{\partial \pi_{ij}}{\partial r_j} + \frac{\rho}{m} F_i(r) - \gamma \rho u_i, \quad (11.2)$$

$$p = \frac{\rho}{m} kT_B,$$

where p is the pressure of Brownian particles, and π_{ij} is the so-far unknown ‘tensor of viscous stress’. Finally, we have the equation for the density of kinetic energy,

$$\begin{aligned} \frac{\partial}{\partial t} \left(\frac{\rho |\mathbf{u}|^2}{2} + \frac{3}{2} \frac{\rho}{m} kT_B \right) \\ + \frac{\partial}{\partial r_i} \left[u_i \left(\frac{\rho |\mathbf{u}|^2}{2} + \frac{3}{2} \frac{\rho}{m} kT_B + \rho \right) + \pi_{ij} u_j + S_i \right] \\ = 3\gamma \rho \left[\frac{kT}{m} - \left(\frac{|\mathbf{u}|^2}{3} - \frac{kT_B}{m} \right) \right] + \rho \mathbf{F} \cdot \mathbf{u}, \quad (11.3) \end{aligned}$$

where S_i represents the so-far unknown components of the ‘vector of thermal flux’. The last term on the right-hand side of Eqn (11.2) and the first term on the right-hand side of Eqn (11.3) are the moments of the ‘collision integral’ in the Fokker–Planck equation.

The distribution function $f(\mathbf{r}, t)$ in the Einstein–Smoluchowski equation is linked with the density of Brownian particles by equation $\rho(\mathbf{r}, t) = mnf$. Accordingly, in order to find the desired equation we must eliminate all ‘excessive’ functions \mathbf{u} , T_B , π_{ij} , S_i from equations of the hydrodynamics of Brownian particles. This can only be done by the methods of perturbation theory, under the assumption that the diffusion process described by the Einstein–Smoluchowski equation is the slowest in time and the smoothest in coordinates. In the case of the diffusion of free particles, the first condition is expressed by the inequality $\tau_D \gg \gamma^{-1}$, and the second by demanding that the relevant Knudsen number $\text{Kn} = (v_T/\gamma)/L$ should be small. In other words, the gradients of hydrodynamic functions are assumed to be small.

In the zero approximation with respect to these parameters, equation (11.3) implies that the temperature of Brownian particles is the same as the temperature of the thermostat,

$$T_B = T. \quad (11.4)$$

In case of free Brownian particles ($\mathbf{F} = \mathbf{0}$), the main terms on the right-hand side of Eqn (11.2) are the first and the last. Then, taking into account Eqn (11.4), we find that

$$\rho \mathbf{u} = -\frac{kT}{\gamma m} \frac{\partial \rho}{\partial \mathbf{r}} = -D_{(r)} \frac{\partial \rho}{\partial \mathbf{r}}. \quad (11.5)$$

Substituting this expression into the continuity equation (11.1), we arrive at the Einstein–Smoluchowski equation in function $\rho(\mathbf{r}, t)$ and hence in the distribution function $f(\mathbf{r}, t)$.

In the presence of an external force, given that the process is slow ($\gamma \gg \omega_0$ in case of oscillator) and the Knudsen number is small, we proceed in a similar way. Equation (11.4) still holds, as the correction is proportional to the gradient and is therefore small. In the equation for the density of momentum we have a new term which is proportional to the gradient of the potential, and is only

small given that the potential is smooth enough (again, $\gamma \gg \omega_0$ in the case of an oscillator). Then Eqn (11.5) becomes

$$\rho \mathbf{u} = -D_{(r)} \frac{\partial \rho}{\partial \mathbf{r}} + \frac{\rho}{M\gamma} \mathbf{F}(r). \quad (11.6)$$

Substitution of this expression into the continuity equation (11.1) results in the Einstein–Smoluchowski equation (10.7).

Now a brief summary. The construction of the Einstein–Smoluchowski equation in this section has been based on the kinetic Fokker–Planck equation (2.6) for the distribution function of Brownian particles $f(r, v, t)$ in six-dimensional phase space of coordinates and velocities. The system of noninteracting Brownian particles is thus regarded as a continuous medium interacting with the thermostat. Interaction with the thermostat is characterised by the coefficient of friction γ and the coefficient of diffusion D . Both these coefficients in Eqn (2.6) are constant, which means that the Brownian motion is linear. The force of friction is also nonzero when the thermostat is regarded as a ‘continuous medium’. The coefficient of diffusion is an integral characteristic of the atomic structure of the surrounding medium (the atomic structure of the thermostat).

Transition to the Einstein–Smoluchowski equation has been carried out in two stages. First we made the transition to equations in hydrodynamic characteristics of Brownian particles. The resulting set of equations (11.1)–(11.3) is not closed, being obtained without any simplifying assumptions.

The closed diffusion equation (11.7) is obtained under several important constraints: smoothness of the potential (in particular, the condition $\gamma \gg \omega_0$), and smallness of the Knudsen number. These constraints, together with the assumption that γ and D are constant (linear approximation of Brownian motion), considerably restrict the applicability of the Einstein–Smoluchowski equation. Because of this, we are again faced with the problem of construction of the generalised kinetic equation for unified description of Brownian motion on the kinetic and hydrodynamic (diffusion) scales [28]. In the next section we shall apply the general results of thermodynamics of irreversible processes to the theory of Brownian motion.

12. Evolution of free energy and entropy in Brownian motion. Lyapunov functionals A_F , A_S

12.1 Master equation. H-theorem

By $f_0(x, t)$ we denote the stationary solution of the master equation (5.9), and represent it as the canonical Gibbs distribution. The effective Hamilton function can be defined in a number of ways.

The intensity of the Langevin source, which also defines the coefficient of diffusion, may act as the effective temperature. When the general form of the master equation (5.9) is used, however, there is no explicit information about the structure of the stationary solution and the generally nonlinear coefficient of diffusion. As a result, a clear-cut definition of the effective temperature is not feasible.

There are two possible definitions of the nonequilibrium free energy of the stationary state.

On the one hand, we may formally put the effective temperature equal to unity, and represent the stationary distribution as

$$f_0(x) = \exp[F_{\text{eff}} - H_{\text{eff}}(x)], \quad F_{\text{eff}} = - \int \exp[-H_{\text{eff}}(x)] dx . \quad (12.1)$$

This representation amounts to the inclusion of the effective temperature into the definition of free energy and temperature.

On the other hand, we may define the effective Hamilton function by the following equation:

$$f_0(x) = \exp(-H_{\text{eff}}) (T_{\text{eff}} = 1, \quad F_{\text{eff}} = 0) . \quad (12.2)$$

This representation is especially suitable when the information is derived directly from experiment — for instance, from time realisations of the process $x(t, a)$. We have used this approach in the formulation of the criterion of the relative degree of order of states of open systems in the form of the S-theorem [17, 38].

Now we return to the master equation (5.9) and represent its stationary solution in the form (12.1), thus introducing the free energy of the nonequilibrium stationary state. A ‘thermodynamic relation’ links this quantity with the mean effective energy and the corresponding entropy:

$$F_{\text{eff}} = \int H_{\text{eff}}(x) f_0(x) dx - \int \ln f_0(x) f_0(x) dx . \quad (12.3)$$

In a similar way we define the nonequilibrium free energy for the process of time evolution

$$F(t) = \int H_{\text{eff}}(x) f(x, t) dx - \int \ln f(x, t) f(x, t) dx . \quad (12.4)$$

The difference in the thus defined free energies is reduced to:

$$A_F = F(t) - F_0 = \int \ln \frac{f(x, t)}{f_0(x)} f(x, t) dx \geq 0 . \quad (12.5)$$

We see that the free energy is at a minimum in the stationary state. Now let us show that the free energy monotonically decreases in the course of time evolution towards equilibrium as described by the master equation (5.9). This will prove that A_F is a Lyapunov functional.

We differentiate Eqn (12.5) with respect to time, and use the master equation (5.9). Taking the normalisation condition into account, we get

$$\begin{aligned} \frac{dA_F}{dt} &= \int \ln \frac{f(x, t)}{f_0(x)} \left[W_{xx'} f(x', t) - W_{x'x} f(x, t) \right] dx dx' \\ &= \int W_{xx'} f_0(x') \left[\frac{f(x', t)}{f_0(x')} \ln \frac{f(x, t)}{f_0(x)} - \frac{f(x', t)}{f_0(x')} \ln \frac{f(x', t)}{f_0(x')} \right] dx dx' . \end{aligned} \quad (12.6)$$

To determine the sign of the integrand we take advantage of the equation

$$\int W_{xx'} f_0(x') \left[\frac{f(x, t)}{f_0(x)} - \frac{f(x', t)}{f_0(x')} \right] dx dx' = 0 , \quad (12.7)$$

where we have noted that $f_0(x)$ is the stationary solution, and therefore

$$W_{xx'} f_0(x') - W_{x'x} f_0(x) = 0 . \quad (12.8)$$

By virtue of Eqn (12.7), we may rewrite Eqn (12.6) as

$$\frac{dA_F}{dt} = \int W_{xx'} f_0(x') \left(-a' \ln \frac{a}{a'} - a + a' \right) \leq 0 , \quad (12.9)$$

where we have used the notation

$$a = \frac{f(x, t)}{f_0(x)}, \quad a' = \frac{f(x', t)}{f_0(x')} \quad (12.10)$$

and the textbook inequality $\ln(a/a') \geq 1 - (a/a')^{-1}$.

Inequalities (12.5) and (12.9) prove that the difference in nonequilibrium free energies A_F is a Lyapunov functional. The process of evolution leads towards the stationary state which, according to this criterion, is stable.

The same result was interpreted in Ref. [11] as an increase in entropy. In this connection we would like to observe the following:

The difference in the free energies and the corresponding Lyapunov functional can serve as the measure of how far the current state is from equilibrium. This information, however, is not sufficient for concluding whether or not the evolution under consideration is a process of self-organisation. To answer this question, we must introduce another Lyapunov functional in place of A_F ,

$$A_S = S_0 - \tilde{S}(t) = \int \ln \frac{\tilde{f}(x, t)}{f_0(x)} \tilde{f}(x, t) dx \geq 0, \quad \frac{dA_S}{dt} \leq 0 , \quad (12.11)$$

where $\tilde{f}(x, t)$ is the renormalised distribution. Renormalisation is based on the assumption that the mean effective Hamilton function H_{eff} remains constant in the course of evolution. The Lyapunov functional A_S can be used as the measure of the relative order of states in the course of evolution towards equilibrium. Inequalities (12.11) express Boltzmann’s H-theorem for processes described by the master equation (5.9).

12.2 The Fokker–Planck equation. The H-theorem

Now we turn to the Fokker–Planck equation (6.9), which follows either from the master equation (5.9) or from the equivalent equation (6.2). The stationary solution is then given by Eqn (6.16) with $v = 0$, which can be represented in the form [cf. Eqn (12.1)]

$$f_0(x) = \exp[F_{\text{eff}} - H_{\text{eff}}(x)], \quad H_{\text{eff}}(x) = \int_0^x \frac{A(x')}{D(x')} dx' . \quad (12.12)$$

Once again we introduce the Lyapunov functional A_F by formula (12.5). Its time derivative is found with the aid of Eqn (6.9),

$$\frac{dA_F}{dt} = - \int D(x) f(x, t) \left[\frac{\partial}{\partial x} \ln \frac{f(x, t)}{f_0(x)} \right]^2 dx \leq 0 . \quad (12.13)$$

We see that in case of the Fokker–Planck equation the free energy decreases monotonically in the course of time evolution towards the equilibrium distribution (12.12). Naturally, the Lyapunov functional A_F in this case also shows how far the system is from equilibrium, but is not an indicator of self-organisation. In order to obtain a criterion of self- organisation, one must again carry out renormalisation to the specified value of mean effective energy.

12.3 The Einstein–Smoluchowski equation. H-theorem

We return to the Einstein–Smoluchowski equation (10.17). The nonlinear thermostat acts on a Brownian particle through the effective force. The stationary solution in the thermostat coincides with the equilibrium solution and is given by Eqn (10.14). When the coefficient of feedback a_f is

zero, the solution is the Boltzmann distribution for a harmonic oscillator.

The Lyapunov functional A_F is defined by an expression similar to Eqn (12.5):

$$A_F = F(t) - F_0 = \int \ln \frac{f(\mathbf{r}, t)}{f_0(\mathbf{r})} f(\mathbf{r}, t) d\mathbf{r} \geq 0. \quad (12.14)$$

The time derivative is found with the aid of Eqn (10.17),

$$\frac{dA_F}{dt} = - \int D(\mathbf{r}) f(\mathbf{r}, t) \left[\frac{\partial}{\partial \mathbf{r}} \ln \frac{f(\mathbf{r}, t)}{f_0(\mathbf{r})} \right]^2 d\mathbf{r} \leq 0. \quad (12.15)$$

The Lyapunov functional A_F again indicates how far the system is from equilibrium. The criterion of self-organisation must be based on a functional defined in terms of the entropy difference.

So, we have demonstrated the feasibility of extending the thermodynamic concept of free energy to a large class of equations which describe Brownian motion in nonlinear media. Notwithstanding a certain artificiality of such a definition, Lyapunov functionals A_F are very helpful in dealing with many problems, some of which will be discussed in the sections to follow.

13. Brownian motion in self-oscillatory systems. The Van der Pol oscillator

In the preceding sections we discussed methods used for describing nonlinear Brownian motion. Now we are going to illustrate the efficiency of the general theory with concrete examples.

We have selected only those examples of nonlinear Brownian motion which are of interest from the standpoint of the general theory (this, of course, does not diminish their own importance). Many of these examples illustrate the difference between traditional and non-traditional approaches to the description of Brownian motion.

Let us begin with the Brownian motion in the Van der Pol oscillator.

The Van der Pol oscillator is a classic example of an electrical self-oscillatory system. It contains a linear electrical oscillatory circuit to which amplified feedback is applied.

Neglecting the fluctuations, the process in the oscillator can be described by a set of nonlinear dynamic dissipative equations in charge and current. The 'electron structure' of the flow of electric charge is taken into account by inclusion of the source of random emf \mathcal{E} into the dynamic equations (the Langevin source).

Consider an oscillator with soft excitation, when the linear component of the coefficient of friction changes its sign when the coefficient of feedback becomes sufficiently large. By electromechanical analogy, we obtain the following set of Langevin equations:

$$\frac{dx}{dt} = v, \quad \frac{dv}{dt} + (-\alpha + \beta v^2)v + \omega_0^2 x + \frac{1}{2} \frac{dD}{dv} = \sqrt{D(v)} y(t), \quad (13.1)$$

where $\alpha = \alpha_f - \gamma$, α_f is the coefficient of feedback, γ and β are the coefficients of linear and nonlinear friction.

Equations (13.1) are similar to the Langevin equations (3.16) for a Brownian particle in the medium with nonlinear friction. This time, however, the steady component of

friction may change its sign. Following the guidelines of Section 3, we go over from Eqns (13.1) to the corresponding Fokker-Planck equation in the kinetic form,

$$\frac{\partial f}{\partial t} + v \frac{\partial f}{\partial r} - \omega_0^2 r \frac{\partial f}{\partial v} = \frac{\partial}{\partial v} \left[D(v) \frac{\partial f}{\partial v} \right] + \frac{\partial}{\partial v} \left[(-\alpha + \beta v^2) v f \right], \quad (13.2)$$

which is similar to Eqn (3.15).

Exact solution of the equations for self-oscillations cannot be obtained even in the dynamic regime described by equations (13.1) without the Langevin source. It is far more difficult to solve the corresponding Langevin and Fokker-Planck equations. The equations can be considerably simplified, however, when the dissipative parameters are much smaller than the frequency of oscillations,

$$|\alpha|, \alpha_f, \delta \langle v^2 \rangle \ll \omega_0. \quad (13.3)$$

To put it differently, this means that all relaxation times are large compared with the period of oscillations. Then the equations under consideration can be made much simpler by carrying out averaging over the oscillation period $2\pi/\omega_0$. Mathematical aspects of the perturbation theory have been worked out by A A Andronov, N M Krylov, and N N Bogolyubov.

There are many papers that discuss the theory of fluctuations in oscillators (see, for example, [5-9, 20-22]). As a rule, the fluctuations are calculated under the assumption that the coefficient of diffusion is constant. This constraint is too strong when the feedback is designed in such a way that only thermal fluctuations remain in the circuit when the feedback is switched off.

Assume that the feedback is designed in exactly this way. Then the coefficient of nonlinear diffusion $D(v)$ at $\alpha_f = 0$ can be determined as explained in Section 3. In the absence of feedback the equilibrium solution is the Maxwell-Boltzmann distribution

$$f(x, v, \alpha_f = 0) = C \exp \left[-\frac{H(x, v)}{kT} \right],$$

$$H(x, v) = \frac{Mv^2}{2} + \frac{M\omega_0^2}{2} x^2, \quad (13.4)$$

where we have introduced the notation for the Hamilton function of a linear oscillatory circuit. By electromechanical analogy, the mass corresponds to the inductance L , the coordinate x to the charge, etc.

To define $D(v)$, we substitute this distribution into Eqn (13.2) and find that

$$D(v) = (\gamma + \beta v^2) \frac{kT}{M} = \gamma(v) \frac{kT}{M}. \quad (13.5)$$

We have once again come to the Einstein relation (3.4).

Assume now that the coefficient of feedback is nonzero. Then, putting the 'collision integral' in Eqn (13.2) equal to zero, we obtain the corresponding stationary solution, which is again represented as the canonical Gibbs distribution:

$$f(x, v, \alpha_f) = \exp \left(-\frac{H_{\text{eff}}}{kT} \right),$$

$$H_{\text{eff}} = H(x, v) - \frac{\alpha_f}{\beta} \ln \left(1 + \frac{\beta}{\gamma} \frac{mv^2}{2} \right). \quad (13.6)$$

If $\alpha_f = 0$, the effective Hamilton function coincides with Eqn (13.4).

Observe that this stationary solution does not, however, satisfy Eqn (13.2), since it does not bring the left-hand side to zero. This is yet another indication of the difficulty of definition of the stationary distribution for the non-averaged Fokker–Planck equation, which can only be done in an approximate way.

As mentioned above, one simplification is possible when condition (13.3) is satisfied. In the first approximation it reduces to averaging over the period of oscillations. Let us consider other possibilities which are based on the use of the simpler model equations for description of Brownian motion in the Van der Pol oscillator.

One such model will be considered in the next section. At this point we shall go to a coarser description by redefining the velocity of a Brownian particle in a non-equilibrium state at $\alpha_f \neq 0$:

$$v \rightarrow v_{\text{eff}} = \frac{\partial H_{\text{eff}}}{\partial Mv} = v - \alpha_f \frac{1}{\gamma + \beta v^2} v. \quad (13.7)$$

Then in place of Eqn (13.2) we come to the following equation:

$$\begin{aligned} \frac{\partial f}{\partial t} + \frac{\partial H_{\text{eff}}}{\partial mv} \frac{\partial f}{\partial r} - \omega_0^2 r \frac{\partial f}{\partial v} \\ = \frac{\partial}{\partial v} \left[D(v) \frac{\partial f}{\partial v} \right] + \frac{\partial}{\partial v} \left[(-\alpha + \beta v^2) v f \right], \end{aligned} \quad (13.8)$$

whose stationary solution has the form of Eqn (13.6).

Owing to the new definition of the velocity, Eqn (13.7), the level of description of Brownian motion in the oscillator is changed. The appearance of a limiting cycle is now not described in an explicit way; it is reflected, however, in the distribution (13.6). When the parameter of feedback is large enough, the distribution with respect to velocity becomes bistable, and the maxima of the distribution are linked with the energy of oscillations in the generation regime.

This simplification is also efficient in the more general case when both dissipative and nondissipative nonlinearities are present simultaneously. Recall that the case of non-dissipative nonlinearity has been considered in Section 10 in connection with the description of Brownian motion in bistable elements. Combination of the two types of nonlinearities gives rise to many new regimes of Brownian motion, as such systems may host both equilibrium and nonequilibrium ‘phase transitions’.

In particular the equilibrium phase transitions may affect considerably the rates of chemical reactions. In this case one may speak of the new kind of chemical catalysis, when the rate of chemical reaction is controlled by the value of the coefficient of feedback α_f .

Now we return to the solution (13.6) and apply the criterion of the S-theorem [19, 39, 40] for calculating the relative degree of order of different regimes of generation which correspond to different values of α_f .

For the ‘reference point’ it would be natural to take the state of equilibrium with the distribution (13.4), which corresponds to $\alpha_f > 0$. Equation

$$\begin{aligned} \frac{3}{2} k \tilde{T}(\alpha_f) &\equiv \int \frac{Mv^2}{2} C_0 \exp \left[-\frac{H(x, v)}{k \tilde{T}(\alpha_f)} \right] dx dv \\ &= \int \frac{Mv^2}{2} C \exp \left[-\frac{H_{\text{eff}}(x, v)}{kT} \right] dx dv, \end{aligned} \quad (13.9)$$

is used for renormalising the equilibrium state to the preassigned value of the mean kinetic energy of oscillations in the state with $\alpha_f > 0$. This equation allows us to find the effective temperature as a function of α_f ,

$$\tilde{T} = \tilde{T}(\alpha_f),$$

at ‘initial condition’

$$\tilde{T}(\alpha_f)|_{\alpha_f=0} = T. \quad (13.10)$$

Since the maximum entropy corresponds to the state of equilibrium, given that the values of mean energy are the same, the degree of order for all states with $\alpha_f > 0$ is higher than that for the equilibrium state with $\alpha_f = 0$. Equation (13.9) also allows one to check whether the degree of order increases monotonically with increasing α_f .

Quantitative assessment of the relative degree of order of different states is based on the difference in the entropies of the renormalised equilibrium state and the state with $\alpha_f > 0$. We shall discuss this issue in Section 18.

14. The Van der Pol oscillator. Symmetrised nonlinearity

In some cases (for instance, in the theory of solid state lasers) the process of generation is described by equations in x and v which are symmetrical with respect to the dissipative nonlinearity [17, 21]:

$$\frac{dx}{dt} + \frac{1}{2}(-\alpha + \beta E)x = v, \quad \frac{dv}{dt} + \frac{1}{2}(-\alpha + \beta E)v + \omega_0^2 x = 0, \quad (14.1)$$

where

$$E = \frac{M}{2}(v^2 + \omega_0^2 x^2) \equiv H(x, v), \quad (14.2)$$

is the energy of oscillations.

An exact equation in E (without averaging over the period of oscillations) follows from Eqn (14.1):

$$\frac{dE}{dt} = (\alpha - \beta E)E, \quad \alpha = \alpha_f - \gamma. \quad (14.3)$$

The solution of these equations can be expressed in terms of fixed values of energy E_0 and phase ϕ_0 (we assume that $M = 1$):

$$x(t) = \omega_0^{-1} \sqrt{2E(t)} \cos(\omega_0 t + \phi_0), \quad (14.4)$$

$$v(t) = -\sqrt{2E(t)} \sin(\omega_0 t + \phi_0),$$

$$E(t) = E_0 \frac{\alpha}{\beta} \left[E_0 - \left(E_0 - \frac{\alpha}{\beta} \right) e^{-\alpha t} \right]^{-1},$$

$$E(t) = \frac{E_0}{1 + E_0 \beta t} \quad \text{at} \quad \alpha = 0. \quad (14.5)$$

The character of time evolution depends on the sign of α . At $\alpha < 0$ the system is at rest with $E = 0$; at $\alpha > 0$ steady oscillations are established with frequency ω and energy of the limiting cycle $E = \alpha/\beta$. Thus, the value of $\alpha = 0$ corresponds to the bifurcation point.

The same expression also defines the relaxation time as a function of the deviation from the bifurcation point. At a finite distance from the point of bifurcation, the system approaches either of the stationary states according to the exponential law with the relaxation time $\tau_{\text{rel}} \sim |\alpha|^{-1}$.

Closer to the bifurcation point the exponential dependence is replaced by the power law. In the limit of $\alpha = 0$, $E(t) \propto 1/t$ and the dependence on E_0 disappears.

Fluctuations are taken into consideration by introducing Langevin sources into these equations, and establishing the appropriate form of the Fokker–Planck equation. As before, we give preference to the K-form of the Fokker–Planck equation.

We shall use the same equations as those developed for describing Brownian motion with nonlinear friction. By analogy with Eqn (13.1), we write the Langevin equation for the energy as

$$\frac{dE}{dt} + (-\alpha + \beta E)E - \frac{1}{2} \frac{d}{dE} [D(E)E] = \sqrt{D(E)E} y(t). \quad (14.6)$$

The moments of the Langevin source $y(t)$ are given by

$$\langle y(t) \rangle = 0, \quad \langle y(t)y(t') \rangle = 2\delta(t - t'). \quad (14.7)$$

The only difference is that in the definition of the coefficient of diffusion we factor out the quantity E , which comes up in the transition from x, v to polar coordinates and also remains there in the case of constant diffusion.

Transition to the equation for the distribution function $f(E, t)$ follows the guidelines set forth in Section 3, and results in

$$\frac{\partial f(E, t)}{\partial t} = \frac{\partial}{\partial E} [D(E)E \frac{\partial f}{\partial E}] + \frac{\partial}{\partial E} [(-\alpha + \beta E)E f]. \quad (14.8)$$

We can also write the equation for the mean energy,

$$\frac{d\langle E \rangle}{dt} + \langle (-\alpha + \beta E)E \rangle = \left\langle \frac{dD(E)E}{dE} \right\rangle, \quad (14.9)$$

which is obviously not closed.

Function $D(E)$ in Eqn (14.8) is defined from the condition that in the absence of feedback the solution should be the Boltzmann distribution. Hence

$$D(E) = D \left(1 + \frac{\beta}{\gamma} E \right), \quad D = \gamma kT. \quad (14.10)$$

The stationary solution of Eqn (14.8) is again represented as the canonical Gibbs distribution with the effective Hamilton function

$$f_0(E) = C \exp \left[-\frac{H_{\text{eff}}(E, \alpha_f)}{kT} \right],$$

$$H_{\text{eff}} = E - \frac{\alpha_f}{\beta} \ln \left(1 + \frac{\beta E}{\gamma} \right). \quad (14.11)$$

At α_f this solution coincides with the Boltzmann distribution.

Diffusion $D(E)$ reflects the ‘atomicity’ of charge transfer. Often employed is the approximation of predetermined noise: $D(E) = D$. In this approximation the Einstein relation $D = \gamma kT$ does not hold, and so the stationary solution is

$$f(E, a) = \exp \frac{F - H_{\text{eff}}}{D/\gamma}, \quad H_{\text{eff}}(E) = -\alpha E + \frac{1}{2} - \beta E^2, \quad (14.12)$$

where F and D/γ are, respectively, the effective free energy and the effective temperature.

Let us compare the stationary distributions corresponding to the different definitions of the Langevin source (and hence to the different definitions of the coefficient of diffusion).

When the coefficient of diffusion is a function of energy, the distribution function falls off more slowly at high energies. The ‘tails’ of distributions are therefore different. At the same time, the locations of maxima coincide:

$$E_{\text{max}} = \frac{\alpha}{\beta} \quad \text{if } \alpha > 0,$$

$$E_{\text{max}} = 0 \quad \text{if } \alpha < 0. \quad (14.13)$$

It is interesting to compare the two distributions in the domain of the Gaussian approximation. For the region of well-developed generation we may rewrite Eqn (14.12) as

$$f_{(3)} = \sqrt{\frac{\beta}{2\pi D}} \exp \left[-\frac{(E - \alpha/\beta)^2}{2D/\beta} \right], \quad \varepsilon = \frac{D\beta}{\alpha^2} \ll 1, \quad (14.14)$$

where ε is a dimensionless parameter which is small in the regime of well-developed generation. Then the expression for the relative dispersion of energy is

$$\langle (\delta E)^2 \rangle \langle E \rangle^{-2} = D\beta\alpha^{-2} = \varepsilon \ll 1. \quad (14.15)$$

To bring Eqn (14.11) to the form of the Gaussian distribution, we expand the exponent in $E - E_{\text{max}}$ and retain the first two derivatives. As a result, we arrive at the distribution similar to that given by (14.14); the relative dispersion, however, is now given by

$$\langle (\delta E)^2 \rangle \langle E \rangle^{-2} = D\beta\alpha^{-2} \left(\frac{\gamma + \alpha}{\gamma} \right) \equiv \varepsilon \frac{\gamma + \alpha}{\gamma} \ll 1. \quad (14.16)$$

Recall that for natural fluctuations we have $D = \gamma kT$.

We see that even in the Gaussian approximation the energy-dependent coefficient of diffusion alters the expression for the relative dispersion. Expression (14.16) only coincides with Eqn (14.15) when $\alpha \ll \gamma$. Accordingly, the applicability of expressions (14.14) and (14.15) is restricted by two inequalities:

$$\varepsilon = D\beta\alpha^{-2} \ll 1, \quad \alpha \ll \gamma. \quad (14.17)$$

This means that we are in the domain of well-developed generation, but not far from the threshold.

A similar two-way inequality defines the limits of applicability of Landau’s theory of phase transitions [41, 42]. This theory only works under conditions similar to those given by inequalities (14.17). Later on we shall return to this problem.

15. Combined action of natural and external noise

Of special practical interest is the case when the system experiences the action of both natural noise [via the function $D(E)$] and external noise. The following example shows that it is necessary to take the natural noise into account even when the external noise is strong [18].

We return to the Langevin equation (14.6) and assume that the value of the coefficient of feedback fluctuates under the action of an external source,

$$\alpha_f \rightarrow \alpha_f + \sqrt{\sigma} y(t), \quad (15.1)$$

where σ is the intensity of parametric noise. The moments

of random function $y(t)$ are given by Eqn (14.7). Then the Langevin equation becomes

$$\begin{aligned} \frac{dE}{dt} + (-\alpha + \beta E)E - \frac{1}{2} \frac{d}{dE} \left[\sqrt{D(E)E} + \sqrt{(\sigma)E} \right]^2 \\ = \left[\sqrt{D(E)E} + \sqrt{(\sigma)E} \right] y(t). \end{aligned} \quad (15.2)$$

Following the scheme of Section 3, we find the corresponding Fokker–Planck equation:

$$\begin{aligned} \frac{\partial f(E, t)}{\partial t} = \frac{\partial}{\partial E} \left\{ \left[\sqrt{D(E)E} + \sqrt{\sigma E} \right]^2 \frac{\partial f}{\partial E} \right\} \\ + \frac{\partial}{\partial E} \left[(-\alpha + \beta E)E f \right]. \end{aligned} \quad (15.3)$$

The stationary solution of this equation is

$$\begin{aligned} f(E) = C \exp \left(\int_0^E \frac{\alpha - \beta E'}{\left[\sqrt{D(E')E'} + \sqrt{\sigma E'} \right]^2} dE' \right), \\ D(E) = D \left(1 + \frac{\beta}{\gamma} E \right), \end{aligned} \quad (15.4)$$

and coincides with Eqn (14.12) if external noise is absent ($\sigma = 0$). If, on the contrary, the source of natural fluctuations is switched off ($D = 0$), the solution (15.4) becomes

$$f(E) = C \exp \int_0^E \frac{\alpha - \beta E'}{\sigma E'} dE', \quad \int_0^\infty f(E) dE = 1. \quad (15.5)$$

We see that the integral in the exponent contains logarithmic divergence at small values of E . At the same time, in the general expression the denominator is finite even when $E = 0$ owing to the presence of natural noise, and the integral does not diverge. This proves that the inclusion of natural noise (even small) may dramatically alter the behaviour of the distribution and lead to a physically correct result. In this way, the known difficulty of calculation of fluctuations in an oscillator with fluctuating parameters is overcome.

16. The symmetrised oscillator. Distribution of coordinates and velocities

Let us now establish the more general Fokker–Planck equation for distribution function $f(x, v, t)$. For this purpose we introduce the appropriate Langevin sources into the dynamic equations (14.1). Recall that energy E is expressed in terms of x, v by the use of Eqn (14.2). Once again we use the method of Section 3, getting as a result the following Fokker–Planck equation:

$$\begin{aligned} \frac{\partial f(x, v, t)}{\partial t} + v \frac{\partial f}{\partial x} - \omega_0^2 x \frac{\partial f}{\partial x} \\ = \frac{1}{2} \left\{ \frac{\partial}{\partial v} \left[D(E) \frac{\partial f}{\partial v} \right] + \frac{1}{\omega_0^2} \frac{\partial}{\partial x} \left[D(E) \frac{\partial f}{\partial x} \right] \right\} \\ + \frac{1}{2} \left\{ \frac{\partial}{\partial v} \left[(-\alpha + \beta E) v f \right] + \frac{\partial}{\partial x} \left[(-\alpha + \beta E) x f \right] \right\}. \end{aligned} \quad (16.1)$$

To obtain from Eqn (16.1) our former Eqn (14.8) we must use the relationship between distribution functions $f(E, t)$ and $f(x, v, t)$.

The nonlinear coefficient of diffusion is obtained from the condition of existence of the Gibbs distribution for the oscillator when $\alpha_f = 0$; the resulting expression is similar to Eqn (14.11):

$$D(E) = D \left(1 + \frac{\beta}{\gamma} E \right), \quad E = \frac{1}{2} (v^2 + \omega_0^2 x^2). \quad (16.2)$$

The stationary solution of Eqn (16.1) is

$$\begin{aligned} f_0(x, v, \alpha_f) = C \exp \left[-\frac{H_{\text{eff}}(x, v)}{kT} \right], \\ H_{\text{eff}} = E - \frac{\alpha_f}{\beta} \ln \left(1 + \frac{\beta}{\gamma} E \right), \end{aligned} \quad (16.3)$$

and coincides with the canonical Gibbs distribution when $\alpha_f = 0$.

17. H-theorem for the Van der Pol oscillator

We represent the distribution (16.3) as

$$f_0(x, v, \alpha_f) = \exp \left[\frac{F_{\text{eff}} - H_{\text{eff}}(x, v, \alpha_f)}{kT} \right], \quad (17.1)$$

(this time we are dealing with the true temperature of the thermostat rather than with the effective temperature). Once again we introduce a functional defined as the difference between the free energy of the state at the time t and the free energy of the stationary state:

$$A_F(t) = F(t) - F_{\text{eff}} = kT \int \ln \frac{f(x, v, t)}{f_0(x, v, \alpha_f)} f(x, v, t) dx dv \geq 0. \quad (17.2)$$

With the aid of the Fokker–Planck equation (16.1) we obtain the derivative of this functional, and find it to be similar to Eqn (12.13). Hence, A_F is a Lyapunov functional.

Let us quote the results for the simpler equation (14.8), which will be required shortly for evaluation of the maximum permissible discrete time step in the numerical solution of the Fokker–Planck equation. The Lyapunov functional satisfies two inequalities:

$$A_F = F(t) - F(T, \alpha_f) = kT \int_0^\infty \ln \frac{f(E, t)}{f_0(E, \alpha_f)} f(E, t) \geq 0, \quad (17.3)$$

$$\frac{dA_F}{dt} = -kT \int_0^\infty D(E) E f \left[\frac{\partial}{\partial E} \ln \frac{f(E, t)}{f_0(E, \alpha_f)} \right]^2 dE \equiv -\sigma_F \leq 0, \quad (17.4)$$

where σ_F is the analogue of entropy production. The stationary solution for f_0 is given by Eqn (14.11).

In this way, the Lyapunov functional is defined as the difference in nonequilibrium free energies. As indicated elsewhere, the functional A_F can serve as a measure of remote-ness from the stationary state. However, this information is in general not sufficient for deciding whether the evolution in question is a process of self-organisation. This problem can only be solved with the aid of a functional based on the difference in the entropies of the stationary and the current nonequilibrium states.

In order to define the Lyapunov functional A_F in the case of evolution described in terms of the distribution

function $f(E, t)$, we must go over to the renormalised distribution $\tilde{f}(E, t)$. Renormalisation is carried out under the assumption that the mean effective Hamilton function (14.11) remains constant in the course of time evolution towards the stationary state. This condition may be expressed as

$$\int_0^\infty H_{\text{eff}}(E, \alpha_f) \tilde{f}(E, t) dE = \int_0^\infty H_{\text{eff}}(E, \alpha_f) f_0(E, \alpha_f) dE. \quad (17.5)$$

Naturally this condition is not satisfied in the course of time evolution towards the stationary state according to the Fokker–Planck equation (14.8). In order to comply with this condition, we must introduce the new temperature \tilde{T} which is a functional of the distribution $\tilde{f}(E, t)$:

$$\tilde{T}\{f\} = \int \frac{(-\alpha + \beta E)^2 E}{\gamma + \beta E} \tilde{f} dE \left\{ \int \frac{d}{dE} [(-\alpha + \beta E) E] \tilde{f} dE \right\}^{-1}. \quad (17.6)$$

The renormalised distribution satisfies the equation which is nonlinear in \tilde{f} :

$$\frac{\partial \tilde{f}}{\partial t} = \frac{\partial}{\partial E} \left[\tilde{D}(E) E \frac{\partial \tilde{f}}{\partial E} \right] + \frac{\partial}{\partial E} [(-\alpha + \beta E) E \tilde{f}], \quad \tilde{D}(E) = k\tilde{T}\{\tilde{f}\}(\gamma + \beta E). \quad (17.7)$$

The stationary solution of this equation coincides with Eqn (14.11), and the temperature \tilde{T} is the same as the thermostat temperature T .

After renormalisation we may introduce the functional defined by the difference in the entropies,

$$A_S = S_0 - \tilde{S}(t) = k \int_0^\infty \ln \frac{\tilde{f}(E, t)}{f_0(E, \alpha_f)} \tilde{f}(E, t) dE \geq 0. \quad (17.8)$$

Using Eqns (17.7), one can prove that this functional decreases monotonically in the course of time evolution towards the stationary state:

$$\frac{dA_S}{dt} = -k \int_0^\infty \tilde{D}(E) E f \left[\frac{\partial}{\partial E} \ln \frac{\tilde{f}(E, t)}{f_0(E, \alpha_f)} \right]^2 dE \equiv -\sigma \leq 0. \quad (17.9)$$

The last two inequalities indicate that $\#$ is a Lyapunov functional. They also prove that the stationary state is stable at any value of the feedback parameter.

In this way we have proved the H-theorem for Eqn (17.7), which is similar to Boltzmann's H-theorem for rarefied gas. There is, however, one question.

As a matter of fact, the Lyapunov functional A_S for the Boltzmann equation is a natural characteristic, since the mean energy is conserved in the course of evolution towards equilibrium. Currently, however, the situation is quite different: to make condition (17.5) hold, we had to change the structure of the initial equation and to replace Eqn (14.8) by Eqn (17.7). It follows that we have proved the H-theorem for a different system. Is it of any value for the original problem? Or should we put up with the results for the functional A_S ? Such questions are certainly not without reason. For the example under consideration, however, the Lyapunov functional A_S is actually relevant to the initial equation (14.1), since it includes, for instance, its stationary solution f_0 . Because of this, the analysis of the relative degree of order based on the H-theorem is useful. We shall illustrate this clearly in the next section with the example of the evolution of stationary states as the parameter of feedback is varied.

18. Self-organisation in the Van der Pol oscillator. The S-theorem

In Refs [17, 39, 40,] the criterion of the relative degree of order of the states of open systems was formulated as the S-theorem. Now we shall apply this criterion for the particular case of evolution of stationary states of the Van der Pol oscillator as the parameter of feedback (the controlling parameter) is varied.

So, let us return to the stationary solution (14.11) and write it out for three selected states:

1. Feedback parameter is zero, $\alpha_f = 0$. The stationary solution (14.11) is then the Boltzmann distribution:

$$f_{(1)} = \frac{1}{kT} \exp\left(-\frac{E}{kT}\right). \quad (18.1)$$

2. Generation threshold, $\alpha_f = \gamma$. We also assume that the nonlinearity is small. Then we can expand Eqn (14.11) in $kT\beta/\gamma$, and find that

$$f_{(2)} = \sqrt{\frac{2\beta}{\pi\gamma kT}} \exp\left(-\frac{\beta E^2}{2\gamma kT}\right). \quad (18.2)$$

3. Regime of well-developed generation. In this case we may use the Gaussian distribution

$$f_{(3)} = \sqrt{\frac{1}{2\pi \langle (\delta E)^2 \rangle}} \exp\left[-\frac{(E - \alpha\beta^{-1})^2}{2 \langle (\delta E)^2 \rangle}\right]. \quad (18.3)$$

The relative dispersion of energy is given by Eqn (14.16). From the above distributions we find the mean energy values,

$$\langle E \rangle_{(1)} = kT, \quad \langle E \rangle_{(2)} = \sqrt{\frac{2}{\pi}} \frac{\gamma kT}{\beta}, \quad \langle E \rangle_{(3)} = \frac{\alpha}{\beta}, \quad (18.4)$$

and the corresponding entropy values (at #):

$$S_{(1)} = \ln kT + 1, \quad S_{(2)} = \ln \left(\frac{\pi\gamma kT}{2\beta} \right)^{1/2} + \frac{1}{2}, \quad S_{(3)} = \ln \sqrt{\frac{2\pi kT}{\beta}} (\gamma + \alpha) + \frac{1}{2}. \quad (18.5)$$

Now let us analyse the implications of these results.

First of all, we note that, under our assumption that $\beta kT/\gamma \ll 1$, the entropy increases as the system moves towards the regime of well-developed generation:

$$S_{(1)} < S_{(2)} < S_{(3)}. \quad (18.6)$$

This could be interpreted as a decrease in the orderliness of states as the generation develops. Intuitively it is clear, however, that the degree of order should increase. What is wrong?

To clear up the situation we shall compare the values of mean energy for the three selected states. From Eqn (18.4) it follows that

$$\langle E \rangle_{(1)} < \langle E \rangle_{(2)} < \langle E \rangle_{(3)}. \quad (18.7)$$

We see that the mean energy also increases as the generation develops. At the same time, we know that the S-theorem must be applied to the states whose mean energy is the same. Accordingly, we have to use an appropriate procedure of renormalisation.

In our current example it would be natural to choose state '1' (the state of equilibrium) for the state of 'physical

chaos'. Since the distribution function is then given by Eqn (18.1), the effective Hamilton function coincides with the energy E :

$$H_{\text{eff}}(E, \alpha_f) = E . \tag{18.8}$$

In this way the renormalisation is carried out at a preassigned value of the mean energy.

In this connection we recall Eqn (13.9), used as the additional condition for renormalizing the velocity distribution so as to comply with the criterion of the S-theorem. Now, taking into account Eqn (18.8), this equation takes on the form

$$k\tilde{T}(\alpha_f) = \int_0^\infty E\tilde{f}_0(E, \alpha_f = 0) dE = \int_0^\infty Ef(E, \alpha_f) dE , \tag{18.9}$$

and allows us to find the effective temperature as a function of the controlling parameter α_f :

$$\tilde{T} = \tilde{T}(\alpha_f) ,$$

at 'initial condition'

$$\tilde{T}(\alpha_f)|_{\alpha_f=0} = T . \tag{18.10}$$

Taking advantage of this result, we now compare state '1' with state '2' at $\langle E \rangle_{(1)} = \langle E \rangle_{(2)}$, and then state '1' with state '3' at $\langle E \rangle_{(1)} = \langle E \rangle_{(3)}$. The renormalised values of temperature and difference in entropies are given by

$$k\tilde{T}_{(1)} = \left(\frac{2}{\pi} \frac{\gamma kT}{\beta}\right)^{1/2} > kT , \quad \tilde{S}_{(1)} - S_{(2)} = \ln \frac{2}{\pi} + \frac{1}{2} > 0 , \tag{18.11}$$

for the first pair of states, and by

$$k\tilde{T}_{(1)} = \frac{\alpha}{\beta} > \sqrt{\frac{2}{\pi} \frac{\gamma kT}{\beta}} > kT , \tag{18.12}$$

$$\tilde{S}_{(1)} - S_{(3)} = \ln \sqrt{\frac{1}{2\pi\varepsilon} \frac{\gamma + \alpha}{\gamma}} > \ln \frac{2}{\pi} + \frac{1}{2} > 0 ,$$

$$\varepsilon = \frac{\beta\gamma kT}{\alpha^2} \ll 1 , \tag{18.13}$$

for the second pair. Recall that ε is a characteristic small parameter for the regime of well-developed generation.

From these results for the three selected states it follows that the effective temperature given by Eqn (18.9) steadily grows, and the entropy steadily decreases as the feedback parameter α_f is increased. Thus, the disorder added in state '1' by 'heating' it to the temperature \tilde{T} , transforms in states '2' and '3' into the more ordered motion, as the entropy of these states is less, given that their mean energy is the same. This allows us to conclude that the evolution towards well-developed generation is a process of self-organisation. What is more, these results confirm that our choice of the feedback parameter # for the controlling parameter is correct.

Note that the S-theorem as a criterion of the relative degree of order was first formulated for the case of the Van der Pol oscillator [39].

18.1 Shannon entropy and 'S-information'

Let us return to expressions (18.5) which define the values of Shannon entropy (or Shannon information)

$$S \equiv I = - \int \ln f(E)f(E) dE , \tag{18.14}$$

for the three selected stationary states of the Van der Pol oscillator. To draw a comparison with the above results based on the criterion of the S-theorem, let us compare the values of Shannon entropy for states '3' and '2':

$$S_{(3)} - S_{(2)} \equiv I_{(3)} - I_{(2)} = \ln \left(2 \frac{\gamma + \alpha}{\gamma}\right) > 0 , \tag{18.15}$$

which implies that the entropy and the information increase as the generation develops. If this result is to be interpreted as the increase in chaoticity, we challenge both our physical intuition and the criterion of the S-theorem.

It is possible, however, to construe this result, in the spirit of information theory, as the information gain: the information about the system increases upon transition to the regime of well-developed generation. The state of well-developed generation is thus regarded as more informative. It is difficult, however, to give a comprehensible physical explanation to this result.

19. Oscillator with inertial nonlinearity

Dynamic motion in the Van der Pol oscillator is two-dimensional, being described by a set of two ordinary differential equations of the first order (13.1) without a Langevin source. In such systems only the simple attractors are possible: the state of rest and the limiting cycle.

Three-dimensional systems may also have the so-called *strange attractors*. This term refers to such regions of phase space where all trajectories are dynamically unstable, and the divergence of trajectories is exponential. Because of this, the Kolmogorov—Krylov—Sinai entropy (K-entropy) is positive.

Quite understandable is the desire to explore the more complicated oscillators, in which the dynamic processes are described by at least three differential equations of the first order. Such systems include, in particular, the so-called oscillators with inertial nonlinearity, first described by K F Teodorichik [23, 43].

The oscillatory circuit of Teodorichik's oscillator includes a thermistor, which gives rise to inertial nonlinearity and, as a consequence, to new regimes of oscillations. This oscillator, however, does not involve strange attractors. In order to produce complex motion, a modified oscillator with inertial nonlinearity was invented [44], in which the inertial converter is placed in the feedback circuit. The nature of motion in the oscillator depends considerably on the asymmetry of the nonlinear characteristic of the inertial element. The most 'universal' spectrum of bifurcations is observed in such an oscillator when a half-wave detector is used.

As a matter of fact, the strange attractor was discovered in the classic paper of E Lorenz in 1963 [45]. As a mathematical object, however, it was first analysed by D Ruelle and F Takens in 1971 [46].

In connection with the discovery of the strange attractor it would be interesting to mark the following. 'Strange' irregular solutions of nonlinear equations of the same type as Lorenz equations were independently discovered by A Grasyuk and A Oraevskii in 1964 [47] in their studies of oscillations in a molecular oscillator. At that time their discovery failed to draw much attention, since complex irregular motions in molecular generators did not seem to be too important. By contrast, for Lorenz the appearance of complex chaotic regimes explained why a long-term weather

forecast was practically impossible. The present-day status of the problem of predictability of complex behaviour is reported in Ref. [48].

The Anishchenko–Astakhov oscillator with inertial nonlinearity proved to be a very convenient device for experimental study of complex ('chaotic') behaviour in relatively simple dynamic systems. In particular, it was observed that as the feedback parameter increases (with fixed value of the inertial parameter), the limiting cycle is surpassed by a sequence of period-doubling bifurcations in accordance with Feigenbaum's scheme [49]. Beyond Feigenbaum's critical point, the strange attractor appears with intricately interlaced regions of chaos and order. What is more, there was good agreement between the results of physical and numerical simulations. In recent years, the fluctuation processes ('Brownian motion') in such oscillators, due to both natural and external noise, have been thoroughly investigated.

Evaluation of the relative degree of order was based on the criterion of the S-theorem [50]; the entropy was calculated by the scheme described above. Renormalisation of the distribution function was carried out at the given intensity of oscillations [conditions similar to Eqn (18.9)]; the range of the feedback parameter extended up to Feigenbaum's critical point.

Calculations indicate that the entropy (renormalised to the preassigned value of the intensity of oscillations) decreases as the system approaches the critical point of transition into the domain of the strange attractor, in the process of a period-doubling sequence. This points to the increasing degree of order. In other words, the sequence of period-doubling bifurcations may be regarded as a process of self-organisation. More detailed analysis shows that the decrease in entropy is nonmonotonic in the neighbourhood of bifurcation points.

For our future discussion it is important that the sequence of period-doubling bifurcations and the main features of bifurcations in the domain of strange attractor can be described on the basis of the so-called *logistic equation*

$$x_{n+1} = \alpha(1 - x_n)x_n, \quad 0 \leq \alpha \leq 4, \quad 0 \leq x \leq 1, \quad (19.1)$$

which describes a one-dimensional process in discrete time with a unit step. It is also referred to as the *equation of sequence*, because it may describe a sequence of locations of traces of trajectory (of the process in question) on the secant plane.

In this way, the one-dimensional equation in discrete time emulates the important properties of the system of three differential equations. This becomes possible because the dimensionality of phase space filled up by the trajectory is two with a small fraction.

Logistic equations were first introduced for describing the behaviour of biological objects. We shall see that equations of this kind offer the possibility of construction of two- and one-dimensional models of complex motion. This complex motion owes its existence not to the escape of trajectory from the plane into three dimensions, as in case of the oscillator with inertial nonlinearity, but to the more sophisticated nonlinearity.

But let us return to the logistic equation. If α varies within the range of $3 \leq \alpha \leq 4$, the logistic equation describes a very complicated motion which corresponds

to the state of the so-called *dynamic chaos*. This state may be characterised by appropriate distribution functions. The most chaotic state occurs when $\alpha = 4$. In general, the distribution function $f(x, \alpha)$ can only be determined from numerical simulations.

For the most chaotic state ($\alpha = 4$) the distribution function was found analytically by Ulam and Neumann:

$$f(x) = \frac{1}{\pi} \sqrt{x(1-x)}, \quad \int_0^1 f(x) dx = 1. \quad (19.2)$$

This distribution can be obtained in the following way [17, 51, 52.]: it is possible to use equation Eqn (19.1) for simulating complex motion as described, for instance, by the dynamic equations of the oscillator with inertial nonlinearity. Naturally, one has to deal with the problem of the description of Brownian motion in such systems. As before, it is important to take into account not only the natural sources of noise, which reflect the 'atomic structure' of individual elements of the oscillator, but also the external noises, which can be used for controlling the sophisticated process of generation.

Problems of this kind are very complicated because, as we have seen, the characteristics of natural noise in nonlinear systems depend on the nonlinear properties of the system. For this reason it will be worthwhile to start by tackling a simpler problem. Namely we shall discuss the structure of the Langevin equation which coincides with the logistic equation (19.1) if the noise is neglected.

We shall see that this approach offers a new opportunity for constructing a hierarchy of mathematical models of oscillators displaying sequences of bifurcations of the energy of the limiting cycle. It is with this problem that we shall start our investigation of stochastic processes in systems with complex behaviour.

20. Bifurcations of the energy of the limiting cycle. Oscillators with multistable stationary states

As indicated above, the transition to 'dynamic chaos' in an oscillator with inertial nonlinearity can be qualitatively described on the basis of the logistic equation.

Now we are going to show that the same equation can be used for constructing a model of the generalised Van der Pol oscillator, in which a cascade of bifurcations of limiting cycle energy is possible as the parameter of feedback increases.

We start by introducing dimensionless variables. The choice of scales is, of course, not unambiguous. A natural time interval for an oscillator is its period of oscillations $T = 2\pi/\omega_0$. Accordingly, the dimensionless variables can be defined as follows:

$$\begin{aligned} t' &= \omega_0 t, & \alpha' &= \alpha \omega_0^{-1}, & E' &= \beta E \omega_0^{-1}, \\ D' &= \beta D \omega_0^{-2} = \gamma' k T'. \end{aligned} \quad (20.1a)$$

In the second case, the time scale is based on the relaxation time $1/\gamma$:

$$\begin{aligned} t' &= \gamma t, & \alpha' &= \alpha \gamma^{-1}, & E' &= \beta E \gamma^{-1}, \\ D' &= D \beta \gamma^{-2} = k T \beta \gamma^{-1} = k T'. \end{aligned} \quad (20.1b)$$

The form of Eqn (14.3) for the energy E' does not depend

explicitly on the choice of scales used for transition to dimensionless variables (further on, the prime is dropped):

$$\frac{dE}{dt} = (\alpha - E)E. \tag{20.2}$$

The distinction becomes important, however, when the differential equation is replaced by the equation in differences. The basic time scale will then define the discrete time step; the choice of time scale also affects the calculation of fluctuations.

Let us generalise Eqn (20.2) in such a way as to obtain a sequence of bifurcations of limiting cycle energy with an increasing feedback parameter. With this purpose we replace Eqn (20.2) by an equation in discrete time with step $\Delta = 1$, which corresponds to the period of oscillations in case of dimensionless variables (20.1a), and to the relaxation time $1/\gamma$ in the case of dimensionless variables (20.1b). As a result, we come to the logistic equation

$$E_{n+1} = (\alpha + 1)E_n - E_n^2 \equiv F(E_n), \quad 0 \leq \alpha + 1 \leq 4, \\ 0 \leq E \leq 4, \tag{20.3}$$

which after the k th iteration becomes

$$E_{n+k} = F^{(k)}(E_n). \tag{20.4}$$

To return from Eqn (20.4) to the differential equation, we define the sequence of derivatives

$$\frac{E_{n+k} - E_n}{k} \longleftrightarrow \frac{dE}{dt}, \quad k = 1, 2, \dots \tag{20.5}$$

As a result, we come to the sequence of differential equations

$$\frac{dE}{dt} = \frac{1}{k} [F^{(k)}(E) - E], \quad k = 1, 2, \dots \tag{20.6}$$

Let us consider a few first equations in this sequence. At $k = 1$ we return to the initial equation (20.2). At $k = 2$ we get a new equation

$$\frac{dE}{dt} = \frac{1}{2}(\alpha - E)E[E^2 - (\alpha + 2)E + \alpha + 2]. \tag{20.7}$$

This equation admits four stationary states:

$$E_1 = 0 \text{ at } \alpha \leq 0; \quad E_2 = \alpha \text{ at } 0 \leq \alpha \leq 2, \\ E_{3,4} = \frac{\alpha + 2}{2} \pm \sqrt{\left(\frac{\alpha + 2}{2}\right)^2 - (\alpha + 2)} \text{ at } \alpha \geq 2. \tag{20.8}$$

We see that ramification of values of the limiting cycle energy occurs at $\alpha = 2$, and we are dealing with a bistable state. In general, the number of stationary states in the range of values of α up to Feigenbaum's critical point is 2^k . The possible values of the limiting cycle energy coincide with the values of energy at the stationary points of the logistic equation (20.3).

Equations (14.1) for functions $x(t)$ and $v(t)$ are generalised in a similar way, by making the substitution

$$\alpha - E \rightarrow \frac{1}{k} \frac{F^{(k)}(E) - E}{E}. \tag{20.9}$$

The solutions for $x(t)$, $v(t)$ are given, as before, by Eqn (14.4), whereas $E(t)$ must be determined from Eqn (20.6).

Now let us consider the relevant Langevin and Fokker–Planck equations, which can be written by analogy with Eqns (14.6) and (14.8). The first of these is

$$\frac{dE}{dt} = \frac{1}{k} [F^{(k)}(E) - E] + \frac{1}{2} \frac{d}{dE} [D_{(k)}(E)E] \\ + \sqrt{D_{(k)}(E)E} y(t). \tag{20.10}$$

The moments of the source $y(t)$ are given by Eqn (14.7). The corresponding Fokker–Planck equation can be represented as

$$\frac{\partial f}{\partial t} = \frac{\partial}{\partial E} [D_{(k)}(E)E \frac{\partial f}{\partial E}] + \frac{\partial}{\partial E} \left\{ -\frac{1}{k} [F^{(k)}(E) - E] f \right\}. \tag{20.11}$$

The nonlinear coefficient of diffusion is again found under the assumption that in the absence of feedback ($\alpha_f = 0$) the system is at equilibrium, and the solution is the Boltzmann distribution (18.1):

$$D_{(k)}(E) = \frac{1}{k} [E - F^{(k)}(E)] \Big|_{\alpha_f=0}, \quad D_{(1)}(E) = kT'(\gamma' + E). \tag{20.12}$$

In dimensional variables [see Eqns (20.1a, b)] the expression for $D_{(1)}$ coincides with Eqn (14.10). Accordingly, Eqn (20.11) with $k = 1$ coincides with the Fokker–Planck equation (14.8). The stationary solution of Eqn (20.11) is

$$f_0(E, \alpha_f) = C \exp \left[\frac{1}{k} \int_0^E \frac{F^{(k)}(E') - E'}{D_{(k)}(E')} dE' \right]. \tag{20.13}$$

Naturally, the structure of the distribution becomes more and more complicated as the number of iteration increases, because there are more and more maxima of the distribution. The locations of maxima are determined by the roots of the equation

$$E_{\max} = F^{(k)}(E_{\max}), \tag{20.14}$$

and therefore coincide with the locations of stationary points of the logistic equation (20.4).

Let us illustrate these results with a simple example, with $k = 2$ and $D_{(k)} = D = \text{const}$. The constant coefficient of diffusion implies that the noise is external (predetermined). At the same time, the noise is not parametric, and so there are no such difficulties as those encountered in Section 15.

Recall that in the dynamic regime at $k = 2$ the value of $\alpha = 2$ is the point of bifurcation, associated with ramification of the energy of the limiting cycle [see Eqn (20.8)].

For the sake of clarity we single out three particular cases.

1. The region below the bifurcation point ($\alpha < 2$). In the Gaussian approximation the stationary solution (20.13) becomes

$$f_0(E) = \left(\frac{1}{2\pi D} \frac{2 - \alpha}{2} \right)^{1/2} \exp \left[-\frac{1}{2D} \frac{2 - \alpha}{2} (E - \alpha)^2 \right], \\ 0 \leq \alpha < 2. \tag{20.15}$$

Here and below the lower limit $E = 0$ in the normalisation conditions is replaced by $E = -\infty$, which is justified if $\varepsilon = D\beta/\alpha^2 \ll 1$. From this distribution it follows that on approaching the critical point the dispersion of fluctuations increases as $2/(2 - \alpha)$ — that is, according to Curie's law for the nonequilibrium phase transition.

2. The critical point ($\alpha = 2$). The stationary solution (20.13) is then

$$f_0(E) = \frac{1}{\Gamma(1/4)} \left(\frac{2}{D}\right)^{1/4} \exp\left[-\frac{(E-\alpha)^4}{8D}\right], \quad \alpha = 2. \quad (20.16)$$

The relative dispersion is proportional to ε rather than to $\sqrt{\varepsilon}$.

3. The region above the bifurcation point ($\alpha > 2$). The distribution displays two maxima, and the partial Gaussian distributions are

$$f_{1,2} = \left(\frac{\alpha-2}{2\pi D}\right)^{1/2} \exp\left[-\frac{\alpha-2}{2D}(E-E_{1,2})^2\right],$$

$$E_{1,2} = 2 \pm \alpha - 2. \quad (20.17)$$

On approaching the critical point from above, the dispersion of fluctuations also increases according to Curie's law, but this time with a factor of $1/(\alpha-2)$.

To end this section, we shall demonstrate that the above transition across the critical point is, according to the criterion of the S-theorem, a process of self-organisation.

There is an entropy jump for the states above and below the critical point in the Gaussian approximation. By S_- and S_+ we denote the value of entropy below and above the critical point, respectively. Then the difference in entropies in the Gaussian approximation is

$$S_- - S_+ = \ln \sqrt{2} > 0. \quad (20.18)$$

So, when the critical point is crossed as α_f increases, the entropy decreases. Since in the neighbourhood of the critical point $|\alpha-2| \ll 2$ the mean energies have the same values above and below ($\langle E \rangle_- = \langle E \rangle_+$) we may use the criterion of the S-theorem, which indicates in this case that the state above the critical point is more ordered. This means that the transition into the region of bistability is a process of self-organisation.

The role of the parameter of order in this example of a nonequilibrium phase transition is played by the difference in the mean energies of the two branches,

$$\eta = \langle E_1 \rangle - \langle E_2 \rangle = 2\sqrt{\alpha-2}, \quad \alpha \geq \alpha_{cr} = 2. \quad (20.19)$$

We see that at the critical point the parameter of order is zero.

These results have been obtained in the Gaussian approximation. Because of this, they are quite similar to those obtained within the framework of Landau's theory of phase transitions of the second kind.

21. Oscillators in discrete time. Bifurcations of the energy of the limiting cycle and of the period of oscillations

Recall that the logistic equation (20.3) can be used for describing the transition to the state of dynamic chaos in discrete time through a sequence of period-doubling bifurcations. Alternatively the set of differential equations (20.6) can be employed for the description of bifurcations of energy of the limiting cycle. For the stationary distribution (20.13), this sequence of bifurcations corresponds to the appearance of new maxima at those values of E_{\max} which coincide with the locations of stationary points of the logistic equation at the k th step of iteration.

At large enough values of k one might expect to observe chaotic behaviour in the arrangement of maxima of the stationary distribution in that range of values of α_f which, according to the logistic equation, corresponds to the domain of dynamic chaos.

Now let us find a generalised logistic equation which would simultaneously describe a superposition of two cascades of bifurcations: bifurcations of energy of the limiting cycle and bifurcations of period doubling [17, 53, 54].

Making the substitution E_{n+k} we reduce the logistic equation (20.4) to the form

$$E_{n+1} = F^{(k)}(E_n), \quad k = 1, 2, \dots \quad (21.1)$$

As a result, we get a family of logistic equations. At $k = 1$ we return to the initial equation (20.3), the solution of which is well known. The sequence of bifurcations of period-doubling starts at $\alpha = 2$ and ends at the critical point at $\alpha_{cr} = 2.58$. The edge of the broadest window of order appears at $\alpha = \sqrt{8}$. The value of $\alpha = 3$ corresponds to the state of the most developed 'dynamic chaos'.

With $k = 2$ the bifurcation pattern becomes very different. At $\alpha = 2$ we now have the bifurcation of energy of the limiting cycle, and bistability occurs. Depending on the initial conditions, the system goes to either the upper or the lower branch. The process of period doubling now only starts at $\alpha = \sqrt{6}$, when for the logistic equation the period would already have quadrupled. A 'chaos-to-chaos' phase transition takes place at $\alpha = 2.6785$, which results in the state of 'dynamic chaos' inherent in the logistic equation.

Recall that the differential equation (20.6) at $k = 2$ describes only the bifurcations of energy values; there are no bifurcations of period doubling.

Finally, let us consider the case of $k = 3$.

For the differential equation (20.6) the limiting cycle with $E = \alpha$ is now stable up to $\alpha = \sqrt{8}$, where three stationary states with different energies are formed. The bifurcation diagram of the generalised logistic equation (21.1) is also changed dramatically.

The comparison of bifurcation diagrams at $k = 1$ and $k = 3$, respectively, reveals the difference between the states within the limits of the broadest window of order. At $k = 1$ we discern the states which correspond to all three values of the energy, whereas at $k = 3$ there is only the state corresponding to the largest energy value. This implies that at $k = 3$ only one of the three states has the domain of attraction of trajectories which is large enough to be observable.

It is possible to demonstrate the extent to which this distinction is detected by different criteria of order. The values of Shannon entropy differ considerably for the states within the window of order at $k = 1$ and $k = 3$, whereas the Lyapunov indices for these states are almost the same.

Now let us show how the Lyapunov functional A_F for Brownian motion in the Van der Pol oscillator can be used for exploring the possibility of transition from continuous to discrete time.

22. Criterion of stability upon transition to discrete time based on the H-theorem [17]

Let us return to the Fokker-Planck equation (14.8) for the distribution function of the energy of oscillations. In particular, this equation describes the process of time

evolution towards the stationary state. For numerical calculations this equation must be replaced by the appropriate equation in discrete time with step Δ . Denoting by $f(E, n)$ the corresponding distribution function, in place of Eqn (14.8) we get

$$\frac{f(E, n + \Delta) - f(E, n)}{\Delta} = \frac{\partial}{\partial E} \left[D(E)E \frac{\partial f}{\partial E}(E, n) \right] + \frac{\partial}{\partial E} [(-\alpha + \beta E)E f(E, n)]. \quad (22.1)$$

Observe that formally the stationary solutions of equations (14.8) and (22.1) in continuous and discrete time are given by the same expression (14.11). The problem is, however, whether these stationary solutions are stable. To answer this question we must analyse the behaviour of small deviations from the stationary solution $f_0(E, \alpha_f)$:

$$f_1(E, t) = f(E, t) - f_0(E), \quad f_1(E, n) = f(E, n) - f_0(E). \quad (22.2)$$

First we shall do this for the equation in continuous time. We make use of the H-theorem, which for the theory of Brownian motion is expressed by two inequalities (17.3) and (17.4) in the Lyapunov functional A_F , defined in terms of the difference in nonequilibrium free energies of the current and stationary states.

Substituting $f = f_0 + f_1$ into expression (17.3) for the difference of free energies, we carry out an expansion in f_1 and retain only the main term. Then for the Lyapunov functional A_F we find that

$$\frac{d}{dt}(F - F_0) = \frac{kT}{2} \frac{d}{dt} \int_{f_0}^1 f_1^2 dE \leq 0. \quad (22.3)$$

Hence it follows that f_1 decreases with time, and the stationary solution of the Fokker-Planck equation (14.8) is stable.

Now let us consider the same problem in discrete time, when the Fokker-Planck equation has the form of Eqn (22.1).

Introduce a factor q which links the values of f_1 at two consecutive instants of discrete time with step Δ :

$$f_1(E, n + \Delta) = q f_1(E, n). \quad (22.4)$$

Now we use definition (17.4) of ‘entropy production’ $\sigma_F \geq 0$, which for small f_1 can be represented as

$$\sigma_F = kT \int_0^\infty \frac{DE}{f_0} \left(\frac{\partial f_1}{\partial E} \right)^2 dE \geq 0. \quad (22.5)$$

The time derivative in Eqn (22.3) we replace by the finite difference,

$$\begin{aligned} \frac{d}{dt}(F - F_0) &= kT \int_{f_0}^{f_1} \frac{\partial f_1}{\partial t} dE \\ &\rightarrow kT \int \frac{f_1(E, n) f_1(E, n + \Delta) - f_1^2(E, n)}{\Delta} dE \\ &= \frac{kT}{\Delta} (q - 1) \int \frac{f_1^2(E, n)}{f_0} dE = -\sigma_F \leq 0, \end{aligned} \quad (22.6)$$

where q is as defined by Eqn (22.4).

From the condition of stability, which requires that $|f_1|$ should decrease as the discrete time passes, it follows that

$|q| < 1$. The necessary and sufficient condition of stability, which follows from Eqn (22.6), is then given by [17]

$$|q| = \left| 1 - \Delta \frac{\sigma_F}{kT \int_{f_0}^1 f_1^2 dE} \right| \leq 1, \quad (22.7)$$

which can be written in a more concise form in terms of the Lyapunov functional A_F :

$$|q| = \left| 1 - \frac{1}{2} \Delta \frac{\sigma_F}{A_F} \right| \leq 1, \quad A_F = \frac{kT}{2} \int_{f_0}^1 f_1^2 dE. \quad (22.8)$$

We see that the criterion of stability of the stationary solution is expressed in terms of the general thermodynamic characteristics of the nonequilibrium process: the analogue of ‘entropy production’ σ_F and the Lyapunov functional A_F .

From Eqn (22.8) we find that the region of stability is delimited by the two-way inequality

$$0 \leq \frac{1}{2} \Delta \frac{\sigma_F}{A_F} \leq 2, \quad \Delta_{\max} = 4 \frac{A_F}{\sigma_F}. \quad (22.9)$$

We see that it is always possible to make the step of discrete time small enough to ensure stability of the stationary distribution with respect to a small deviation of arbitrary form.

To particularise this criterion of stability, let us specify the form of a nonequilibrium distribution. We assume that the nonequilibrium distribution can be obtained from the stationary distribution by varying the parameter of feedback (the controlling parameter). Then the effective Hamilton function in Eqn (14.11) must be replaced: $H_{\text{eff}} \rightarrow H_{\text{eff}} + \delta\alpha_f E$. In the simplest case, when the coefficient of diffusion is constant [see Eqn (14.12)], and the variation $\delta\alpha_f$ is small, the values of σ_F and A_F can be defined as

$$\sigma_F = \gamma (\delta\alpha_f)^2 \langle E \rangle, \quad A_F = (\delta\alpha_f)^2 \frac{\langle (\delta E)^2 \rangle}{2kT}, \quad (22.10)$$

and are thus expressed in terms of the mean energy and the dispersion of energy for the stationary distribution. The condition of stability then becomes

$$0 \leq \Delta \gamma kT \langle E \rangle / \langle (\delta E)^2 \rangle \leq 2. \quad (22.11)$$

In particular, the value of the largest permissible time step is thus given by

$$\Delta_{\max} = 2 \langle (\delta E)^2 \rangle / (\gamma kT \langle E \rangle). \quad (22.12)$$

Observe that $\#$ implicitly depends on the parameter of feedback via the mean energy and the dispersion. For the regime of well-developed generation these results can be reduced to a very simple form with the aid of Eqn (14.15):

$$0 \leq \Delta \alpha \leq 2, \quad \Delta_{\max} = 2\alpha^{-1}. \quad (22.13)$$

These expressions do not contain either the temperature or the coefficient of diffusion, because they pertain to the region of well-developed generation, which corresponds to the limit of $T \rightarrow 0$ when the dynamic distribution holds good. In our present case the distribution follows from Eqn (14.14) and has the form

$$f_0(E, \alpha_f) = \delta(E - \alpha\beta^{-1}), \quad \langle E \rangle = \alpha\beta^{-1}. \quad (22.14)$$

These results can also be used in a different way. Namely if we define the value of the time step (for instance, by

setting it equal to one, $\Delta = 1$), from Eqn (22.13) we find that the stationary solution is stable when

$$0 \leq \alpha \leq 2, \quad \alpha_{\max} = 2. \quad (22.15)$$

This condition coincides with the condition of stability of the limiting cycle, when the evolution in discrete time is described by the logistic equation (20.3). This is natural because we started with the Fokker–Planck equation (14.8), from which we obtained Eqn (22.1) in discrete time.

Now we can redo all our calculations, and consider the more general Fokker–Planck equation (20.11) for the next level with $k = 2$. It corresponds to the stationary solution (20.13) with $k = 2$, which describes the ramification of the energy of the limiting cycle. Bifurcation occurs exactly at $\alpha = 2$, at the limit of stability as defined by Eqn (22.15).

By way of generalisation, we may tackle the problem of stability of the most general stationary distribution (20.13) at arbitrary numbers k . The regions in which dynamic distributions hold good will be narrowed as k increases; because of this, the analysis of the limits of stability of stationary states on the basis of the H-theorem becomes even more important.

Recall that the final expressions (22.13)–(22.15) have been obtained for the region of well-developed generation. For comparison let us consider another distinct state, the threshold of generation. This will show us how strong the dependence of the limits of stability on the value of the controlling parameter can be.

From distribution (18.2) it follows that the relative dispersion of energy is of the order of unity, and that $\langle E \rangle \sim \sqrt{D/\beta}$. Given this, from Eqn (22.12) we find the maximum permissible time step at the generation threshold: $(\Delta_{\max})_{\text{th}} \sim \sqrt{\beta D}$. Then

$$(\Delta_{\max})_{\text{th}} A_{\max}^{-1} \sim \sqrt{\alpha^2 (\beta D)^{-1}} \sim \sqrt{\varepsilon^{-1}} \gg 1, \quad (22.16)$$

where ε is the small parameter from the theory of well-developed generation.

We see that upon transition to discrete time the state of well-developed generation loses stability with much smaller values of Δ_{\max} than does the state at the generation threshold. Comparing this conclusion with calculations of the relative degree of order of the same states on the basis of S-theorem, we find that the higher organised state is more vulnerable upon transition to discrete time. In other words, it has less ‘stability margin’ than the more chaotic state at the threshold of generation. One might ask whether this is good news or bad news. There is no categorical answer to this question. Sometimes the loss of stability may result in a higher organised state, which is good from the standpoint of self-organisation. Such was the case with the ramification of the values of energy of the limiting cycle, and with the period-doubling bifurcations.

If, however, the loss of stability results in transition to a more chaotic state, this is bad as far as self-organisation is concerned. In this respect the higher organised state is more fragile.

So far we have been considering examples based on various modifications of the Van der Pol oscillator. Now we shall give some examples from other areas.

23. Brownian motion in chemically reacting systems. Partially ionised plasma

Consider an example of Brownian motion when the ‘Brownian particles’ are represented by the fluctuations of density of electrons, ions, and atoms in a partially ionised plasma. By ‘chemical reactions’ we refer to the processes of ionisation (for instance, electron impact ionisation), and the processes of recombination involving three charged particles.

The theory of such fluctuations can be based on the set of kinetic equations for the distribution functions of electrons, ions and atoms [33, 55–57]. In particular, one may define the appropriate Langevin sources in the kinetic equations for the distributions of electrons, ions, and atoms.

Relaxation towards the equilibrium or (in open systems) the stationary state proceeds, as a rule, in several stages. In many cases, local equilibrium with respect to translational degrees of freedom is achieved at the first stage, followed by the stage when local equilibrium with respect to internal degrees of freedom is established, and followed finally by the stage when chemical equilibrium sets in.

Consider the state of the plasma after completion of the first two stages of relaxation. Neglecting fluctuations, the evolution towards chemical equilibrium can be described on the basis of equations for the mean densities of electrons n_e , ions n_i , and atoms n_a . Of all possible processes of conversion of particles in a plasma we select just one, the process of ionisation by electron impact. By α and β we denote the coefficients of ionisation by electrons and recombination of an electron and an ion in the presence of an electron, respectively. Then the dynamic equations can be written as

$$\frac{dn_e}{dt} = \alpha n_e n_a - \beta n_e^2 n_a, \quad n_e = n_i, \quad n_e + n_a = n. \quad (23.1)$$

The second equation states that the average numbers of electrons and ions are equal, and the third equation ensures that the total number of charged particles (for instance, electrons and atoms) is constant. At equilibrium, the so-called Saha’s formula holds:

$$\frac{n_e n_i}{n_a} = \frac{\alpha}{\beta}, \quad \left\{ \frac{\alpha}{\beta} = \left[\frac{\mu k T}{(2\pi\hbar)^2} \right]^{3/2} \frac{1}{Z} \right\}. \quad (23.2)$$

Next we define the Langevin sources in the kinetic equations so as to take the atomic structure of the ‘continuous medium’ into account. This will also allow us to find the relevant Langevin sources in the equations in random functions n_e , n_i , n_a . The intensity of the Langevin source, which acts as the coefficient of diffusion in the corresponding Fokker–Planck equation for the distribution function $f(n, t)$, is

$$D_{n_e} = \frac{1}{2V} (\alpha n_e n_a + \beta n_e^2 n_i), \quad (23.3)$$

where V is the volume, and thus the particles are assumed to be uniformly distributed in space. Under this condition, two equations in Eqn (23.1) also remain valid when fluctuations are taken into account. Then Eqn (23.3) can be rewritten as

$$D_{n_e} = \frac{1}{2V} [\alpha n_e (n - n_e) + \beta n_e^3] = \frac{1}{2} (r_{n_e} + g_{n_e}). \quad (23.4)$$

The coefficient of nonlinear friction follows from the dynamic equation (23.1):

$$A_{n_c} = -[\alpha n_c(n - n_c) - \beta n_c^3] \equiv r_{n_c} - g_{n_c}. \quad (23.5)$$

To establish correspondence with the structure of the master equation for the one-step processes of Section 9, we have again introduced the coefficients of generation and recombination. Then, by analogy with Eqns (9.5) and (9.17), we may immediately write the Fokker–Planck equation for distribution function $f(n_c, t)$:

$$\frac{\partial f}{\partial t} = \frac{\partial}{\partial n_c} \left(D_{n_c} \frac{\partial f}{\partial n_c} \right) + \frac{\partial}{\partial n_c} (A_{n_c} f), \quad \int f(n_c, t) dn_c = 1. \quad (23.6)$$

The stationary solution of this equation is

$$f(n_c) = C \exp \left[- \int_0^{n_c} \frac{A(n'_c)}{D(n'_c)} dn'_c \right]. \quad (23.7)$$

Hence it follows that the maximum of the distribution is defined by the solution of the equation

$$A(n_{\max}) = -[\alpha n_{\max}(n - n_{\max}) - \beta n_{\max}^3] = 0. \quad (23.8)$$

The location of the maximum of the distribution satisfies Saha’s formula.

In the Gaussian approximation the dispersion $\delta n_c = n_c - n_{\max}$ is given by

$$\langle (\delta n_c)^2 \rangle = \frac{n_c}{V} \frac{n - n_c}{(2n - n_c)}. \quad (23.9)$$

The dispersion vanishes in two extreme cases: when the degree of ionisation is zero (the gas of neutral particles, $n_c = 0$), and when the plasma is completely ionised ($n_c = n$). This indicates that our source of fluctuations reflects the fact that the acts of chemical transformation are discrete; the dispersion is zero in the above two cases because chemical reactions do not take place.

For small deviations from equilibrium, the fluctuation δn satisfies the linear Langevin equation with the coefficient of friction $\lambda = \alpha(2n - n_c)$. The expression for the diffusion coefficient can then be reduced to the standard form of the fluctuation–dissipation relation:

$$D = \lambda \langle (\delta n_c)^2 \rangle, \quad \lambda = \alpha(2n - n_c), \quad (23.10)$$

whence it follows that the coefficient of diffusion also vanishes in the two extreme cases specified above.

Making use of the definitions (23.4), (23.5) for the coefficients of generation and recombination, we may write the appropriate master equation. In the context of statistical theory, of the two possible definitions of transition probabilities (9.2) and (9.11) the latter is more ‘natural’. It is only then that the K-form of the Fokker–Planck equation follows from the master equation.

In the case of the master equation it is more natural to use the total numbers of particles rather than the densities:

$$N_c = Vn_c, \quad N_i = Vn_i, \quad N_a = Vn_a, \quad N = Vn. \quad (23.11)$$

In these variables the dynamic equation (23.1) becomes

$$\frac{dN_c}{dt} = \alpha_v N_c N_a - \beta_v N_c^2 N_i, \quad \alpha_v = \frac{\alpha}{V}, \quad \beta_v = \frac{\beta}{V^2}. \quad (23.12)$$

In the presence of an external source (for instance, photoionisation in a given light field), it is possible to monitor the degree of order of a partially ionised plasma as a function of, for instance, the strength of the external field.

24. The Malthus–Verhulst process

The Malthus–Verhulst model was proposed many years ago for describing the survival of a population, for instance, of bacteria [11, 15]. Let N be the number of species in the population. If γ and α are, respectively, the rates of death and of birth, and $\beta(N - 1)$ is the rate of extinction because of competition between the species, then the process of time evolution at $N \gg 1$ can be described by the following differential equation:

$$\frac{dN}{dt} = (\alpha - \gamma - \beta N)N \equiv (g_N - r_N)N. \quad (24.1)$$

Here we have factored out the common multiplier N in the definitions of coefficients g_N, r_N . Equation (24.1) is similar in structure to Eqn (14.3) for the energy of oscillations in the Van der Pol oscillator. Observe that the coefficient of feedback α_f corresponds here to the birth rate α .

In this connection let us recall that the nonlinear coefficient of diffusion, given by Eqn (14.10), in the Fokker–Planck equation (14.8) has been defined from the condition that the stationary distribution in the absence of feedback ($\alpha_f = 0$) should coincide with the Boltzmann distribution. Following the same hypothesis, we must now define the nonlinear coefficient of diffusion as

$$D(N) = (\gamma + \beta N)T, \quad (24.2)$$

where T is the dimensionless counterpart of the temperature, which, as we shall see, can be assumed to be unity.

Alternatively one may define the coefficient of diffusion in terms of the coefficients of ionisation and recombination by formulas (9.5) and (23.4). In this case, for Eqn (24.1) we have

$$g_N = \alpha N, \quad r_N = (\gamma + \beta N), \quad D = (\gamma + \beta N) - \frac{1}{2}\alpha. \quad (24.3)$$

Thus defined, the coefficient of diffusion depends on the ‘feedback parameter’. As the feedback parameter increases, the system displays a phase transition which results in a stationary state with a nonzero number N . However, the sign of the coefficient of diffusion may change. To avoid this complication, we shall stick to the definition (24.2) with $T = 1$. As before, we give preference to the K-form of the Fokker–Planck equation:

$$\frac{\partial f(N, t)}{\partial t} = \frac{\partial}{\partial N} \left[(\gamma + \beta N)N \frac{\partial f}{\partial N} \right] + \frac{\partial}{\partial N} \left[(-\alpha + \gamma + \beta N)NF \right]. \quad (24.4)$$

If the birth rate α is zero, the stationary distribution takes on the form

$$f_0(N) = \exp(-N), \quad \int f_0 dN = 1. \quad (24.5)$$

As this description holds for $N \gg 1$, the exponential decrease means that the population is actually doomed to extinction.

By analogy with Eqn (14.11), the general stationary solution can be represented as

$$f_0(N, \alpha) = C \exp[-H_{\text{eff}}(N, \alpha)],$$

where

$$H_{\text{eff}} = N - \frac{\alpha}{\beta} \ln \left(1 + \frac{\beta}{\gamma} N \right), \quad (24.6)$$

is the relevant Hamilton function. The maximum of the distribution is given by

$$N_{\max} = (\alpha - \gamma)\beta^{-1}, \quad (24.7)$$

which expresses a quite natural requirement: birth rate must be high enough ($\alpha > \gamma$) to ensure viability of the population.

If the relative dispersion of the number of species is small, we may go over from Eqn (24.6) to the Gaussian distribution by expanding in small deviation $\delta N = N - N_{\max}$. The condition of smallness of the relative dispersion is similar to that given by Eqn (14.16):

$$\frac{\langle(\delta N)^2\rangle}{N_{\max}^2} = \frac{\gamma}{\alpha - \gamma} \frac{1}{N_{\max}} \ll 1. \quad (24.8)$$

The appropriate master equation must be used for describing processes with a small number of species. In this case also, there are strong arguments which shift the balance in favour of the kinetic representation of the master equation.

Practical applications of the nonlinear theory of Brownian motion are numerous; they are discussed in specialised literature. Our main task consisted of elucidating the important new aspects which are necessary for further development of the theory. Of course, some important matters had to be left out — in particular, the feasibility of the unified kinetic description of dissipation and diffusion both in the space of velocities and in conventional space. This is briefly discussed in Refs [27, 28].

The use of appropriate generalised kinetic equations in the theory of Brownian motion allows one not to resort to the methods of perturbation theory used in Sections 10 and 11 for transition from the Fokker–Planck equation to the Einstein–Smoluchowski equation. This opens the possibility of describing Brownian motion in systems which display both dissipative and nondissipative nonlinearity. The simplest example of such a system is the Van der Pol–Duffing oscillator, which involves at least two controlling parameters: the parameter of feedback in the Van der Pol oscillator, and the ‘parameter of effective field’ in the Duffing oscillator. By varying the second parameter it is possible to produce a transition similar to an equilibrium phase transition of the second kind. The potential may then change from parabolic to, for example, bistable.

New phenomena, caused by the interplay of equilibrium and nonequilibrium phase transitions, may take place in such systems. For instance, in the well-known Kramers problem of surpassing a barrier, the characteristic time of transition can be controlled by varying the parameter of feedback in the Van der Pol oscillator. In systems with chemical reactions the nonequilibrium dissipative processes may affect the reaction rates through different channels. Accordingly, one may speak of the kinetic theory of catalysis, when the reaction rate is controlled by varying the parameter of feedback in the Van der Pol oscillator.

Obviously, such problems are very complicated. The author may only hope that this paper will stimulate further development of the nonlinear theory of Brownian motion.

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