lowance has been made for the effect of the corneal curvature on the image size, but the possible effect of diffusion on the values has not yet been estimated. The dispersion of the values taken in succession is considerable (range $\pm$ 20%), which may be explained by the fact that the number of slit images covering the small vesicle is low (4–5). The precision of the mean flow value may be increased by decreasing the distance between successive sections or by taking a greater number of consecutive determinations.

There does not seem to be any hindrance to applying the method to other species besides rabbits[2].

Zusammenfassung. Wird der Vorkammerinhalt des Auges von aussen mit Fluorescin gefärbt, so ist das aus der Pupille herausfliessende Kammerwasser kurzfristig als ungefärbte «Blase», deren Volumen photogrammetrisch bestimmbar ist, sichtbar. Das Minutenvolumen ist durch zwei zeitlich getrennte Messungen feststellbar.

O. HOLM and C. E. T. KRAKAU

Department of Experimental Ophthalmology, University Eye Clinic, Lund (Sweden), August 19, 1966.

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THEORIA

A Quantum Theory of Brownian Movement

It is believed that Brownian movement is a fundamental type of motion of matter at a molecular level. Although such a motion cannot be found at a submolecular level, in which it is governed strictly by the mechanical laws of motion without irreversibility, Brownian particle-like motion may appear as a result of space-time correlation of irregular fluctuating forces. According to the theorem of fluctuation-dissipation, there may occur dissipation where there is fluctuation. Furthermore, it must be considered that the fluctuation comes from some generalized Brownian motion of a moving body in the system. The investigation of Brownian motion will be important in the boundary field between the world of microscopic moving bodies and that of macroscopic moving particles, in order to seek the essential cause of irreversibility which is a characteristic property in the macroscopic world. Regarding the theory of Brownian movement[1], it is well known that there are two kinds of approach: one is Einstein's and the other is Langevin's. Here, we will deal with the problem along the line of Langevin's approach from the statistical mechanical point of view.

In the present article, the problem is to obtain a generalized form of Langevin's equation of motion, not on the ordinary stochastic basis but in the systematic way, from the quantum mechanical equation of motion in the Heisenberg picture, and to express the relation of Einstein-Nernst in a general sense.

The operator of an arbitrary observable A changes temporally according to the equation of motion

$$dA/dt = i\,\hbar^{-1}[H, A] \equiv i\,\omega^*\,A\,, \qquad (1)$$

whose solution is given by

$$A(t) = \exp(i\,t\,H/\hbar)\,A(0)\exp(-\,i\,t\,H/\hbar) \equiv \exp(i\,t\,\omega^*)\,A(0)\,, \qquad (2)$$

where H is the Hamiltonian of the system, $\hbar$ is Planck's constant divided $2\,\pi$, and ω^* is defined by Eq. (1) as an operator of angular frequency of transition whose classical analogue is Liouville's operator.

By making use of a projection operator P, the operator of A can be split identically into $\overline{A} \equiv P\,A$, projected onto a subspace of the operator Hilbert-space, and $A' \equiv (1-P)\,A$, projected onto an orthogonal complement of the P-subspace:

$$A = P\,A + (1-P)\,A = \overline{A} + A'\,. \qquad (3)$$

According to ZWANZIG's technique[2], we obtain the following coupled equations from Eq. (1):

$$d\overline{A}/dt = i\,P\,\omega^*\,(\overline{A} + A')\,, \qquad (4a)$$

$$dA'/dt = i\,(1-P)\,\omega^*\,(\overline{A} + A')\,. \qquad (4b)$$

By means of the Laplace transformation of

$$A(s) = \int_0^\infty \exp(-\,s\,t)\,A(t)\,dt, \qquad \text{(real part of } s \text{ being positive)},$$

we can deal with Eqs. (4a and b) in an algebraic manner. These are transformed into

$$s\,\overline{A}(s)\overline{A} - (0) = i\,P\,\omega^*\,\{\overline{A}(s) + A'(s)\}\,, \qquad (5a)$$

$$s\,A'(s) - A'(0) = i\,(1-P)\,\omega^*\,\{\overline{A}(s) + A'(s)\}\,, \qquad (5b)$$

where $\overline{A}(s)$ and $A'(s)$ are the Laplace transforms of $\overline{A}(t)$ and $A'(t)$, respectively. At first, from Eq. (5b), we obtain

$$A'(s) = [s - i\,(1-P)\,\omega^*]^{-1}\cdot A'(0) + [s - i\,(1-P)\,\omega^*]^{-1}\cdot i\,(1-P)\,\omega^*\,\overline{A}(s)\,. \qquad (6)$$

Inserting this into Eq. (5a), we obtain without approximation a unified equation of

$$s\,\overline{A}(s) - \overline{A}(0) = i\,P\,\omega^*\,\overline{A}(s) + i\,P\,\omega^* \times [s - i\,(1-P)\,\omega^*]^{-1}\cdot i\,(1-P)\,\omega^*\,\overline{A}(s) + i\,P\,\omega^* \times [s - i\,(1\,\%\,P)\,\omega^*]^{-1}\cdot A'(0)\,. \qquad (7)$$

[1] R. EISENSCHITZ, *Statistical Theory of Irreversible Processes* (Oxford University Press, London 1958).

[2] R. ZWANZIG, J. chem. Phys. *33*, 1338 (1960).

It may be understood easily by means of the inverse Laplace transformation that Eq. (7) is the Laplace transform of

$$d\bar{A}(t)/dt = i P \omega^* \bar{A}(t) + \int_0^t dt' \, i P \omega^* \times \text{oxp}\{i t' (1 - P) \omega\} \cdot i (1 - P) \omega^* \bar{A} (t - t') + i P \omega^* \cdot \exp\{i t (1 - P) \omega^*\} \cdot A'(0). \quad (8)$$

Now the following form is taken for the projection operator:

$$P A(t) = (Tr\, \varrho\, A^+ A)^{-1} \cdot \{Tr\, \varrho\, A^+(0) A(t)\} A(0) = \Xi(t) A(0), \quad (9)$$

where

$$\Xi(t) \equiv (Tr\, \varrho\, A^+ A)^{-1} \cdot \{Tr\, \varrho\, A^+(0) A(t)\}. \quad (9')$$

In this case, it is noted that P fulfils the property of

$$P A(0) = \bar{A}(0) = A(0), \quad \text{or} \quad (1 - P) A(0) = 0. \quad (10)$$

Thus Eqs. (7) and (8) become

$$s \bar{A}(s) - A(0) = \{i P \omega^* + i P \omega^* \times [s - i (1 - P) \omega^*]^{-1} i (1 - P) \omega^*\} \bar{A}(s), \quad (11)$$

and

$$d\bar{A}(t)/dt = i P \omega^* \bar{A}(t) + \int_0^t dt' \, i P \omega^* \times \exp\{i t' (1 - P) \omega^*\} \cdot i (1 - P) \omega^* \bar{A} (t - t'), \quad (12)$$

respectively.

By making use of the Laplace transform of $\Xi(t)$, i.e.,

$$\Xi(s) = \int_0^\infty dt \exp(-s t) \, \Xi(t),$$

Eq. (9) leads to

$$\bar{A}(s) = \Xi(s) A(0). \quad (13)$$

From Eq. (11), we obtain

$$s \Xi(s) - 1 = \{i P \omega^* - \zeta(s)\} \Xi(s),$$

or

$$\Xi(s) = \{s - i P \omega^* + \zeta(s)\}^{1-}. \quad (14)$$

Here, we have defined

$$\zeta(s) \equiv - i P \omega^* \cdot [s - i (1 - P) \omega^*]^{-1} \cdot i (1 - P) \omega^*. \quad (15)$$

Because we obtained the Laplace transform of the projected operator, $\bar{A}(s)$, from Eqs. (13) and (14), we will attempt to recover the Laplace transform of the original total operator, $A(s)$, after the complement with the part of orthogonally complementary operator, $A'(s)$. From Eq. (6) with Eq. (10), we obtain

$$A'(s) = \Xi(s) \cdot [s - i (1 - P) \omega^*]^{-1} i (1 - P) \omega^* A(0). \quad (16)$$

Here it must be mentioned that this equation has been derived with the help of a commutability of

$$i [(1 - P) \omega^*, \Xi(s)] = 0. \quad (17)$$

Hence, by adding Eq. (13) to Eq. (16), we recover the required quantity of

$$A(s) = \bar{A}(s) + A'(s) = \Xi(s) \cdot \{1 + [s - i (1 - P) \omega^*]^{-1} \times i (1 - P) \omega^*\} A(0). \quad (18)$$

By rewriting Eq. (18) with Eq. (14), the following equation can be obtained:

$$s A(s) - A(0) = i P \omega^* A(s) - \zeta(s) A(s) + \mathfrak{F}(s), \quad (19)$$

whose inverse transform is

$$dA(t)/dt = i P \omega^* A(t) - \int_0^t dt' \, \zeta (t - t') A(t') + \mathfrak{F}(t), \quad (20)$$

where

$$\mathfrak{F}(s) = [s - i (1 - P) \omega^*)^{-1} \cdot \mathfrak{F}(0), \quad (21)$$

$$\mathfrak{F}(t) = \exp\{i t (1 - P) \omega^*\} \mathfrak{F}(0), \quad (21')$$

$$\mathfrak{F}(0) = i (1 - P) \omega^* A(0) = (1 - P) \dot{A}(0), \quad (21'')$$

By making operation of $(1 - P)$ to the both sides of Eq. (21″) and taking account of the idempotent property of $[1 - P)^2 = (1 - P)$, it follows that

$$\mathfrak{F}(0) = i \omega^* A(0) = \dot{A}(0). \quad (22)$$

Recently, MORI[3] derived an equation quite similar to Eq. (20) in an alternative way, which is called a quantum analogue of Langevin equation. It will be ascertained that the above equation of Eq. (20) is also equivalent to that, but the present derivation is more systematic than MORI's method.

In order to execute this project, by taking account of

$$[s - i (1 - P) \omega^*]^{-1} \cdot i (1 - P) \omega^* = \mathfrak{F}(s) \cdot A(0)^{-1},$$

we obtain from Eq. (15)

$$\zeta(s) = - i P \omega^* \mathfrak{F}(s) \cdot A(0)^{-1} = - (Tr\, \varrho\, A^+ A)^{-1} \cdot \{Tr\, \varrho\, A^+(0)\, i\, \omega^* \mathfrak{F}(s)\} = (Tr\, \varrho\, A^+ A)^{-1} \cdot \{Tr\, \varrho\, \dot{A}^+(0) \mathfrak{F}(s)\} = (Tr\, \varrho\, A^+ A)^{-1} \cdot \{Tr\, \varrho\, \mathfrak{F}(0) \mathfrak{F}(s)\}, \quad (23)$$

or

$$\zeta(t) = (Tr\, \varrho\, A^+ A)^{-1} \cdot \{Tr\, \varrho\, \mathfrak{F}(0) \mathfrak{F}(t)\}. \quad (24)$$

It has been shown that $\zeta(s)$ or $\zeta(t)$ is a sort of friction coefficient which is given by the time-correlation function of $\mathfrak{F}$, and $\mathfrak{F}(t)$ is called correctly the fluctuating term with respect to the temporal change of an observable A. In other words, Eq. (19) or (20) is a quantum analogue of Langevin equation. Here we would remark that, in spite of $\mathfrak{F}(t) \neq i \omega^* A(t) = \dot{A}(t)$, it may be shown to be approximately $\mathfrak{H}(t) \cong \dot{A}(t)$, since

$$\mathfrak{F}(t) = \exp\{i t (1 - P) \omega^*\} \cdot (1 - P) \dot{A}(0) \cong (1 - P) \exp(i t \omega^*) \dot{A}(0) + O(P \dot{A}).$$

Thereby, we obtain to this approximation the relation of

$$\zeta(t) \cong (Tr\, \varrho\, A^+ A)^{-1} \cdot \{Tr\, \varrho\, \dot{A}^+(0) \dot{A}(t)\}, \quad (24')$$

which agrees with the expression of this quantity in the well-known KUBO formalism of the conductivity theory[4].

Because Eq. (12) can be written in the form of

$$d\bar{A}(t)/dt = i P \omega^* \bar{A}(t) - \int_0^t dt' \, \zeta (t - t') \bar{A}(t'), \quad (25)$$

[3] H. MORI, Prog. theor. Phys. Kyoto 33, 423 (1965).
[4] R. KUBO, J. phys. Soc. Japan 12, 570 (1957).

with $\bar{A}(0) = A(0)$, in comparison with Eq. (20), it may be understood that a sort of Langevin equation including fluctuating term appears after removing the projection operator in Eq. (25) to furnish with a kind of causality.

Finally, it may be shown that a generalized relationship of Einstein and Nernst is established as an Abel limit of Eq. (14) with Eq. (15).

In the limit of $s \rightarrow +0$ of Eq. (14), the following equation is obtained:

$$\lim_{s \rightarrow +0} \int_0^\infty dt \exp(-s\, t)\, \{Tr\, \varrho\, A^+(0)\, A(t)\}$$

$$= \int_0^\infty dt \exp(-t/\tau)\, \{Tr\, \varrho\, A^+(0)\, A(0)\}, \quad (26)$$

where

$$\tau = \{-i\, P\, \omega^* + \zeta_{s\,t}\}^{-1}$$

$\zeta_{s\,t}$ is nothing but a friction coefficient in a stationary Markoffian process, and

$$\zeta_{s\,t} = \lim_{s \rightarrow +0} \int_0^\infty dt \exp\,(-s\, t)\{Tr\, \varrho\, \mathfrak{F}^+(0)\, \mathfrak{F}(t)\} \times (Tr\, \varrho A^+\, A)^{-1}$$

Because of

$$-i\, P\, \omega^* = (Tr\, \varrho\, A^+\, A)^{-1} \cdot \{Tr\, \varrho\, \dot{A}^+(0)\, A(0)\},$$

Eq. (26) becomes a generalized Einstein-Nernst relation of

$$(Tr\, \varrho\, A^+\, A)/\tau = Tr\, \varrho\, \mathfrak{F}^+(0)\, A(0)$$

$$+ \lim_{s \rightarrow +0} \int_0^\infty dt \exp(-s\, t)\{Tr\, \varrho\, \mathfrak{F}^+(0)\, \mathfrak{F}(t)\}. \quad (27)$$

For example, according to Langevin's formalism, the equation of motion of a Brownian particle, whose mass is m, the momentum p, is written as

$$dp/dt = -\zeta(p/m) + \mathfrak{F}(t),$$

where the property of dissipation is due to the first term on the right-hand side, and the second term is a fluctuating force. Because of the momentum relaxation time $\tau_p = m/\zeta$, the Einstein-Nernst relation leads to

$$(Tr\, \varrho\, p^2)/\tau_p = m\, k\, T/\tau_p$$

$$= \lim_{s \rightarrow 0} \int_0^\infty dt \exp(-s\, t)\{Tr\, \varrho\, \mathfrak{F}(0)\, \mathfrak{F}(t)\},$$

which is usually written as

$$m/\tau_p = \zeta = (1/k\, T) \int_0^\infty dt\{Tr\, \varrho\, \mathfrak{F}(0)\, \mathfrak{F}(t)\}.$$

Here, it is evident that the phase-space distribution function in equilibrium has been used in place of quantum density matrix, and also an integral referring to the elementary volume of the phase space was taken instead of a trace of a matrix, Tr.

In this article, we have derived several equations having wide ranges of validity and applicability, from Heisenberg's equation of motion in quantum mechanics with Zwanzig's technique of projection operator[5].

Zusammenfassung. Die Gleichung von Langevin wurde mit Hilfe der Zwanzigschen Methode von Projektionsoperator aus der quantenmechanischen Bewegungsgleichung von Heisenberg geführt, die allgemein als Grundgleichung der Brownschen Bewegung gilt. Zum Derivieren der Gleichung, ist es notwendig, eine Definition für Projektionsoperator P mit einem statistischen Operator ϱ aufzunehmen, das ist:

$$P\, A(t) \equiv (A(0),\ \varrho\, A(0))^{-1} \cdot (A(0),\, \varrho\, A(t))\, A(0)$$

$$= \{\mathrm{Spur}\, \varrho\, A^+(0)\, A(0)\}^{-1} \cdot \{\mathrm{Spur}\, \varrho\, A^+(0)\, A(t)\}\, A(0)$$

$$= \Xi(t)\, A(0),$$

wobei (A', A'') ein inneres Produkt zwischen zwei Vektoren A' und A'' ist. Die Zeitveränderlichkeit einer Observablen A im Heisenberg-Bild gegeben ist:

$$A(t) = \exp(i\, t\, H/\hbar)\, A(0) \exp(-i\, t\, H/\hbar); \text{ mit } \hbar = h/2\,\pi,$$

$$\Xi(t) = \{\mathrm{Spur}\, \varrho\, A^+(0)\, A(0)\}^{-1} \cdot \{\mathrm{Spur}\, \varrho\, A^+(0)\, A(t)\},$$

A^+ zeigt den adjungierten Operator von A, und

$$(i/\hbar)\, [(1-P)\, H,\ \Xi(t)] = 0.$$

Hierauf $(1 - P)$ ist der orthogonalkomplementale Projektionsoperator zu P.

N. TAKEYAMA[6]

Department of Applied Chemistry, Faculty of Engineering, Kyushu University, Fukuoka (Japan), May 22, 1966.

[5] The main contents of this article were published in Japanese in Bussei Kenkyu *5*, 275 (1966).

[6] The author would like to express his sincere thanks to Professor T. SEIYAMA for his encouragement.

CORRIGENDA

C. PARADISI: *Mitochondrial Changes Induced by Diphtheria Toxin in Chicken Embryo Heart Cell Cultures*, Experientia *22*, fasc. 6, p. 373 (1966). On page 373, line 15 reads correctly as follows: 'This toxin induces a clear swelling in isolated mitochondria'. The author points out that Dr. Kadis has never stated or implied that murine plaque toxin causes uncoupling of oxidative phosphorylation.

S. GELFAND and A. GANZ: *Further Observations of the Angiotensin Vasopressive Effect in Rabbits*, Experientia *22*, fasc. 7, p. 478 (1966). The author has noted that the dosage of angiotensin should read **μg** instead of mg.