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Brownian Motion and Nonequilibrium Statistical Mechanics

Ryogo Kubo

This article is a personal reflection of the branch of nonequilibrium statistical mechanics called the linear response theory that has as its heart the fluctuation-dissipation thereom, which states that irreversible processes in nonequilibrium are necessarily related to thermal fluctuations in equilibrium. Its origin lies in the Einstein relation for the diffusion constant and the mobility of a Brownian particle. The short history of the fluctuation-dissipation theorem is described. Then the linear response theory is briefly summarized and the meaning of stochastization is considered. The Langevin equation approach and its extensions are reviewed.

Much of my work is concerned with the nonequilibrium theory of statistical mechanics that aims to establish a theoretical framework to treat a macroscopic system in nonequilibrium dynamic states from a microscopic standpoint. In contrast to equilibrium statistical mechanics, which is the microscopic foundation of thermodynamics, nonequilibrium statistical mechanics is far from complete. The concept of nonequilibrium is perhaps too broad to be unified by a few principles. In spite of this there has been great progress in our understanding of this field in the last few decades.

The nonequilibrium aspect of statistical mechanics is not new, but rather the origin of statistical mechanics more than 100 years ago. In 1872, Ludwig Boltzmann published the Boltzmann equation for gaseous systems (*1*). This equation determines the evolution of the distribution function of gaseous molecules. For equilibrium states it derives the Maxwell-Boltzmann distribution of molecular velocities. For nonequilibrium states, where mass and heat flows are present, it gives the macroscopic laws with the kinetic coefficients (for example, the viscosity coefficient or the heat conductivity) in terms of the intermolecular forces governing collisions between molecules. If the function $f(\mathbf{r}, \mathbf{v}, t)\, d\mathbf{r}d\mathbf{v}$ is the number of molecules to be found at time t in the elementary volume $d\mathbf{r}d\mathbf{v}$ with the spatial coordinate $\mathbf{r}$ and the velocity $\mathbf{v}$, the equation is expressed in the form

$$\frac{\partial}{\partial t} f(\mathbf{r}, \mathbf{v}, t) = -\mathbf{v} \cdot \frac{\partial}{\partial \mathbf{r}} f - \frac{\mathbf{K}}{m} \cdot \frac{\partial}{\partial \mathbf{v}} f + \Gamma(f) \qquad (1)$$

where the first two terms on the right are the drift terms representing the change of f by the free motion of each molecule, $\dot{\mathbf{r}} = \mathbf{v}$, and $\dot{\mathbf{v}} = \mathbf{K}/m$. $\mathbf{K}$ is the force acting on a molecule and m is the molecular mass. The last term $\Gamma(f)$ is the collision term representing the change of f by collisions between molecules. Boltzmann wrote this equation by intuition, making the Stoss-Zahl Ansatz assumption for collisions taking place in a random fashion. This may be justified in the limit of a dilute gas and in the scale of time and space much larger than the mean free time and mean free path. Derivation of the Boltzmann equation and its extensions to denser systems remain outstanding problems even today.

The Boltzmann equations are commonly used not only for gases

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but also, with necessary modifications, for electrons and phonons in solids. This approach in nonequilibrium statistical mechanics is called the kinetic method. It is useful in many problems, but only within its limitation, which is that the particles must be in nearly free motion, with mutual collisions or scattering occurring only occasionally. Because of this limitation, the simple kinetic method cannot be applied to denser systems—say, liquids or amorphous solids. Considerable work has been done to extend the kinetic method to dense systems of interacting particles (*2*). Although such extensions are important, they are usually complicated and unsuitable as a general basis for studying nonequilibrium physical processes.

The Linear Response Theory

A different approach to nonequilibrium statistical mechanics is generally called the linear response theory (*3*). In this approach, one is limited to nonequilibrium states near equilibrium. From the analysis of such states, a general framework is constructed.

In the presence of an electric field, a conductor carries an electric current in proportion to the field strength. If there is a temperature gradient in a system, heat flow is produced in proportion to the gradient. Such linear phenomena are common and fundamental in physics. The physical constants appearing here are called kinetic coefficients, susceptibilities, or, more generally, admittance. They are usually functions of the frequency ω and the wave number k, which characterize the spatial and temporal nonuniformity of the phenomena. Can statistical mechanics give us a general method by which we calculate the kinetic coefficients from a basic knowledge of the microscopic structure and the dynamics of the system of interest? Again, the traditional kinetic method is not quite satisfactory for this purpose.

The linear response theory answers this question, giving an expression for an admittance in terms of the correlation function of fluctuations of physical quantities relevant to the problem under consideration. A good example is the conductivity formula, often called the Kubo formula,

$$\sigma(\omega) = \int_0^\infty dt\, e^{-i\omega t} \langle J(0); J(t)\rangle / V\, k_B T \qquad (2)$$

where σ is the conductivity at the frequency ω of the applied electric field and the bracketed terms inside the integral constitute the correlation function of the electric current, which fluctuates spontaneously in thermal equilibrium without any external driving forces and is regarded as a stationary stochastic process. V is the volume of the conductor under consideration, k_B is the Boltzmann constant, and T is absolute temperature. [See (*3a*) for the definition of the bracket.] Theoretical methods have been developed to calculate such an expression by quantum mechanics. Thus, this approach has proved to be powerful in solving a number of problems in the physics of condensed matter, for example, electrical properties of amorphous semiconductors and the conduction properties of two-dimensional systems, including the topical problem of the quantized Hall effect.

Einstein's Theory of Brownian Motion

This approach differs from Boltzmann's and, in fact, its origin is attributed to Einstein. In 1905 Einstein wrote three papers, one on special relativity (*4*), one on the photoelectric effect (*5*), and the third on Brownian motion (*6*). The last is perhaps the least well known but is as important as the others. Einstein showed that the diffusion constant D and the mobility of Brownian particles, μ, are related to each other in terms of the equation

$$D = \mu\, k_B T \qquad (3)$$

where T is the absolute temperature of the fluid in which Brownian particles are immersed. Einstein came to this conclusion while trying to prove the atomic theory. (It is hard to realize that atomism was not yet established 80 years ago.) Einstein reasoned that if heat is really irregular motion of atoms, the thermal motion of fluid molecules must be transmitted to a particle floating in the fluid. If the particle is large enough, he thought, its motion should be observed directly. Einstein did not know at first that such motion had already been observed by the botanist Robert Brown in 1827 when he was working with small particles originating from pollen floating on water (*7*). A quantitative test of Eq. 3 was crucial, and Einstein himself checked it with existing data on sugar molecules in water. Soon after, Jean Baptiste Perrin successfully observed Brownian motion and confirmed Einstein's theory. This finally convinced even the strongest opponents of the truth of atomism.

The outcome of the Einstein theory was not only the confirmation of atomism. Brownian motion turned out to be an ideal model. From Einstein's theory, mathematical theories of stochastic processes have emerged. Numerous applications have been developed in physics, chemistry, biology, and almost every other discipline of science. In fact the linear response theory is, in a sense, the most natural extension of the Einstein theory of Brownian motion. The heart of the linear response theory is the so-called fluctuation-dissipation theorem (FD theorem).

A Brownian particle is observed to be in incessant zigzag motion. This causes diffusion, which can be observed for a solution of such particles. The diffusion constant D is defined by the equation

$$D = \lim_{t\to\infty} \langle \Delta x(t)^2\rangle / 2t \qquad (4)$$

where $\Delta x(t) = x(t) - x(0)$ is the displacement of a Brownian particle along the x-axis in a time interval t, and the pointed bracket designates the statistical average. The displacement obeys a Gaussian distribution with the variance equal to $2Dt$. This means that

$$D = \ell^2/2\tau = \langle u^2\rangle\, \tau = k_B T\, \tau/m \qquad (5)$$

where ℓ is the mean free path, τ is the mean free time, and u is the velocity. The form of Eq. 5 is due to the equipartition law $m\langle \mu^2\rangle = k_B T$, where m is the mass of a Brownian particle. If the particles are in a force field such as gravity, the zigzag motion is somewhat ordered in the direction of the force K, and a steady motion is induced in this direction. If the average drift velocity is u_d, the average acceleration is u_d/τ, which must be equal to K/m. Therefore the mobility μ is equal to τ/m. Substitution of μ for τ/m in Eq. 5 gives the Einstein relation (Eq. 3). This derivation differs from the original one, but it is instructive. Diffusion is a direct consequence of fluctuations of the velocity of the Brownian particle. The mobility characterizes the response of the particle to a driving force and to the process of dissipation in which the potential energy of the driving force is turned into heat. Thus, fluctuation and dissipation are two aspects of a single phenomenon and thus are necessarily related to each other. This is the general concept of the FD theorem, of which the Einstein relation (Eq. 3) was the first example; Eqs. 3 and 4 give, with the use of $\Delta x(t) = \int_0^t u(t')dt'$, the relation

$$\mu = \frac{1}{k_B T}\int_0^\infty \langle u(0)\, u(t)\rangle dt \qquad (6)$$

in which the mobility is represented by the integrated correlation of the velocity of the Brownian particle. This is almost equivalent to Eq. 1, since the conductivity σ is equal to $e^2 n\mu$ if the density of charge carriers is n and their charge is e.

The FD Theorem

In 1927, Nyquist developed a theorem that an electric resistor produces a spontaneously fluctuating voltage difference between its terminals (*8*). This is called thermal or Nyquist noise. Although it is weak, it can be heard by ear if properly amplified. The power of this noise voltage is on average equal to the impedance multiplied by k_BT, T being the temperature of the resistor. Nyquist derived this theorem by a thermodynamic consideration of detailed balance.

In 1931, Onsager proved the celebrated theorem of reciprocity by proposing the FD theorem (*9*). Consider, for example, a block of solid in contact with a heat bath. At equilibrium, the temperature is almost uniform in space, but thermal motion of atoms necessarily causes fluctuations in temperature distribution and associated heat currents. This is a sort of Brownian motion. Onsager assumed that the average decay of such fluctuations follows the macroscopic law of heat conduction. This assumption led him to an expression of the heat conductivity, in terms of a correlation function of energy flow, similar to Eq. 1. The dynamics of fluctuation is essentially microscopic, however, and has time-reversal symmetry. Onsager proved the reciprocity of the heat conductivity tensor of an anisotropic substance. The same reasoning can be used to prove the reciprocity in a set of kinetic coefficients. Onsager's reciprocity was used later as the basis of nonequilibrium thermodynamics as it was developed in the late 1940's by Prigogine and others (*10*).

In the early 1950's, interest in the FD theorem arose afresh in different parts of the world almost independently. In the United States, Callen and Welton (*11*) made a quantum-mechanical formulation and, in Japan, Takahashi (*12*) made a classical formulation following the Gibbs formalism. Green (*13*) considered a generalized theory of Brownian motion of macrovariables and derived Fokker-Planck equations for their evolution. In doing so, he showed that the kinetic coefficients are expressed as integrated correlations of relevant physical quantities. Such expressions are sometimes referred to as the Green-Kubo formulas. I came across the problem at about the same time, but a full account of my work was not published until 1957 (*14*). Perhaps my derivation of the theorem was the most transparent and elucidated many points that had not been realized. In any event, that remains one of my most frequently cited papers.

I had been interested in the theory of Brownian motion as a statistical problem since I was a physics student. But I approached this problem from a different angle through my theoretical work on semiconductors and nuclear magnetic resonance (NMR). Thus, my first motivation was not abstract. During World War II I worked on some problems in semiconductors, and I came back to them after the war. Electrons trapped by impurities are ionized thermally by absorbing many phonons, a phenomenon now called nonradiative transitions. I formulated a theory by expressing the quantum-mechanical transition probability in terms of a correlation function of a perturbative quantity, in this case a nonadiabatic term arising from the break of the Born-Oppenheimer approximation (*15*). I used this method later to formulate a unified theory of radiative and nonradiative transitions in condensed matter (*16*). This Kubo-Toyozawa theory has been applied to many problems, such as electrochemical processes.

The International Conference on Theoretical Physics in Kyoto in 1953 marked the return of Japanese science to the international science community. It may even be called Japan's first appearance, because before the war Japanese scientists, with a few exceptions, remained almost unrecognized by the West. Fortunately I was able to present a paper together with Tomita on a general theory of NMR absorption (*17*), which was again an application of the correlation function formulation, namely the FD theorem. This work has since been known as the Kubo and Tomita theory. The full paper was published 1 year later (*18*). In the appendix I gave a simple derivation of the basic formula, which was further elaborated in the 1957 paper (*14*). Nuclear magnetic resonance absorption spectra are generally considered the power spectra of fluctuating nuclear magnetic moments, a sort of Brownian motion. Our theory was simply a perturbative treatment of dipolar interactions, which are modulated by atomic motion or exchange interactions, and was an extension of the work of Bloembergen, Purcell, and Pound (*19*).

A Brief Summary of the Linear Response Theory

My 1957 paper (*14*) may be summarized as follows. Suppose a system in thermal equilibrium is exposed to an external force $F(t)$. The effect of the force on the system is represented by an additional term $H' = -AF(t)$ of the Hamiltonian, where A is the quantity conjugate to the force. We observe a physical quantity B of the system. If F is a short pulse of unit strength exerted on the system at $t = 0$, the response observed as the induced change of B at a later time t is called the response function $\phi_{BA}(t)$. It is given by

$$\phi_{BA}(t) = \langle [A(0), B(t)] \rangle \qquad (7)$$

where the notation (X, Y) means the Poisson bracket. $X(t) = X(p_t, q_t)$ denotes the temporal evolution of a dynamic quantity X as the system changes its dynamic states following the law of microscopic dynamics, either classical or quantum-mechanical, in the absence of the external force. In quantum mechanics the Poisson bracket is equal to a commutator, namely, $(X, Y) = [X, Y]/i\hbar$, where $\hbar$ is the Planck constant divided by 2Π and i is the imaginary unit.

If the force $F(t)$ is in action continuously from the infinite past to the present, the effect is represented by

$$\overline{B(t)} = \int_{-\theta}^{t} \phi_{BA}(t - t')\, F(t')dt' \qquad (8)$$

as the linear response.

If the system is near equilibrium at temperature T, the response function (Eq. 7) can be transformed to a correlation function of the form

$$\phi_{BA}(t) = \frac{1}{k_BT} \langle \dot{A}(0); B(t) \rangle \qquad (9)$$

where $\dot{A}$ is the time derivative of A. For example, if we observe the velocity of a Brownian particle under the action of a force, we set $B = \mu$, $A = x$, and $\dot{A} = \dot{x} = \mu$. Then Eq. 9 gives Eq. 6. Equation 9 is a correlation function with the usual meaning in the classical cases, but it contains some necessary complications in the quantal cases.

A general admittance at the finite frequency ω is given in the form

$$\chi(\omega) = \int_0^{\infty} e^{-i\omega t}\, \phi_{BA}(t)dt \qquad (10)$$

This is a complex function. Either its real or its imaginary part corresponds to the dissipative response, for example, the resonance absorption. It is then shown to be the power spectrum of fluctuation of a proper physical quantity. This is the FD theorem. Note, however, that Eq. 10 contains the nondissipative part also expressed in terms of correlation functions. In this sense a better name for the theorem would be the fluctuation-response theorem. The Heisenberg-Kramers dispersion formula, Onsager's reciprocity, and some other basic laws follow from this theorem.

The linear response theory gave only the expressions for the response function and the admittance. It is difficult to calculate such an expression for a many-body system in which a great number of particles are interacting with each other. It is important, however, that the linear response theory give the exact expression to start with. In some cases, one cannot really write any Boltzmann equation. One example is electronic conduction in a strong magnetic field (*20*). The calculation of response functions for many-body systems has been greatly facilitated by developments of Green's function method, which has become the basic method of modern quantum statistical mechanics (*21*). It was perhaps not a mere coincidence that this method was so rapidly developed just after the appearance of the linear response theory.

Coarse-Graining and Stochastization

As the name indicates, statistical mechanics uses some probabilistic assumptions. Obviously, the probabilistic elements come in with some sort of coarse-graining (that is, when observations are made cruder). Microscopically, a physical system consists of a large number of particles, say 10^{23} molecules in a drop of water. In a macroscopic observation, one is interested only in a small number of physical variables that are called, for brevity, the gross variables. So information is reduced enormously, which necessarily requires a probabilistic description. It is extremely difficult, however, to determine the a priori probabilities from the first principle. In equilibrium statistical mechanics, the fundamental postulate is the principle of equal weight, which means that every microscopic state has equal a priori probability in equilibrium. Boltzmann introduced the ergode hypothesis to make this plausible. Efforts have been made to support this hypothesis, but it still remains an outstanding problem, even though the validity of the principle of equal weight and of equilibrium statistical mechanics are not in doubt.

In nonequilibrium cases, the probabilistic description must be stochastic, that is, it must describe the temporal evolution of the probability for the gross variables that adequately define the coarse-grained states of the system under observation. Switching from microscopic dynamics to a stochastic description may be called stochastization. The Boltzmann equation is the oldest example of stochastization. The gross variables there are the distribution function $f(\mathbf{r}, \mathbf{v}, t)$. For a Brownian motion, the gross variables are the space coordinates and the velocities of a Brownian particle. If the velocities are not observed, the gross variables are coarse-grained to the space coordinates. In general, there are different successive stages of coarse-graining. For a gas, the hydrodynamic equations, which can be derived from the Boltzmann equation, are the crudest description.

Mathematical theories of stochastic processes have been developed greatly in this century. Much has been learned about Markovian and Gaussian processes. An ideal Brownian motion is a good example of the Gaussian-Markovian process. In a Markovian process, the evolution of the probability in the next instant is determined by its state at present. This is satisfactory from a physicist's point of view. It is, so to say, second best if a deterministic description is not possible. A Boltzmann equation describes a Markovian evolution.

It is a general rule of stochastization that merely reducing the variables is not enough to obtain a simple and clear description such as a Markovian description. One must simultaneously make the scales of time and space cruder. For example, a description of Brownian motion as diffusion is possible only for a time scale larger than the mean free time (τ) and a spatial scale larger than the mean free path (ℓ). However, physics is not always limited to that crudeness; when it is not, one can no longer enjoy the elegance of Markovian theory. In these circumstances, one uses the linear response theory approach in which the stochastization, if it is ever made, is performed at a later stage. If the system is simple enough, stochastization is unnecessary, but equations of the linear response theory still work.

Equation 7 was derived with the use of a simple perturbation expansion. In this respect, the linear response theory was once criticized because the phase-space trajectories of a dynamic system are generally unstable against perturbation (*22*). My view is that this instability instead favors the stability of distribution functions, working as the cause of mixing. I used perturbative calculations for a family of smooth distribution functions in the phase space or the space of density matrices, not for each of the phase-space trajectories. Rigorous verification of this assertion, however, is very difficult.

Langevin Equations and Their Generalizations

One standard method for treating Brownian motion is the use of Langevin equations. For example, the equation of motion for a Brownian particle can be written as

$$m\dot{u}(t) = -m\gamma u(t) + R(t) + K \qquad (11)$$

where $m\gamma$ is the friction constant, and R represents a random force. $F = -m\gamma u + R$ is the force coming from the molecules in the surrounding fluid. This is divided into a systematic friction part and a random part. The FD theorem, in the form of the Nyquist theorem, shows that the friction constant is essentially the integrated correlation of the random force, namely

$$m\gamma = \frac{1}{k_B T}\int_0^\infty \langle R(0)R(t)\rangle dt \qquad (12)$$

For an ideal Brownian motion, $R(t)$ is assumed to be a white Gaussian noise, that is, its correlation time is infinitely short. This is consistent with the assumed form of friction, which is that the force acts on the particle without delay; however, this idealization may not be adequate for describing realistic physics. For example, electrical resistance usually depends on frequency. To include such cases the Langevin equation has to be generalized to

$$m\dot{u}(t) = -m\int_{-\infty}^{t} \gamma(t-t')\, u(t')dt' + R(t) + K(t) \qquad (13)$$

namely, to a retarded friction (*3a*). The FD theorem then shows that the kernel of retardation is essentially the correlation function of the random force $R(t)$; in such cases R is a colored (nonwhite) noise. Mori showed that a microscopic equation of motion can be cast into a form of Eq. 13 (*23*).

A Langevin equation is a stochastic equation, which describes a stochastic process generated by an underlying stochastic process. By Eq. 12 the random force $R(t)$ drives the process $u(t)$, and the equation $\dot{x}(t) = u(t)$ generates the process $x(t)$. In the procedure of successive coarse-graining, it is sometimes useful to take this point of view. In general, the stochastic equation can be nonlinear. Such is the case for the Brownian motion of the resultant spin of a spin system that is modeled by a random local field. After this issue was treated by Anderson and Weiss (*24*), I showed that the problem is generally formulated in a solvable way if the driving process is Markovian (*25*). Since the driving process is merely assumed as a model, this approach is necessarily phenomenological, but it is useful for obtaining some insight into the physics of a complex process. Interesting applications were made to relaxation and resonance phenomena of spins in zero or low external fields (*26*), to second order optical processes, and to other problems (*27*).

Much more remains to be discussed and to be done on the generalization of the concept of Brownian motion and nonequilibrium statistical mechanics. For example, interest in the Brownian motion of quantum systems which are sometimes called dissipative quantum systems seems to have revived in recent years in connection with supercurrents in superconductors, although a large amount of work on this problem has been done.

I did not intend here to give a comprehensive review of the whole subject, but have tried to describe how I understand the problem and how I have worked on this subject. In this sense this essay is a personal reflection.

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Evolution of Meson Science in Japan

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Forty years after Yukawa predicted the existence of mesons, experimental research activities with the use of mesons were started in Japan. Particles of the "second generation," which have nothing to do with the structure of ordinary materials, such as muons, K mesons, and other exotic particles have been exploited as unique probes to study new constituents of matter.

About 50 years ago Yukawa predicted the existence of a new particle mediating the nuclear force (*1*). Nowadays, this particle, called the π meson (or pion), is produced abundantly by means of high-energy accelerators. Long before pions were identified, another particle had been discovered unexpectedly in cosmic rays. That particle was called the μ meson and is now called the muon (μ^+ and μ^-), which is believed to have the same properties as the electron except for mass (207 times heavier). Muons and muon neutrinos (ν_μ, $\bar{\nu}_\mu$) are abundantly produced from the decay of charged pions: $\pi^+ \rightarrow \mu^+ + \nu_\mu$, $\pi^- \rightarrow \mu^- + \bar{\nu}_\mu$.

Because of the maximum violation of parity in the weak interaction, the muon from the pion decay is nearly 100% polarized in the pion rest frame. Furthermore, the muon decays with a 2.2-μsec lifetime by weak interaction as $\mu^+ \rightarrow e^+ + \bar{\nu}_\mu + \nu_e$, $\mu^- \rightarrow e^- + \nu_\mu + \bar{\nu}_e$, where ν_e and $\bar{\nu}_e$ are electron neutrinos, and the positron (e^+) or electron (e^-) thus produced is very asymmetrical with respect to the muon spin. This basic property permitted the study of the behavior of muon spin in matter, and this aspect of study has been growing under the name of μSR (muon-spin rotation, relaxation, and resonance) (*2*). Positive muons behave like "light protons" and probe internal fields at interstitial sites of crystals, where they are located. They hop from one site to another, and their diffusive motion shows strong quantum effects because of the small mass of the muons (*3, 4*). Negative muons are used to create atoms of pseudonuclear charge $(Z - 1)e$ and to probe electron spin densities outside the nuclear region (*5*).

Because of the unique masses and interactions of pions and muons, they constitute a rich arena of exotic objects for scientists. This study is called meson science. Around 1975 so-called meson factories with high-intensity proton accelerators were launched at Los Alamos (LAMPF), Vancouver (TRIUMF), and Zurich (SIN), and a number of interdisciplinary research programs were initiated to study not only particle and nuclear physics but also atomic and solid-state physics, chemistry, and biomedical applications.

Birth of a Pulsed-Meson Facility in Japan

Although, theoretically, mesons were discovered in Japan, this country used to be very much behind other nations in the development of medium- and high-energy physics. In 1975, however, with the birth of the National Laboratory for High Energy Physics (so-

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