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ON THE THEORY OF IRREVERSIBLE PROCESSES IN NONSTATIONARY SYSTEMS

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The formalism of Kubo[3] developed for the analysis of irreversible processes in systems close to thermodynamic equilibrium is generalized to the case of essentially nonequilibrium periodically-nonstationary systems. General expressions are obtained for the response function and for the admittance of such systems and their fundamental properties are investigated. In particular, the generalization is obtained of the symmetry relations for the Onsager kinetic coefficients to the case of periodically-nonstationary systems. A possible generalization of the fluctuation-dissipation theorem as applied to classical nonstationary systems is also considered. The results obtained have direct relation to the problem of obtaining and using nonequilibrium systems for the amplification and generation of electromagnetic oscillations.

1. INTRODUCTION

THE general quantum theory of irreversible processes in stationary systems has by now attained a level of development marked by considerable success (cf., for example, [1]). The stationary nature of the system is manifested in the fact that its Hamiltonian H_0 (in the absence of a possible external variable perturbation acting on the system the response to which is of interest) is independent of the time, while the statistical operator ρ_0 describing the state of the system has the form $\rho_0 = C \times \exp(-\beta H_0)$ (it is assumed that the system is in thermal contact with a heat bath).

In the papers of Callen et al[2] and of Kubo[3] quite general expressions were obtained for the linear response functions (or the aftereffect) of the system to an external perturbation variable in time. It was shown that the complex admittance matrix (the Fourier transform of the response functions) satisfies symmetry relations which should be regarded as the generalization of the symmetry principle for Onsager's kinetic coefficients.[4,5] Moreover, it was shown that the matrix for the admittance of the system can in the general case be related to the matrix for the spectral intensity of fluctuations in a state of thermodynamic equilibrium of appropriate (corresponding to the chosen admittance) physical quantities (the fluctuation-dissipation theorem).

At the same time, while the successes noted above have been achieved with stationary systems, there exists no general theoretical development with reference to irreversible processes in nonstationary systems. We take the latter to be such systems in thermal contact with a heat bath whose Hamiltonian explicitly depends on the time: $H_0 = H_0(t)$. The dependence of H_0 on the time physically means that the system is situated in an external variable field which is treated classically.

In the present paper we give a generalization of the formalism of Kubo[3] to the case of periodically-nonstationary systems, i.e., systems which are situated in a periodic external field $[H_0(t) = H_0(t+T)]$. The results of this paper in addition to being of general physical interest are also of direct interest for the problem of obtaining and utilizing nonequilibrium media for the purposes of amplification and generation of electromagnetic oscillations ("the problem of quantum and parametric amplifiers and generators").

In all those cases when the deviation from equilibrium of the medium utilized in quantum or parametric amplifiers is due to the action on this medium of a strong external alternating field (for example, in the so-called three-level quantum amplifier or in the optical parametric amplifier proposed by Akhmanov and Khokhlov[6]) from the point of view of macroscopic electrodynamics we are dealing with electromagnetic processes occurring in a medium with properties which are variable in time.[1)] Of fundamental practical interest in this connection is the case of a periodic external

[1)]From this point of view it is, in general, not meaningful to subdivide such systems (media) into "quantum amplifiers" and "paramagnetic amplifiers" (as has been the custom until now). This has already been pointed out earlier[7].

perturbation (monochromatic "pumping").

The connection referred to above of the topics discussed in this paper with the problem of quantum and parametric amplifiers [2] consists of the fact that in this article we have essentially obtained a general form of the material equation for periodically-nonstationary media and have investigated its fundamental properties.[3] In particular, a generalization of Onsager's symmetry relations has been obtained. At the end of this paper some problems are discussed dealing with fluctuations in classical nonstationary systems.

2. RESPONSE AND ADMITTANCE OF A NONSTATIONARY SYSTEM

We consider a macroscopic system described by the Hamiltonian $H_0(t) = H_0(t+T)$ ($T = 2\pi/\Omega$) which is in thermal contact with a heat bath. In deriving the symmetry relations (Sec. 3) we shall take into account the fact that the dependence of H_0 on the time is due to an external variable electromagnetic field, and that, therefore, the time t appears in H_0 only through the vector potential $\mathbf{A}(t)$ of this field:[4] $H_0 = H_0(\mathbf{A}(t))$. For the purposes of the present section this fact is not essential.

We first of all establish the general properties of the statistical $\rho_0(t)$ which describes the state of the system. By definition

$$\rho_0(t) = \mathrm{Sp}_1\, \rho(t), \tag{2.1}$$

where $\rho(t)$ is the statistical operator of the "large system," i.e., of the system under consideration and of the heat bath with which it interacts. The subscript 1 carried by the trace symbol denotes that the operation Sp is performed only with respect to the variables which refer to the heat bath. The operator $\rho(t)$ satisfies the Schrödinger equation:

$$i\hbar\dot{\rho} = [H_0(t) + H_1 + \mu V, \rho], \tag{2.2}$$

where H_1 is the Hamiltonian of the heat bath, while μV is the interaction Hamiltonian. We assume that the operators H_1 and V do not explicitly depend on the time.[5]

Further, we assume that in the case of a periodic Hamiltonian $H_0(t)$ the operator $\rho(t)$ is also periodic: $\rho(t) = \rho(t+T)$. This condition means that there is no accumulation with time of information (positive or negative) with respect to the "large system" and in the case of a sufficiently large number of particles in the heat bath this condition can, evidently, always be considered to be satisfied. In accordance with (2.1) the operator $\rho_0(t)$ will also have the same property of periodicity, and this will hold independently of the magnitude of the interaction μV. The magnitude of this interaction can affect only the rate of establishment in the system of the periodic state $\rho_0(t)$.

Since we assume that the external perturbation has been applied to the system at $t = -\infty$, and that, therefore, we do not discuss transient processes, then we have at all times for arbitrarily small μ

$$\rho_0(t) = \rho_0(t+T). \tag{2.3}$$

For a macroscopic system the steady state operator $\rho_0(t)$ which satisfies condition (2.3) is fundamentally determined by the properties of the system itself, and its dependence on the interaction with the heat bath is negligible. Therefore, with the same justification which holds in the case of the establishment of Gibbs' canonical distribution for stationary (equilibrium) systems we shall assume that the periodically-nonstationary system under consideration is described by the limiting (for $\mu \to 0$) expression for the statistical operator $\rho_0(t)$, which also satisfies condition (2.3). The role played in this case by the transition to the limit consists of selecting the correct "zero order approximation" to the exact expression for the operator $\rho_0(t)$.[6]

The "zero order approximation" itself (the

[2]In speaking only of amplifiers we wish to emphasize that we are dealing only with the properties of a nonstationary system with respect to small perturbations (the domain of linear electrodynamics of nonstationary media).

[3]Some other properties of such media have been investigated earlier and by means of a different approach in a number of articles[8-11].

[4]We adopt that gauge for the potentials for which the scalar potential $\varphi = 0$.

[5]Physically this means that the external variable field A(t) is strong with respect to the system H_0, but is weak with respect to the heat bath H_1 (i.e., it does not lead to significant deviations from the equilibrium state of the latter). It is just such situations that are of principal practical interest.

[6]A similar situation exists in the case of stationary systems for which we must take the interaction with the heat bath into account only in order to obtain in the "zero-order approximation" $\rho_0 = C\exp(-\beta H_0)$ the correct value of the constant $\beta = 1/kT$. We note that the relevant mathematical problem of finding the "zero-order approximation" to the periodic solution of a system of nonlinear differential equations which are close to linear equations with periodic coefficients has been discussed by Mandel'shtam[12].

limiting operator $\rho_0(t)$) must satisfy the Schrödinger equation

$$i\hbar\dot{\rho}_0 = [H_0(t), \rho_0] \tag{2.4}$$

and correspondingly must satisfy the relation[7)]

$$\rho_0(t) = S_0(t, t')\rho_0(t')S_0^{-1}(t, t'),$$

$$S_0(t, t') = T_t \exp\left\{-\frac{i}{\hbar}\int_{t'}^{t} H_0(t_1)\,dt_1\right\}. \tag{2.5}$$

From the compatibility of (2.3) and (2.5) it follows that the following commutation relation holds

$$[\rho_0(t), S_0(t+T, t)] = 0. \tag{2.6}$$

We now assume that in addition to an external field which determines the dependence of the Hamiltonian H_0 on the time the system is also acted upon by a small external "force" $F_a(t)$ the effect of which can be described by the perturbation operator

$$H'(t) = -\sum_a x_a F_a(t), \tag{2.7}$$

where x_a is the operator which refers to the system under consideration. We shall be interested in the reaction of the system to this force in the linear approximation.

The statistical operator $\rho_0'(t)$ describing the behavior of the system in the presence of the perturbation $H'(t)$ satisfies Eq. (2.4) in which the operator H_0 must be replaced by $H_0 + H'$. On assuming

$$\rho_0'(t) = \rho_0(t) + \Delta\rho(t),$$

we obtain in the case of a small increment $\Delta\rho(t)$

$$i\hbar\Delta\dot{\rho} = [H_0(t), \Delta\rho] - \sum_a [x_a, \rho_0(t)]F_a(t). \tag{2.8}$$

The solution of this equation in the case when the perturbation is switched on adiabatically at $t = -\infty$ has the form

$$\Delta\rho(t) = \sum_a \frac{i}{\hbar}\int_{-\infty}^{t} S_0(t, t')[x_a, \rho_0(t')]S_0^{-1}(t, t')F_a(t')\,dt'. \tag{2.9}$$

The reaction of the system, defined by the change $\Delta\langle x_b\rangle_t$ in the average value of a certain physical quantity x_b, is given by

$$\Delta\langle x_b\rangle_t = \mathrm{Sp}\,(\Delta\rho(t)\,x_b).$$

On the basis of (2.9) we obtain from this

$$\Delta\langle x_b\rangle_t = \sum_a \int_{-\infty}^{t} \overline{K}_{ba}(t, t')F_a(t')\,dt'$$

$$= \sum_a \int_0^{\infty} K_{ba}(t, \tau)F_a(t-\tau)\,d\tau, \tag{2.10}$$

where the response functions (or the aftereffects) $\overline{K}_{ba}(t, t')$ and $K_{ba}(t, \tau)$ are determined by the following expressions:

$$\overline{K}_{ba}(t, t') = \frac{1}{i\hbar}\mathrm{Sp}\,\{\rho_0(t')[x_a, x_b(t, t')]\}, \tag{2.11}$$

$$K_{ba}(t, \tau) = \overline{K}_{ba}(t, t-\tau), \tag{2.12}$$

$$x_{ba}(t, t') = S_0^{-1}(t, t')\,x_b S_0(t, t'). \tag{2.13}$$

The formulas (2.10)–(2.13) provide a direct generalization of the corresponding formulas of the paper by Callen et al [2] which define the response function for stationary equilibrium systems.[8)]

In the transition to stationary systems the function $\overline{K}_{ba}(t, t')$ begins to depend only on the difference $t - t'$, while the function $K_{ba}(t, \tau)$ becomes independent of t. In the case that the system is periodically nonstationary, when the operator $\rho_0(t)$ satisfies (2.3), the function $K_{ba}(t, \tau)$ is periodic with the same period T with respect to the variable t.

Just as in the case of stationary systems, a convenient characteristic of the dynamic properties of nonstationary systems is provided by the concept of admittance (cf., [8-11]) which is defined

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7)The question can be raised whether the limiting operator $\rho_0(t)$ satisfies "its own" Schrödinger equation (2.4). This can be proved in the following manner. On the basis of (2.1) and (2.2) we have

$$\rho_0(t) = \mathrm{Sp}_1 T_t \exp\left\{-\frac{i}{\hbar}\int_{t'}^{t}(H_0(t_1) + H_1 + \mu V)\,dt_1\right\}\rho(t')T_{-t}$$

$$\times \exp\left\{\frac{i}{\hbar}\int_{t'}^{t}(H_0(t_1) + H_1 + \mu V)\,dt_1\right\}.$$

Utilizing the Feynman technique of "disentangling" the operator (μV) from the T_t-exponential[13] and expanding in powers of μ we obtain

$$\rho_0(t) = S_0(t,t')\rho_0(t')S_0^{-1}(t,t')$$

$$+ \frac{\mu}{i\hbar}S_0(t,t')\mathrm{Sp}_1\int_{t'}^{t}[V(t_1), \rho(t')]\,dt_1 S_0^{-1}(t,t') + \ldots,$$

$$V(t) = S^{-1}(t)\,VS(t), \qquad i\hbar\dot{S} = (H_0(t) + H_1)S.$$

From this it follows that for $\mu \to 0$ the operator $\rho_0(t)$ actually does satisfy (2.5), and consequently also (2.4).

8)We note that formulas (2.10)–(2.13) are consequences of the solution of equation (2.8) in which the operator $\rho_0(t)$, generally speaking, can be arbitrary [i.e., it need not satisfy relations (2.3) and (2.5)], and are therefore valid for arbitrary nonstationary (not necessarily periodically-nonstationary) systems.

by the equation

$$\chi_{ba}(\omega, t) = \int_0^\infty K_{ba}(t, \tau)\, e^{i\omega\tau}\, d\tau. \qquad (2.14)$$

Such a definition of $\chi_{ba}(\omega, t)$ directly generalizes the commonly adopted concept of admittance for stationary systems. Indeed, for $F_a(t) = \mathrm{Re}\, F_{0a} \times \exp(-i\omega t)$ we have in virtue of (2.10), (2.12), and (2.14)

$$\Delta \langle x_b \rangle_t = \sum_a \mathrm{Re}\, \chi_{ba}(\omega, t)\, F_{0a} e^{-i\omega t}. \qquad (2.15)$$

However, formula (2.14) requires a somewhat more precise restatement. The point is that in the case of periodically-nonstationary systems the function $K_{ba}(t, \tau)$ can both decrease as $\tau \to \infty$ (in analogy to the case of equilibrium systems), and also it can increase. In the case of increasing $K_{ba}(t, \tau)$ the function $\chi_{ba}(\omega, t)$ can be defined for real values of ω as the analytic continuation along the real axis of ω of the integral (2.14) in which the quantity ω is complex and satisfies the condition $\mathrm{Im}\, \omega > \sigma_0$, where σ_0 indicates the rate of increase of $K_{ba}(t, \tau)$.

Such a formal definition of $\chi_{ba}(\omega, t)$ in the case of increasing $K_{ba}(t, \tau)$ has an entirely clear physical meaning. The fact that $K_{ba}(t, \tau)$ increases as $\tau \to \infty$ means that at least in a certain range of frequencies ω the system has a negative absorption for the perturbing energy $-\Sigma x_a F_{0a} \exp(-i\omega t)$, i.e., the value of the energy losses

$$Q_\omega \equiv -\sum_b \overline{\Delta \langle x_b \rangle_t \dot{F}_b(t)}$$

$$= \frac{i\omega}{4} \sum_{a,b} [\overline{\chi^*_{ba}(\omega, t)} - \overline{\chi_{ab}(\omega, t)}]\, F_{0b} F^*_{0a}, \qquad (2.16)$$

where $\chi_{ba}(\omega, t)$ is evaluated by the previously indicated method (the bar over the formula indicates averaging with respect to time), turns out to be negative over a certain frequency range.

The formula (2.16) agrees with the corresponding formula which defines energy losses in stationary systems [after replacement of $\overline{\chi_{ab}(\omega, t)}$ by $\chi_{ab}(\omega)$]. Just as in the case of the latter systems the sign of the losses Q_ω is determined by the behavior of the function $\chi_{ab}(\omega, t)$ on the real axis. For isotropic systems we have

$$Q_\omega = (\omega/2) \sum_a |F_{0a}|^2\, \mathrm{Im}\, \overline{\chi_{aa}(\omega, t)}$$

and the sign of Q_ω is determined by the sign of $\mathrm{Im}\, \overline{\chi_{aa}(\omega, t)}$. From the point of view of macroscopic electrodynamics of media with variable properties the case of increasing $K_{ab}(t, \tau)$ corresponds to media with negative absorption. In this case the system as a whole, i.e., the material medium (described by the polarizability tensor $\chi_{ik}(\omega, t)$) and the electromagnetic field determined by Maxwell's equations, can be either stable or unstable. In the former case we are dealing with an amplifier, and in the latter case with a generator of electromagnetic oscillations. Obviously, it is not possible to restrict oneself to linear electrodynamics in analyzing the latter case.

3. BASIC PROPERTIES OF THE RESPONSE FUNCTION AND OF THE ADMITTANCE

First of all we establish the symmetry relations of the function $K_{ab}(t, \tau)$ which follow from its definition (2.12) and from the invariance properties of the Schrödinger equation with respect to time reversal.

By utilizing the obvious property of the S-matrix: $S_0(t, t') = S_0^{-1}(t', t)$ we have for the function $\overline{K}_{ba}(t, t')$ on the basis of (2.11) and (2.5)

$$\overline{K}_{ba}(t, t') = -i\hbar^{-1}\, \mathrm{Sp}\, \{\rho_0(t')\, [x_a, S_0(t', t)\, x_b S_0^{-1}(t', t)]\}$$
$$= i\hbar^{-1}\, \mathrm{Sp}\, \{S_0(t, t')\, \rho_0(t')\, S_0^{-1}(t, t')\, [x_b, S_0^{-1}(t', t)\, x_a$$
$$S_0(t', t)]\} = -K_{ab}(t', t). \qquad (3.1)$$

From here in accordance with (2.12) we obtain for the function $K_{ab}(t, \tau)$

$$K_{ab}(t, \tau) = -K_{ba}(t - \tau, -\tau). \qquad (3.2)$$

Further, as we have already noted, the Hamiltonian H_0 depends on the time through the vector potential $\mathbf{A}(t)$ of the external field, which we shall represent in the form of a sum of a symmetric part $\mathbf{A}_s$ ($\mathbf{A}_s(t) = \mathbf{A}_s(-t)$) and an antisymmetric part $\mathbf{A}_a$ ($\mathbf{A}_a(t) = -\mathbf{A}_a(-t)$) (the constant magnetic field is included in $\mathbf{A}_s$). Since a simultaneous reversal of the sign of t and transition to the complex conjugate Hamiltonian does not alter the sign of the antisymmetric part $\mathbf{A}_a$, the symmetry properties of operators under time reversal in the case of nonstationary systems under discussion have the following form:[9]

$$\rho_0(-t, -\mathbf{A}_s) = \rho_0^*(t, \mathbf{A}_s),$$
$$x_a(-\mathbf{A}_s) = x_a^*(\mathbf{A}_s),$$
$$S_0(-t, -\mathbf{A}_s) = S_0^*(t, \mathbf{A}_s). \qquad (3.3)$$

On the basis of (2.11)—(2.13), (3.3) and of the reality of the function $K_{ba}(t, \tau)$ we obtain

$$K_{ba}(-t, -\tau; -\mathbf{A}_s) = -K_{ba}(t, \tau; \mathbf{A}_s). \qquad (3.4)$$

[9]It is assumed that the classical quantities corresponding to the operators x_a are even functions of the velocities.

If we take properties (3.2) and (3.4) into account at the same time we obtain

$$K_{ab}(t, \tau; \mathbf{A}_s) = K_{ba}(-t + \tau, \tau; -\mathbf{A}_s). \quad (3.5)$$

This last symmetry relation should, evidently, be regarded as a generalization of the principle of the symmetry of Onsager's kinetic coefficients [4,5] to the case of periodically-nonstationary systems. In spectral form it has the following appearance:

$$\chi_{ab}^{(n)}(\omega; \mathbf{A}_s) = \chi_{ba}^{(-n)}(\omega + n\Omega; -\mathbf{A}_s), \quad (3.6)$$

where $\chi_{ab}^{(n)}(\omega)$ is the Fourier coefficient in the series expansion of the admittance $\chi_{ab}(\omega, t)$ i.e.,

$$\chi_{ab}^{(n)}(\omega) = \frac{1}{T}\int_{-T/2}^{T/2} \chi_{ab}(\omega, t)\, e^{in\Omega t}. \quad (3.7)$$

For stationary systems n can assume only the one value n = 0 and the symmetry relation (3.6) reduces to the usual Onsager's symmetry relation.

In concluding this section we shall establish several other properties of the functions $K_{ab}(t, \tau)$ which are analogous to the sum rules well known in dispersion theory. From (2.11) and (2.12) it can be seen that if $[x_a, x_b] = i\hbar x_{ba}$, then

$$K_{ab}(t, 0) = \mathrm{Sp}(\rho_0(t)\, x_{ab}) = \langle x_{ab}\rangle_t. \quad (3.8)$$

Using the well known result from the theory of Fourier integrals (Abel's theorem):

$$-i \lim_{\omega\to\infty} \omega\chi_{ab}(\omega, t) = K_{ab}(t, 0), \quad (3.9)$$

we obtain

$$\chi_{ab}(\omega, t) \sim i\,\langle x_{ab}\rangle_t/\omega, \quad (3.10)$$

where the symbol ~ denotes that equality holds asymptotically as $\omega \to \infty$.

For example, for the components M_x and M_y of the magnetic moment per unit volume of the medium we have

$$[M_x, M_y] = i\hbar \sum_i \gamma_i^2 I_{iz}, \quad (3.11)$$

where I_{iz} is the component of the angular momentum of the i-th particle associated with the magnetic moment (γ_i is the gyromagnetic ratio). The formula (3.10) for the magnetic susceptibility tensor of a nonstationary medium yields in this case

$$\chi_{xy}(\omega, t) \sim \frac{i}{\omega}\sum_r \gamma_r \langle M_{rz}\rangle_t, \quad (3.12)$$

while $\chi_{ii}(\omega, t)$ is a small quantity of order higher than $1/\omega$; $\langle M_{rz}\rangle_t$ is the magnetization of the medium at the time t due to the external field A(t) and associated with the r-th component of the system.

Applying formula (3.8) to the problem of the electrical conductivity of a nonstationary medium (for example, of a plasma in a strong variable field[8,9]) and correspondingly assuming that

$$x_\nu = \sum_i e_i x_{i\nu}; \quad \dot{x}_\mu = \sum_i e_i \dot{x}_{i\mu} = \frac{i}{\hbar}\sum_i e_i [H_0(t), x_{i\mu}],$$

$$H_0(t) = \sum_i \frac{1}{2m_i}\left\{p_{i\nu} - \frac{e_i}{c}A_\nu(\mathbf{r}_i, t)\right\}^2 + V(\mathbf{r}_1, \mathbf{r}_2, \ldots, \mathbf{r}_N),$$

we obtain

$$K_{\mu\nu}(t, 0) = \sum_{i=1}^{N} \frac{e_i^2}{m_i}\delta_{\mu\nu} = \sum_r \frac{n_r e_r^2}{m_r}\delta_{\mu\nu}, \quad (3.13)$$

where n_r is the density of the current carriers of the r-th kind of mass m_r. We thus obtain for the electrical conductivity tensor (with respect to the total current) $\sigma_{\mu\nu}(\omega, t)$ the same asymptotic representation (for $\omega \to \infty$) as in the case of stationary media

$$\sigma_{\mu\nu}(\omega, t) \sim \frac{i}{\omega}\sum_r \frac{n_r e_r^2}{m_r}\delta_{\mu\nu}. \quad (3.14)$$

4. FLUCTUATIONS IN CLASSICAL NONSTATIONARY SYSTEMS

For stationary systems (in thermodynamic equilibrium) the fluctuation-dissipation theorem holds, one of the many possible forms of which consists in establishing a relation between the relaxation function of the system and the correlation function of the equilibrium fluctuations of corresponding physical quantities [3]. It can be easily shown that a similar relation holds for nonstationary systems. However, our discussion will now be restricted only to classical systems characterized by positive absorption of energy along the whole axis of frequencies ω (so that the response function $K_{ab}(t, \tau)$ always dies away for $\tau \to \infty$).

We introduce the concept of the relaxation function $\varphi_{ba}(t, s)$ for a nonstationary system as a response to the force $F_a(t)$ which is constant and equal to unity until the time s and is equal to zero for t > s. In accordance with (2.10) we have

$$\varphi_{ba}(t, s) = \int_{t-s}^{\infty} K_{ba}(t, \tau)\, d\tau, \quad t > s. \quad (4.1)$$

For periodically-nonstationary systems, when $K_{ba}(t, \tau) = K_{ba}(t+T, \tau)$, the function $\varphi_{ba}(t, s)$ satisfies the condition

$$\varphi_{ab}(t, s) = \varphi_{ab}(t + T, s + T). \quad (4.2)$$

The correlation function $\psi_{ba}(t, \tau)$ of nonstationary fluctuations of the quantities $x_a(t)$ and $x_b(t)$ is by definition equal to

$$\psi_{ba}(t, \tau) = \langle \Delta x_b (t+\tau)\, \Delta x_a (t)\rangle = \langle\langle \Delta x_b (t+\tau) \mid \Delta x_i (t)\rangle\, \Delta x_a (t)\rangle_{\Delta x_j(t)}, \qquad (4.3)$$

where the notation $\langle \ldots \mid \Delta x_i(t)\rangle$ denotes averaging corresponding to the time $t+\tau$, under the condition that at the time t the quantities Δx_i ($i = 1, 2, \ldots, n$) have given values; the notation $\langle \ldots \rangle_{\Delta x_j(t)}$ denotes averaging over all posible values of the fluctuations Δx_j at time t.

Just as in the development of the phenomenological spectral theory of fluctuations in equilibrium systems, we shall utilize for the determination of conditional averages Onsager's postulate with respect to the regression (decay) of fluctuations [4,5], and, in particular, we shall assume

$$\langle \Delta x_b (t+\tau) \mid \Delta x_i (t)\rangle = \sum_{i,j} \varphi_{bi}(t+\tau, t)\, \varphi_{ij}^{-1}(t, t)\, \Delta x_j (t). \qquad (4.4)$$

Here $\Sigma_j \varphi_{ij}^{-1}(t,t)\Delta x_j(t)$ is, evidently, that value of the constant force F_i acting until the time t which produces at the time t the given values of the responses Δx_j ($j = 1, 2, \ldots, n$).

On substituting (4.4) into (4.3) and on averaging over $\Delta x_j(t)$ we obtain the desired relation between the correlation function for the fluctuations in a nonstationary system and its relaxation function:

$$\psi_{ba}(t, \tau) = \sum_{i,j} \varphi_{bi}(t+\tau, t)\, \varphi_{ij}^{-1}(t, t)\, \psi_{ja}(t, 0); \quad \tau > 0. \qquad (4.4')$$

This relation, naturally, does not have the same universal character as in the case of equilibrium systems since its right hand side contains the matrix of the intensities

$$\psi_{ij}(t, 0) \equiv \langle \Delta x_i (t)\, \Delta x_j (t)\rangle, \qquad (4.5)$$

which in the case under discussion, generally speaking, depends on the time and can be evaluated only on the basis of a specific kinetic model of the fluctuations. Thus, the relation (4.4′) enables us only to calculate the correlation function of the nonequilibrium fluctuations in the system for $\tau > 0$ if we are given its dynamic properties (i.e., $K_{ba}(t, \tau)$ or $\varphi_{ba}(t, \tau)$), and also the intensity (4.5) of these fluctuations at each instant of time.

The correlation function $\psi_{ab}(t, \tau)$ for negative τ can be defined in accordance with the obvious relation which is valid for any arbitrary nonstationary fluctuations [9]:

$$\psi_{ab}(t, -\tau) = \psi_{ba}(t-\tau, \tau). \qquad (4.6)$$

The combined relations (4.4′) and (4.6) enable us to determine the spectral intensity $G_{ab}(\omega, t)$ of the fluctuations in a nonstationary system if we are given the total (integral) intensity (4.5) of these fluctuations. For this we need only use the natural generalization of the Khinchin-Wiener [9] formula:

$$G_{ab}(\omega, t) = \frac{1}{2\pi}\int_{-\infty}^{\infty} \psi_{ab}(t, \tau)\, e^{i\omega\tau}\, d\tau = \frac{1}{2\pi}\Big\{\int_0^{\infty} [\psi_{ab}(t, \tau) + \psi_{ba}(t-\tau, \tau)] \cos\omega\tau\, d\tau + i\int_0^{\infty} [\psi_{ab}(t, \tau) - \psi_{ba}(t-\tau, \tau)] \sin\omega\tau\, d\tau\Big\}. \qquad (4.7)$$

In going over to systems in thermodynamic equilibrium when $\varphi_{ab}(t, s)$ depends on the difference $(t-s)$, while the correlation function ψ_{ab} does not depend on t, we have [14]

$$\psi_{ik}(0) = kT\left(\frac{\partial^2 S}{\partial \Delta x_i\, \partial \Delta x_k}\right)^{-1}_{\Delta x_j = 0}, \quad \varphi_{ik}^{-1}(0) = \left(\frac{\partial^2 S}{\partial \Delta x_i\, \partial \Delta x_k}\right)_{\Delta x_j = 0}, \qquad (4.8)$$

where $S = S(\Delta x_1, \Delta x_2, \ldots, \Delta x_n)$ is the entropy of the system. Substitution of (4.8) into (4.4′) leads to the fluctuation-dissipation theorem [3].

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[1] Voprosy kvantovoĭ teorii neobratimykh protsessov (Problems in the Quantum Theory of Irreversible Processes), Collection of Articles edited by V. L. Bonch-Bruevich, IIL, 1961.

[2] Callen, Barasch, and Jackson, Phys. Rev. **88**, 1382 (1952).

[3] R. Kubo, J. Phys. Soc. Japan **12**, 570 (1957).

[4] L. Onsager, Phys. Rev. **37**, 405, **38**, 2265 (1931).

[5] H. B. G. Casimir, Revs. Modern Phys. **17**, 343 (1945).

[6] S. A. Akhmanov and R. V. Khokhlov, JETP **43**, 351 (1962), Soviet Phys. JETP **16**, 252 (1963).

[7] F. V. Bunkin, Radiotekhnika i élektronika (Radio Engineering and Electronics) **5**, 296 (1960).

[8] V. N. Lugovoĭ, JETP **41**, 1562 (1961), Soviet Phys. JETP **14**, 1113 (1962).

[9] F. V. Bunkin, JETP **41**, 1859 (1961), Soviet Phys. JETP **14**, 1322 (1962).

[10] F. V. Bunkin, Izv. Vuzov, ser. radiofizika **5**, 714 (1962).

[11] V. N. Lugovoĭ, Izv. Vuzov, ser. radiofizika **5**, 901 (1962).

[12] L. I. Mandel'shtam, JETP **15**, 605 (1945).

[13] R. Feynman, Phys. Rev. **84**, 108 (1951).

[14] L. D. Landau and E. M. Lifshitz, Statisticheskaya fizika (Statistical Physics) Gostekhizdat, 1951.

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250