Generalization of the Langevin Equation in Fluctuation Dissipation Systems

Toshio TAKEHARA

Department of Physics, Faculty of Science, Shimane University, Matsue, Japan
(Received September 14, 1985)

Langevin's equation as an adequate description of Brownian motion is applicable in large variety of systems which are close to equilibrium. For these systems hydrodynamic equations describe the long wave-length, low-frequency phenomena. The equation of motion for these systems of continuous media is generalized to the similar type of Langevin equation and correlation of fluctuation is discussed.

§1. An Abridged Description of Brownian Motion

Fluctuations about the equilibrium state decay on the average by the linear macroscopic law which describes the decay of the system from a nonequilibrium state to equilibrium state. A sufficiently small macroscopic particle immersed in a liquid exhibit a random motion, which is called Brownian motion, and this phenomenon reveals very clearly the statistical fluctuations which occur in a system in thermal equilibrium. There are a variety of similar phenomena which are recognized earlier. For instance, random motion of the mirror suspended by a fiber of a sensitive galvanometer, or fluctuating noise voltage due to thermal fluctuating current in an electric resistor. Thus Brownian motion can serve as a prototype case, and its analysis is described in standard texts[1] and provides considerable insight into mechanisms responsible for the existence of fluctuations and dissipation of energy.

For the simplicity we shall treat one-dimensional motion of the Brownian particle. The equation of motion of a particle is expressed phenomenologically

$$m\frac{dv}{dt} = -m\gamma v + F(t) \tag{1.1}$$

where the term $-m\gamma v$ represents the usual viscous drag on a particle moving with velocity v, and $F(t)$ is a random force due to density fluctuations in the medium with average value zero. For the given function $F(t)$ the equation is integrated and one obtains

$$v(t) = v(t_0)e^{-\gamma(t-t_0)} + \frac{1}{m}\int_{t_0}^{t} e^{-\gamma(t-s)} F(s)ds \qquad (t \geq t_0) \tag{1.2}$$

For simplicity, $v(t_0)$ is assumed finite and in the limit $t_0 \to -\infty$

$$v(t) = \frac{1}{m} \int_{-\infty}^{t} e^{-\gamma(t-s)} F(s) ds, \quad \text{and} \quad \langle v(t) \rangle = 0 \tag{1.3}$$

The correlation function of the fluctuating force $F(t)$ is assumed to be

$$\langle F(t)F(t') \rangle = 2\Gamma\delta(t-t') \tag{1.4}$$

To derive the correlation function of velocities

$$\langle v(t)v(t') \rangle = \frac{1}{m^2} \int_{-\infty}^{t} e^{-\gamma(t-s)} ds \int_{-\infty}^{t} e^{-\gamma(t'-s')} 2\Gamma\delta(s-s') ds'$$

$$= \frac{1}{m^2} \int_{0}^{\infty} e^{-\gamma u} du \int_{0}^{\infty} e^{-\gamma u'} 2\Gamma\delta(t-t'-u+u') du'$$

$$= \frac{2\gamma\Gamma}{m^2} e^{\gamma(t-t')} \int_{0}^{\infty} e^{-2\gamma u} du \int_{0}^{\infty} \delta(t-t'-u+u') du' \tag{1.5}$$

$$= \frac{2\gamma\Gamma}{m^2} e^{\gamma(t-t')} \int_{\max(0,t-t')}^{\infty} e^{-2\gamma u} du$$

we obtain

$$\langle v(t)v(t') \rangle = \frac{2\Gamma}{m} e^{-\gamma(t-t')} \tag{1.6}$$

This functional dependence means that the velocity correlations are only in a time interval of the order of $1/\gamma$. The larger the magnitude of the friction constant this time interval is shorter. Mean square displacement is also evaluated easily and we find the relation

$$\langle x^2 \rangle = 2Dt \tag{1.7}$$

where D is the diffusion coefficient, and famous Einstein relation

$$D = \mu k_B T$$

is derived, and which relates D and mobility μ of the particle in the system of temperature T.

§2. Phenomenological Equation for Transport Processes

We consider systems which are not in equilibrium but are close to equilibrium. We assume that all disturbances are slowly varying in space and have small amplitude. If the system is in equilibrium and its states can be specified via the thermodynamic variables. However, the values of such state variable can vary from one part to the

other gradually in the system.   When a system is disturbed from its equilibrium state, most of the quantity decay rapidly to their equilibrium through the impact of molecular collison, several quantities which are not conserved during molecular collison remain out of equilibrium.   These quantities are densities of state variables and characterize the nonequilibrium behavior of the fluid after long times.   For the equation of motion of these densities of such quantity we call the hydrodynamic equation from the phenomenological point of view.   Examples of these quantities are the number of particles, the total momentum of particles, and the total kinetic energy of particles.   If there are inhomogeneities in the densities of these quantities, the particles, momentum, or kinetic energy must be transported from one part of the fluid to another to achieve equilibrium.   The rate at which the currents induced by the density gradients to return back to equilibrium state is determined by the transport coefficients.   These transport phenomena are the self-diffusion, viscosity, and thermal conductivity and many other processes.   The macroscopic equations — Fick's law for diffusion, Fourier's law for thermal conduction — are derivable from a more rigorous microscopic equation, the Boltzmann equation.   The Boltzmann equation is a nonlinear integrodifferential equation for the distribution, but for small amplitude of disturbance we can linearize the equation.

As an example of linearized hydrodynamic equation, we illustrate the self diffusion process.   For the particle density $m(r, t)$ at some point r

$$\frac{\partial}{\partial t} m(r, t) = D\nabla^2 m(r, t) \tag{2.1}$$

To find the dispersion relation for hydrodynamic modes we define the Fourier transform

$$m(r, t) = \frac{1}{(2\pi)^4} \int dk \int d\omega\, e^{-(k \cdot r - \omega t)} \mu(k, \omega) \tag{2.2}$$

which allows us to examine each Fourier component of diffusion equation.   If we substitute Eq. (2.2) into Eq. (2.1) we obtain

$$-i\omega\mu(k, \omega) + Dk^2\mu(k, \omega) = 0 \tag{2.3}$$

Note that different Fourier components do not couple, because the hydrodynamic equation is linear.   From Eq. (2.3) we obtain the following dispersion relation for the self-diffusion mode:

$$\omega = -iDk^2. \tag{2.4}$$

The diffusion frequency is imaginary, which means that the contribution to the density $m(r, t)$ with wave vector k, decay out in a time which depends on the diffusion coefficient D and the wave number k,

$$m(r, t) \simeq e^{ik \cdot r} e^{-Dk^2 t}. \tag{2.5}$$

Thus, component of very long wavelength disturbance take a long time to decay. This behavior is characteristic of a hydrodynamic mode.

§3. Time-Dependent Correlation Function and Microscopic Reversibility

Onsager showed that the reversibility of the dynamical laws on the microscopic level requires that, without any reference to a particular physical system, certain relation exist between the transport coefficients for coupled transport processes.

We assume that the macroscopic state of the system is described by n independent macroscopic state variable A_i ($i=1, 2,..., n$), and in an equilibrium state A_i^0. Let us denote α_i the fluctuations

$$\alpha_i = A_i - A_i^0 \tag{3.1}$$

The time dependent correlation functions of macroscopic fluctuations, $\alpha = (\alpha_1, \alpha_2,..., \alpha_n)$, obey the relations

$$\langle \alpha_i \alpha_j(\tau) \rangle = \langle \alpha_i(\tau)\alpha_j \rangle \tag{3.2}$$

Eq. (3.2) tells us that the correlation between a fluctuation α_i at time $t=0$ and a fluctuation α_j at time $t=\tau$ is the same as that of a fluctuation α_j at time $t=0$ and a fluctuation α_i at time $t=\tau$. The quantities α_i and α_j can correspond to fluctuations in the same state variables at different points in space. Thus, Eq. (3.2) can also be understood as an equation relating correlations between space- and time-dependent fluctuations.

Probability distribution for fluctuations about the equilibirum state is obtained by Einstein[3] in the following way. Let us consider a closed adiabatic isolated system, which is ergodic so that all possible microscopic states of the system are equally probable. Let $\Gamma(E)$ denote the number of microscopic systems with energy $(E, E+dE)$. Then the entropy of the system is given by

$$S = k_B \ln \Gamma(E) \tag{3.3}$$

We assume that, in addition to the energy E, the macroscopic state of the system is describable in terms of n independent macroscopic state variables A_i ($i=1,..., n$). Then the probability that the system is in a macroscopic state described by variables E, $A_1, A_2,..., A_n$ is given by

$$p(E, A_1,..., A_n) = \frac{\Gamma(E, A_1,..., A_n)}{\Gamma(E)} \tag{3.4}$$

The entropy of the system with variables $(E, A_1,..., A_n)$ is given by

$$S(E, A_1,..., A_n) = k_B \ln \Gamma(E, A_1,..., A_n) \tag{3.5}$$

Hence

$$p(E, A_1,..., A_n) = \frac{1}{\Gamma(E)} \exp\left[S(E, A_1,..., A_n)/k_B\right] \tag{3.6}$$

The entropy will be a maximum when the system is in an equilibrium state $A_1^0,..., A_n^0$. Any fluctuation about the equilibrium state must cause the entropy to decrease. If we let α_i denote the fluctuations $\alpha_i = A_i - A_i^0$ then we can expand the entropy about its equilibrium value to obtain

$$S(E, A_1,..., A_n) = S(E, A_1^0,..., A_n^0) - \frac{1}{2}\sum_{i=1}^{n}\sum_{j=1}^{n} g_{ij}\alpha_i\alpha_j + \cdots \tag{3.7}$$

where

$$g_{ij} = -\left[\frac{\partial^2 S}{\partial A_i \partial A_j}\right]_{)0} \tag{3.8}$$

The matrix (g_{ij}) is positive definite and the first order term in Eq. (3.5) must be identically zero since the quantities are symmetric. We can obtain the following expression for the probability distribution of fluctuations about the equilibrium state

$$p(\boldsymbol{\alpha}) = C \exp\left\{-\frac{1}{2k_B}\boldsymbol{G\alpha\alpha}\right\} \tag{3.9}$$

where $\boldsymbol{\alpha} = (\alpha_1,..., \alpha_n)$ is a vector and

$$\boldsymbol{G\alpha\alpha} = \sum_{i,j} G_{ij}\alpha_i\alpha_j$$

A normalization constant is defined

$$C\int_{-\infty}^{\infty} p(\boldsymbol{\alpha})d\boldsymbol{\alpha} = C\left[\frac{(2\pi k_B)^n}{|G|}\right]^{1/2} = 1 \tag{3.10}$$

where $d\boldsymbol{\alpha} = d\alpha_1,..., d\alpha_n$. Thus

$$C = \left[\frac{|G|}{(2\pi k_B)^n}\right]^{1/2} \tag{3.11}$$

and $|G|$ is the determinat of the matrix $\boldsymbol{G}$.

The correlation matrix has the property

$$\boldsymbol{C}_{\alpha\alpha}(\tau) \equiv \langle\boldsymbol{\alpha}(\tau)\boldsymbol{\alpha}\rangle = \langle\boldsymbol{\alpha}(t+\tau)\boldsymbol{\alpha}(t)\rangle = \langle\boldsymbol{\alpha}(-\tau)\boldsymbol{\alpha}\rangle^T = \boldsymbol{C}_{\alpha\alpha}^T(-\tau) \tag{3.12}$$

where we have taken $t = -\tau$ and T denotes the transpose of the correlation matix. From the Onsager relation $\langle\boldsymbol{\alpha}(\tau)\boldsymbol{\alpha}\rangle = \langle\boldsymbol{\alpha}(-\tau)\boldsymbol{\alpha}\rangle^T$, and therefore,

$$\boldsymbol{C}_{\alpha\alpha}(\tau) = \boldsymbol{C}_{\alpha\alpha}^T(\tau) \tag{3.13}$$

To find the moments of the fluctuation, it is convenient to introduce a more generalized integral

$$C \int_{-\infty}^{\infty} \exp \left\{ -\frac{1}{2k_B} (G\alpha\alpha) + h \cdot \alpha \right\} d\alpha = \exp \left\{ \frac{1}{2} k_B G^{-1} hh \right\} \qquad (3.14)$$

where h is a vector of n dimension.

The second moment $\langle \alpha_i \alpha_j \rangle$ is derived in the following relation.

$$\langle \alpha_i \alpha_j \rangle = \lim_{h \to 0} \frac{\partial}{\partial h_i} \frac{\partial}{\partial h_j} \left[C \int_{-\infty}^{\infty} \exp \left\{ \frac{1}{2k_B} G\alpha\alpha + h \cdot \alpha \right\} d\alpha \right] = k_B (G^{-1})_{ij} \qquad (3.15)$$

and we have

$$C_{\alpha\alpha}(0) = \langle \alpha\alpha \rangle = k_B G^{-1} \qquad (3.16)$$

§4. Time Evolution of Fluctuations

We must remind that α is a macroscopic variable. Thus for each value of α the given initial value of α was α_0. Then for the conditional average of α at time t we can write

$$\langle \alpha(t) \rangle_{\alpha_0} = \int P(\alpha_0 | \alpha, \tau) d\tau. \qquad (4.1)$$

$P(\alpha_0 | \alpha, \tau)$ is the conditional probability that a system turn from the state α_0 to the state α at time $t = \tau$. Onsager assumed that, on the average, the fluctuations decay according to the similar law by the hydrodynamic equations. Thus the average fluctuation $\langle \alpha(t) \rangle_{\alpha_0}$ obeys an equation of the form

$$\frac{d}{dt} \langle \alpha(t) \rangle_{\alpha_0} = -Q \cdot \langle \alpha(t) \rangle \qquad (4.2)$$

Eq. (4.2) has the solution

$$\langle \alpha(t) \rangle_{\alpha_0} = e^{-Qt} \cdot \alpha_0. \qquad (4.3)$$

Eq. (4.1) imposes a condition on the matrix Q, if we expand Eq. (4.3) for short times,

$$\langle \alpha(\tau) \rangle_{\alpha_0} = \alpha_0 - Q \cdot \alpha_0 t + O(t^2), \qquad (4.4)$$

and substitute into Eq (3.2)

$$\langle \alpha_0 Q \alpha_0 \rangle = \langle Q \alpha_0 \alpha_0 \rangle \qquad (4.5)$$

If we now use the fact that $Q\alpha = \alpha^T Q^T$ and use Eq. (3.13), we obtain

$$G^{-1} Q^T = Q G^{-1} \qquad (4.6)$$

We can define a new tensor

$$L = QG^{-1};$$ (4.7)

then Eq. (4.6) becomes

$$L = L^T \quad \text{or} \quad L_{ij} = L_{ji}$$ (4.8)

Eqs. (4.8) are Onsager relations. If the system is subject to the generalized force F, which is introduced from the equation with $\varDelta S = S - S_0$

$$F = -\left(\frac{\partial \varDelta S}{\partial \alpha}\right) = G\alpha$$ (4.9)

the time rate of change of the fluctuation can be written

$$\frac{d}{dt}\langle \alpha(t)\rangle_{\alpha_0} = -L \cdot \langle \alpha(t)\rangle_{\alpha_0}$$ (4.10)

This is the general expression of linearized hydrodynamic equations. These equations describe the evolution of slowly varying modes. We can account for the effects of rapidly fluctuating modes on hydrodynamic evolution by adding a random noise term to these equations. We can write for α_i

$$\frac{d\alpha_i(t)}{dt} = -L_i\alpha_i(t) + F_i(t)$$ (4.11)

where L_i is an Onsager coefficient and is related to the inverse damping of the mode and $F_i(t)$ is a delta correlated noise source, $\langle F_i(t)F_j(t')\rangle = 2\Gamma_i\delta_{ij}\delta(t - t')$ and Γ_i is the strength of the noise. Assume that $\langle \alpha_i(t)F_i(t)\rangle = 0$ (This means that $\alpha_i(t)$ varies slowly compared to $F_i(t)$). As in Brownian motion

$$\Gamma_i = L_i\langle \alpha_i^2\rangle$$ (4.12)

The power spectral density is given by the Wiener-Khinchin's theorem

$$S_{\alpha\alpha}(\omega) = \int_{-\infty}^{\infty} \langle \alpha\alpha(\tau)\rangle e^{i\omega\tau}d\tau$$

and

$$S_{ij}(\omega) = 2\Gamma_i\delta_{ij}/[\omega^2 + L_i^2]$$ (4.13)

§5. Spin Waves in a Macroscopic Continuum Model

In the ferromagnetic system, the exchange interaction which give rise to the spontaneous magnetization M_s resist strongly any change in the direction of M_s from point to point. The excitation of spin waves of precessing magnetization can be interpreted

quantitatively by treating the magnetic material as a continuous field of magnetization
M, having constant magnitude and with the variation in direction from point to point
there is associated an excess energy density. This continuum approach for gradual
disturbances of a uniform magnetization will break down if the spin on one site vary
markedly in direction, but a phenomenological approach is justified for gradual vari-
ations, and is analogous to continua theories of the elastic properties of solids.

We need an expression for the excess energy associated with nonuniform magneti-
zation to the lowest order in the deviation of M. Herring and Kittel[4] pointed out
that, for a field of constant magnitude, $-M \cdot \nabla^2 M$ is the expression of the lowest order
among those which are invariant to rotations of coordinate axes for isotropic exchange
interactions and which give the uniform field as uniquely that of least energy.

The existence of an excess energy implies that a torque acts on M whenever M is
non-uniform, tending to make it uniform. To determine this torque we can recognize
the excess energy $-M \cdot \nabla^2 M$ as a potential energy in an effective field equal to $\nabla^2 M$,
and it follows that the torque (due to the effective field) is $M \times \nabla^2 M$. Therefore the
torque due to exchange interaction acting on M equal to

$$AM \times \nabla^2 M \tag{5.1}$$

where the stiffness constant A measures the strength of the exchange interaction.

The magnetization is associated with the angular momenta of electrons. If they
are related by a magneto-mechanical ratio γ, the equation of motion equating the local
rate of change of angular momentum to the local torque is

$$\frac{dM}{dt} = \gamma M \times (A\nabla^2 M + H) \tag{5.2}$$

where H is the applied field and other anisotropic effects are neglected.

The solutions we seek are deviations from the uniformly ordered state and we
therefore write

$$M = M_0 + m \tag{5.3}$$

where M_0 is the uniform magnetization. To find the travelling wave solutions for
m having the following form

$$m = m_0 \sin (k \cdot r - \omega t)i + m_0 \cos (k \cdot r - \omega t)j \tag{5.4}$$

where k is a wave vector and i and j are unit vectors in the x and y directions. It is
because if M_0 is in the z-direction, and m is small compared to M_0, m is nearly in the
xy plane. Substitution in the equation of motion above gives directly the dispersion
relation between ω and k, if we put $H = 0$ for the present,

$$\omega = -\gamma A M_0 k^2 = -Dk^2 \tag{5.5}$$

for the spin waves.   Suitable combination of these waves produce the standing wave solutions, of course.

Thus, the magnetization density also obeys the linearized hydrodynamic equation

$$\frac{dM(k,\, t)}{dt} = -Dk^2 M(k,\, t) + F(k,\, t) \tag{5.6}$$

where D is the diffusion coefficient and the random noise $F(k,\, t)$ is delta correlated

$$\langle F(k,\, t)F(k',\, t')\rangle = 2\Gamma k^2 \delta(k - {}'k)\delta(t - t') \tag{5.7}$$

As in the previous section, the correlation strength is

$$\Gamma = D\langle M^2(k)\rangle \tag{5.8}$$

Near the critical point

$$\langle M^2(k)\rangle \approx (k^2 + \xi^2)^{-1} \tag{5.9}$$

where ξ is the correlation length and $\xi \to \infty$ as $T \to T_c$, where T_c is the critical temperature, because near the critical point

$$\xi \approx (T - T_c)^{-1/2} \tag{5.10}$$

Power spectral density is also able to evaluate.   Critical behavior of the dispersion relation is $\omega(k) \sim k^4$ as $T \to T_c$, whereas away from the critical point $\omega(k) \sim k^2$ for small k.   Thus we say that long wave length mode of magnetization fluctuation have very long relaxation times, and refered to as critical slowing down.

References

1) F. Reif,   Fundamentals of Statistical and Thermal Physics (McGraw-Hill) 1965.
2) L. Onsager,   Phys. Rev. **37** (1931) 405; **38** (1931) 2265.
3) A. Einstein,   Investigations on the Theory of Brownian Movement (Methuen) 1926.
4) C. Herring and C. Kittel,   Phys. Rev. **81** (1951) 869.