Fluctuation-dissipation relations for nonequilibrium processes in open systems

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The complete set of nonlinear fluctuation-dissipation relations previously derived by the authors [Sov. Phys. JETP 45, 125 (1977)] for an arbitrary closed thermodynamic system is extended to open systems and applied to the analysis of the universal relations between dissipation and fluctuation processes in nonequilibrium stationary states of an open system. General expressions are found for the nonlinear transport coefficients in terms of the fluctuation characteristics of the system (diffusion coefficients). As an application of the general theory, the close relation between the statistics of charge transport through a p-n junction and the shape of the volt-ampere characteristic of the junction is demonstrated. The general structure of the Markov model constructed for fluctuations in nonequilibrium states constructed in accord with the exact fluctuation-dissipation relations is considered. Special models of the system, for which the fluctuation-dissipation theorem in its usual form is valid even in nonequilibrium states are also considered.

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1. INTRODUCTION

In a paper of the authors, a working formula was obtained for the complete set of universal fluctuation-dissipation relations (FDR). These relations, which are the consequence of the reversibility of the microscopic motion in time and of the extremal properties of the thermodynamic equilibrium states, connect the statistical characteristics of the equilibrium thermal fluctuations with the characteristics of nonequilibrium (and nonlinear) processes in a system subject to a dynamic external perturbation.

Greatest interest attaches to the thermodynamic consequences of the FDR, and to their application to the theory of irreversible phenomena. The standard formal method of construction of a phenomenological or semi-phenomenological model of irreversible processes consists of singling out some set of macrovariables and assuming that this set is closed in the statistical sense, i.e., that its evolution is Markovian. The FDR, in conjunction with the Markovian hypothesis, yields directly the connection between the (generally speaking, nonlinear) transport coefficients and the statistical characteristics of the fluctuation "sources" in the stochastic equations of the system (the Langevin form of the fluctuation-dissipation theory). In the general case, the fluctuation sources are non-Gaussian and depend on the macrostate of the system. The Markovian FDR were studied in detail in a number of papers by Stratonovich (see, for example, Refs. 2-4), who showed that although the nonlinear FDR carries less information that the linear ones (the ratio of the number of $n$-index FDR to the number of $n$-index parameters of the theory decreases with increase in $n$), cases are possible in which additional physical assumptions on the character of the fluctuations, together with the nonlinear FDR, lead to an unambiguous reconstruction of the entire kinetic operator of the Markov process from the nonlinear relaxation equations.

It was shown later in Ref. 1 that the Stratonovich relations are also applicable to nonstationary fluctuations whose kinetic operator depends on the time through the external forces. However, the derivation of all these relations was inseparably connected with the assumption that the system is finite and closed in the sense that the constant external forces do not upset the thermodynamic equilibrium but only change its parameters, and that the motion of the macrovariables is finite.

At the same time it is of interest to study the FDR for open systems, in which the constant external forces $x(t)$ induce undamped fluxes of momentum, energy, entropy, charge, and other quantities and, at the same time bring the system into a stationary nonequilibrium state (SNS). The macrovariables $Q(t)$ conjugate to the forces $x(t)$ then experience an unrestricted diffusion, so that it is impossible to ascribe to them a stationary distribution normalized to unity. It is therefore natural to take the currents $I(t) = dQ(t)/dt$ as the defining macrovariables. In equilibrium and in SNS these are stationary random processes; that is, the currents can be considered to be Markovian in the construction of a phenomenological model that includes irreversibility explicitly.

The aim of the present work lay in the derivation of the FDR for currents in SNS from the general formulas obtained in Ref. 1. We emphasize that the basic results of Ref. 1—the symmetry formulas for the characteristic functional of the current and for the probability functions—are applicable in principle also for the description of SNS. In this case, it is only necessary to assume that the transition to the thermodynamic limit is carried out (in complete system—macrovariables plus thermal) and deal in corresponding fashion with the fluctuation moment (correlation) functions. Thus, the present work represents a direct continuation of the work of Ref. 1.

The dynamic FDR for currents are considered in Sec. 2 in more detail than before, and as applied to SNS. In Sec. 3, the Markov relations are derived and their simple special realizations studied (in particular, systems...
to which the fluctuation-dissipation theorem in its usual equilibrium form is applicable also in SNS). The general theory is illustrated in Sec. 4 by the example of the use of nonlinear FDR for the construction of dynamical and statistical characteristics of charge transport in semiconductors.

2. RIGOROUS STATISTICAL DESCRIPTION OF NONEQUILIBRIUM STATIONARY STATES OF THE SYSTEM

Let the external forces $\mathbf{v}(t)$ in thermodynamic equilibrium start to act on a system and alter the Hamiltonian of the system:

$$H(t) = H_{eq}(t) - H_{int}(t)G_{n},$$

where $Q_{m}$ are the internal variables conjugate with the forces. If the forces are constant after their being switched on, then the system returns to thermodynamic equilibrium during after characteristic time $T$, but now with other parameters that depend on $x$ (as in Ref. 1), we shall assume the system contains a subsystem—a thermostat of such large size that its temperature can be assumed to be unchanged in all the nonequilibrium processes. If the time $t_{r}$ (which has the meaning of the relaxation time of some of the macrovariables $Q(t)$ from the initial equilibrium state to the final state) is sufficiently large, then we can isolate an interval during which the system is in a quasistationary state close to an SNS and characterized by quasistationary transport processes. By increasing the dimensions of the system unrestrictedly and going to the thermodynamic limit, we obtain an SNS (the state of infinitely dragged out relaxation process) in which the currents $\mathbf{J}(t) = 0$, and not the macrovariables $Q(t)$ themselves are stationary random processes. Their mean values give a macroscopic description of the SNS.

Since the complete set of nonlinear FDR obtained in Ref. 1 is applicable to an arbitrarily large closed system, we can extend these FDR to open systems in the SNS with the help of a transition to the thermodynamic limit, which is actually effected only conceptually, and reduces to the assumption that certain integrals $\langle \mathbf{J}, \mathbf{J}(t) \rangle$ and others similar to them have non-zero finite values. Thus, almost all the formulas of Ref. 1 can be applied to open systems. As a result, we obtain universal relations that do not depend on the specific physical nature of the transport process between the dissipative and fluctuation characteristics of the SNS.

We denote by $P(x(t); x(t))$ the probability functional of the currents in a specified realization $x(t)$ of the external forces. The following symmetry relation for it follows from the results of Ref. 1:

$$P(x(t); x(t)) = \exp\left\{ -\beta \left[ \mathbf{J}(t) \cdot \mathbf{J}(t) \right] \right\},$$

where $\beta = 1/T$, $T$ is the temperature of the thermostat contained in the considered system. For the characteristic functional of the currents

$$\mathbf{B}(x(t); x(t)) = \exp\left\{ -\frac{1}{\beta} \left[ \mathbf{J}(t) \cdot \mathbf{J}(t) \right] \right\},$$

the formula equivalent to (1) has the form

$$\mathbf{B}(x(t); x(t)) = \exp\left\{ -\frac{1}{\beta} \langle \mathbf{J}, \mathbf{J}(t) \rangle \right\}. \quad (2)$$

The angular brackets with comma inside (Malakhov's cumulant brackets) denote the cumulant functions, for example, $(\mathbf{A}, \mathbf{B}) = \langle \mathbf{A} \mathbf{B} \rangle - \langle \mathbf{A} \rangle \langle \mathbf{B} \rangle$.

In an open system, certain macrovariables $Q(t)$ experience diffusion. We introduce the diffusion coefficients $D_{m}(x)$ at constant forces by means of the generating relation

$$\langle \mathbf{J}, \mathbf{J}(t) \rangle = \frac{1}{\beta} \int \frac{d\mathbf{J}}{T} D_{m}(x) \mathbf{J},$$

which are tensors that are symmetric relative to the upper and lower indices (the Greek indices enumerate the variables). The two-index relations have the form

$$D_{mi} = \frac{1}{\beta} \int \frac{d\mathbf{J}}{T} D_{m}(x) \mathbf{J}_{i} \mathbf{J}_{j},$$

at $\varepsilon _{i} \varepsilon _{j} = +1$ and

$$D_{mi} = \frac{1}{\beta} \int \frac{d\mathbf{J}}{T} D_{m}(x) \mathbf{J}_{i} \mathbf{J}_{j},$$

at $\varepsilon _{i} \varepsilon _{j} = 0$. This leads to the Onsager-Casimir relations

$$(D_{mi} + D_{mj}) = \frac{1}{\beta} \int \frac{d\mathbf{J}}{T} D_{m}(x) \mathbf{J}_{i} \mathbf{J}_{j},$$

at $\varepsilon _{i} \varepsilon _{j} = +1$. This is the analogue of the three-index formulas: at $\varepsilon _{i} \varepsilon _{j} \varepsilon _{k} = +1$ and

$$D_{mi} = \frac{1}{\beta} \int \frac{d\mathbf{J}}{T} D_{m}(x) \mathbf{J}_{i} \mathbf{J}_{j},$$

$$D_{mj} = \frac{1}{\beta} \int \frac{d\mathbf{J}}{T} D_{m}(x) \mathbf{J}_{i} \mathbf{J}_{j},$$

at $\varepsilon _{i} \varepsilon _{j} \varepsilon _{k} = 0$, and at $\varepsilon _{i} \varepsilon _{j} \varepsilon _{k} = -1$. This leads to the Onsager-Casimir relations

$$(D_{mi} + D_{mj} + D_{mk}) = \frac{1}{\beta} \int \frac{d\mathbf{J}}{T} D_{m}(x) \mathbf{J}_{i} \mathbf{J}_{j} \mathbf{J}_{k}.$$
The tensor functional possesses the following properties:

1. **Causality Condition**: Independence of the correlators of currents by the causality condition: independence of the structure of the characteristic functional of the system on the time parameter.

2. **Simple Analysis**: If we identify \( D_{k}\) with the kinetic-operator coefficients that depend on the structure of the characteristic functional of the system, then the system is averaged over the equilibrium distribution of \( Q \). The absence of universal nonlinear reciprocity relations of the system does not exclude the possibility of such relations for specific systems possessing dynamical symmetries.

3. **Practically Markov Model**: Fluctuations of quasi-equilibrium state. Since the diffusion coefficients depend on the energy dissipation, they can be found in real systems from the dynamic nondissipative model for the macrovariables and expressed in terms of the parameters of the quasi-equilibrium state. Since the fundamental property means that the functional \( \Gamma_{\text{f}} \) is constructed from quasi-equilibrium cumulant expressions for the averages. As a result, we obtain

\[
\Gamma_{\text{f}}(x(t)) = \frac{1}{2\pi i} \int \text{d}x \left\{ x \Gamma_{\text{f}}(x(t)) \right\} \frac{1}{x - \text{FDR}(x(t))}.
\]

From this formula and (5) we obtain the relations between the real and the quasi-equilibrium cumulant functions:

\[
\Gamma_{\text{f}}(x(t)) = \frac{1}{2\pi i} \int \text{d}x \left\{ x \Gamma_{\text{f}}(x(t)) \right\} \frac{1}{x - \text{FDR}(x(t))}.
\]

In the case of constant forces this yields, in particular, the nonlinear transport equations in the form

\[
\Gamma_{\text{f}}(x(t)) = \frac{1}{2\pi i} \int \text{d}x \left\{ x \Gamma_{\text{f}}(x(t)) \right\} \frac{1}{x - \text{FDR}(x(t))}.
\]

The linear reciprocity relations \( \Gamma_{\text{f}}(x(t)) = \Gamma_{\text{f}}(x(t)) \) follow from the property 2; however, \( \Gamma_{\text{f}}(x) \) does not possess such symmetry at \( x \neq 0 \).

As the separate and reversible and irreversible transport coefficients in the right side of (7):

\[
\Gamma_{\text{f}}(x(t)) = \frac{1}{2\pi i} \int \text{d}x \left\{ x \Gamma_{\text{f}}(x(t)) \right\} \frac{1}{x - \text{FDR}(x(t))}.
\]

The formula (5) in terms of the even diffusion coefficients \( D^{\text{e}} \):

\[
N_t(x) = \sum_{k=1}^{N_{\text{e}}} \sum_{l=0}^{N_{\text{e}}} C_k \left( \frac{1}{2} \right)^{k} D^{\text{e}}(x)^{2k} + D_{\text{e}}(x)^{2k},
\]

where the numbers \( C_k \) are determined by the generating function (see also Ref. 2).

**Remarks**

1. It depends only on \( x(0) \) and \( x(t) \) at \( t > \tau > 0 \).
2. \( \Gamma_{\text{f}}(x(t)) = \Gamma_{\text{f}}(x(t)) \) is the mean value of the current vector at the time \( t \).
3. \( \text{FDR}(x(t)) = \text{FDR}(x(t)) \) is the mean value of the current vector at the time \( t \).

As for the inverse coefficients \( \Gamma_{\text{f}}^{-1} \) that are not connected with the FDR through the functional relations and with the dissipation, they can be found in real systems from the dynamic nondissipative model for the macrovariables and expressed in terms of the parameters of the quasi-equilibrium state. Since the fundamental property means that the functional \( \Gamma_{\text{f}}^{-1} \) is constructed from quasi-equilibrium cumulant expressions for the averages. As a result, we obtain

\[
W_t(x(t)) = \left\{ \frac{1}{2\pi i} \int \text{d}x \left\{ x \Gamma_{\text{f}}(x(t)) \right\} \right\} \frac{1}{x - \text{FDR}(x(t))}.
\]

where \( \text{FDR}(x(t)) = \text{FDR}(x(t)) \), \( W_t(x(t)) = W_t(x(t)) \), and \( x(t) \) denotes the conditional mean value (under the condition that at the time \( t = 0 \) we have \( x(0) = x(t) \) and prior to this moment the system had been in equilibrium). This formula expresses the distribution of the
and the cumulant power function of the currents in SNS in terms of the equilibrium distribution of forces. Although the energy absorbed by the system has a diffusion behavior, the mean value of the exponential in (10) is finite, since the contributions from this exponential can be obtained by transforming it with the help of the FDR and going over to the characteristic function.

The result has the form

$$Z(t) = \exp \left[ \int_{0}^{t} \text{d}t' \delta F(0) \right]$$

where

$$\delta F(0) = \sum_{n=1}^{\infty} \frac{1}{n!} \text{d}n \delta F(0) \mu_n.$$ 

The resultant expressions for the cumulant currents in the SNS can also be obtained as a particular case of (6).

In the general case, the cumulant power function of the currents in SNS in terms of the equilibrium distribution is given by the expression

$$\langle N(t) \rangle = \int_{0}^{t} \text{d}t' \delta F(t') \mu_n.$$ 

The derivation of the symmetry formula for the kinetic operator of the currents from the FDR is given in the Appendix, Sec. 3. Just as in Ref. 1, we can show that this operator depends on the external forces in an instantaneous fashion. In standard (tensor) notation, the kinetic operator and its conjugate have the form

$$W(x) = W(x) \langle \frac{\delta F}{\delta x} \rangle,$$

where

$$W(x) = \frac{1}{2} \langle \frac{\delta^2 F}{\delta x^2} \rangle.$$ 

The operator equation

$$L \langle x, \frac{\delta}{\delta x} \rangle W(x) = W(x) \langle \frac{\delta^2 F}{\delta x^2} \rangle$$

follows from (1). This operator equation is equivalent to the following Markov FDR between the kinetic coefficients:

$$W(x) = W(x) \langle \frac{\delta^2 F}{\delta x^2} \rangle.$$ 

The resultant expressions for the cumulant currents in the SNS can also be obtained as a particular case of (6).

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Substituting these formulas in the expression for the kinetic operator, we find

$$L(x, I) = \frac{1}{2} \left( \frac{\partial}{\partial x} + A(x) \right) \cdot L(x, I) \cdot \left( \frac{\partial}{\partial x} + A(x) \right),$$

where the number $S_n$ (in the multidimensional case it is necessary to distinguish between the vector indices $n = [n_1, n_2, \ldots]$ and the scalar indices $n = n_1 + n_2 + \ldots$, which is not difficult to do) are determined in the following way:

$$\sum_n S_n n^n = 2^{(n + 1)^2 - 1}.$$  

The coefficients $K_n$ are free (formally, if we disregard their physical meaning) parameters of the Markov model and are not connected with one another by the FDR. We consider separately the special case in which the fluctuation sources in the Langevin stochastic equations for the currents can be regarded as locally Gaussian. In this case, $K_n(x, I) = 0$ at $n > 3$ and the kinetic equation transforms into the Fokker-Planck equation. Formulas (15)-(17) reduce to the form

$$K_n(x, I) = K_n(I), n > 3.$$  

The functions $K_n$ represent by definition the reversible components of the relaxation (phenomenological) equations for the currents. Therefore, the two terms in the expression (19) for the kinetic operator can be regarded as the dynamic and thermodynamic terms. In the same fashion, the operator (17) splits into parts. The dependence of $K_n$ on $x$ at $n > 2$ indicates that the external forces, generally speaking, influence the state of the thermostat. However, the FDR (15) admits of such a possibility when this influence is absent and $K_n(x, I) = K_n(I), n > 2$. In this simplest case, formula (19) follows from (17) as well as the equality

$$L = \frac{1}{2} \left( \frac{\partial}{\partial x} + A(x) \right) \cdot L(x, I) \cdot \left( \frac{\partial}{\partial x} + A(x) \right).$$

The condition (19) is satisfied in natural and simple fashion if

$$K_n(x, I) = K_n(I) + A(x).$$

Here

$$L = \frac{1}{2} \left( \frac{\partial}{\partial x} + A(x) \right) \cdot L(x, I) \cdot \left( \frac{\partial}{\partial x} + A(x) \right).$$

As is well known, the fluctuation-dissipation theorem (FDT) is not satisfied in the SNS and it is impossible to determine the correlation functions of the fluctuations in general form from the linear response to the weak perturbation of the SNS. We consider this process in the Markov model. We set $x(t) = x + \delta x(t)$. The kinetic operator $L = L(I)$ depends on time through the force. If the perturbation acts over a finite interval, then

$$f(t) = f(t) \int dt' \exp \left\{ \int_{t'}^t L(x, I) \cdot \delta x(t) \right\} W(I(t)).$$

where $\delta x(t)$ is the chronologically ordered exponential. We then find for the linear response in the case (20')

$$\frac{\delta}{\delta x(t)} W(I(t)) = \int dt' \exp \left\{ \int_{t'}^t L(x, I) \cdot \delta x(t) \right\} W(I(t)).$$

In equilibrium, only the first term remains, yielding the usual FDT. In the SNS, the second term is expressed, generally speaking, in terms of the higher cumulant functions. But in the special case in which $A(x) = 0$, the operator (21) splits into parts.

$$L(W(I) = W(I)), I > 0.$$  

The functions $K_n$ represent by definition the reversible components of the relaxation (phenomenological) equations for the currents. Therefore, the two terms in the expression (19) for the kinetic operator can be regarded as the dynamic and thermodynamic terms. In the same fashion, the operator (17) splits into parts. The dependence of $K_n$ on $x$ at $n > 2$ indicates that the external forces, generally speaking, influence the state of the thermostat. However, the FDR (15) admits of such a possibility when this influence is absent and $K_n(x, I) = K_n(I), n > 2$. In this simplest case, formula (19) follows from (17) as well as the equality

$$L = \frac{1}{2} \left( \frac{\partial}{\partial x} + A(x) \right) \cdot L(x, I) \cdot \left( \frac{\partial}{\partial x} + A(x) \right).$$

The condition (19) is satisfied in natural and simple fashion if

$$K_n(x, I) = K_n(I) + A(x).$$

Here

$$L = \frac{1}{2} \left( \frac{\partial}{\partial x} + A(x) \right) \cdot L(x, I) \cdot \left( \frac{\partial}{\partial x} + A(x) \right).$$

As an example, we consider a one-dimensional Markov process. From (20') and (22), we find

$$W(I) = \frac{1}{A(I)} \exp \left\{ \frac{1}{A(I)} \left[ \int_{-\infty}^\infty \frac{dI}{A(I)} \int_{-\infty}^\infty \frac{dI'}{A(I')} \right] \right\},$$

Consequently, the temperature $T(x)$ of the macrovariables in the nonequilibrium system with properties (22), (23) can be determined, analogously to the equilibrium system, from the relation between the diffusion coefficient and the linear differential response.

As an example, we consider a one-dimensional Markov process. From (20') and (22), we find

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Definite conditions follow then from this and from (24) on the kinetic operator $L_k$. In particular, if $L_k$ is the Fokker-Planck operator, then it is completely determined by the function $A(I)$. For the case $x = 1$, we find, with account taken of the FDR,

$$L_k = \frac{d}{dx} \left[ (x - g) A(x) \right] + \frac{d}{dx} \left[ \frac{d}{dx} \left( \frac{d}{dx} \right) \right].$$

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In the case $c=-1$ ($f$ is an even variable in time) 
\[
L = -\frac{\partial}{\partial x} \{ (t-x) I(t) - 2 T x^2 \} \left\{ \frac{1+I}{2} \right\}.
\]
\[
A(t)=A(I)(y+n), T(x)=-\frac{y}{1-I}, I(x)=-\frac{y}{1-I}, A=const.
\] (27)

Here $\{AI\}$ has the meaning of a current relaxation time that depends on $I$. These two cases exhaust the set of one-dimensional Markov models (with a Gaussian noise source), for which the non-equilibrium FDT (23) holds.

We now discuss briefly the results of this section. The condition (22) at which the FDT (28) holds was introduced formally and can be shown to be artificial; therefore it is necessary to define the considered class of systems in more detail. Of course, complete clarity can be achieved only in a specific physical application. In this context, we should like to remark that actual usefulness of nonlinear FDR turns out to be considerably greater than could be assumed beforehand when they are supplemented by information (exact or model-derived) on the specifics of the fluctuations in the actual system.

For simplicity, we consider the case $b(x)=0$, $T(x)=T$. We introduce the new variable $P_x$ by the relations $3/2 P_x = A_x(I)$, which is always possible if (22) is satisfied. Then the non-equilibrium distribution $W_t(P)$ and the kinetic operator for the variables $P$ have the form
\[
W_t(P)=\exp\left\{ -\frac{1}{2T} \int d(b) \right\} L = -\frac{\partial}{\partial x} \{ (t-x) I(t) - 2 T x^2 \} \left\{ \frac{1+I}{2} \right\}.
\]
\[(27') \]
A general interpretation of $P$ is suggested by the example of Brownian motion, where $Q$ is the coordinate of a particle diffusing under the action of a constant force, $I$ is its velocity, and $P$ is its momentum. The $P(t)$ dependence is nonlinear, for example, in the case in which $I$ and $P$ are the velocity and momentum of an electron in a crystal lattice. Equation (27') is analogous to the formulation of the transport is also very important, the stochastic model of the system. We consider now some special cases.

Therefore $P$ and $I$ can be regarded as thermodynamic conjugate variables for a fictitious equilibrium system with the perturbed Hamiltonian $H=H_0 - PT$.

Then, nonequilibrium systems for which the conditions (27') are satisfied, are systems close to equilibrium, in particular in the sense that the FDT (23) follows from (27'). On the other hand, such nonlinear systems are in many respects similar to linear ones, since they allow us to reconstruct unambiguously the equilibrium fluctuation coefficients of diffusion, in accord with (24') (and, as can be shown, the entire operator $L$) from the nonequilibrium dissipation characteristics. A more detailed review of this circle of problems goes beyond the theme of this paper.

4. EXAMPLE. NONLINEAR FDR FOR STATIONARY CHARGE TRANSPORT

1. Being primarily interested in illustrations of the general FDR, we consider the case of a single time-
even variable $Q(t)$, which represents, for example, the electric charge (or mass, energy).

In the limit (in correspondence with (2) and (3), we have the universal generating FDR
\[
B(u) = B(\tilde{u})\exp\left\{ -\frac{1}{2T} \int d(b) \right\} B(\tilde{u})
\]
\[(29)\]
Obviously, $\exp\{B(\tilde{u})\}$ as a function of $u$ is a characteristic function of an infinitely divisible distribution (see, for example, Ref. 7). Consequently, $B(u)$ can be represented in the form
\[
B(u)=\exp\left\{ -\frac{1}{2T} \int d(b) \right\} B(\tilde{u})
\]
\[(29)\]
where $\tilde{u}(u)$ is a nonnegative function normalized to unity, $\tilde{T}=T_x$ and $D=D_1$ are, as before, the average value of the current and the coefficient of diffusion, respectively (the spectral density of the current fluctuations at zero frequency). The nonlinearity of the variables $a$ in (29) and $Q$ are the same. Equation (29) enables us to consider the random process $Q(t)$--the value of the transported charge--as the superposition of independent Poisson processes, in each of which the charge is transported in discrete portions of value $a$ with the mean value of each portion per unit time proportional to $\tilde{T} = (1-\tilde{T})^T (1-\tilde{T})^T$. The following FDR result from (29)

\[
\tilde{I}(u)=\int \frac{d(a)}{\tilde{T}} \tilde{P}(u) \{ 1 - \tilde{T} \} \left\{ 1 - \tilde{T} \right\}.
\]
\[(30)\]
with only the first two of these relations independent, while the third is a consequence of the first two. Equation (30) is a special case of the general expression for the average current in terms of the fluctuations characteristics of the transport process. Naturally, the description of this process in terms of only some of the diffusion coefficients $D_j(a)$ is far from complete and says nothing about the properties of the process that are local in time and space, for example, the correlation function of the current fluctuations and their spectrum at high frequencies. Nevertheless, knowledge of the global characteristics of the transport is also very important, while the FDR for them are useful in the construction of the stochastic model of the system. We consider now some special cases.

2. Let $I(t)$ be the electric current flowing through a nonlinear one-port network to which a voltage $x(t)$ is applied. We assume that the charge is transported according to the Poisson law by particles of a single type and definite charge $q$. Then
\[
\phi(x) = \frac{1}{T} \exp\left\{ -\frac{1}{2T} \int d(b) \right\} B(\tilde{u})
\]
\[(29)\]
We then find from (29), (30) (setting $z=x/T$)
\[
\tilde{I}(u)=\int \frac{d(a)}{\tilde{T}} \tilde{P}(u) \{ 1 - \tilde{T} \} \left\{ 1 - \tilde{T} \right\}.
\]
\[(31)\]
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It then follows from the FDR (28) (if we introduce we get from (34)

\[ I(z) = g(z)(e^{m^2} - 1) \]

This is the simplest case of connection between the statistics of charge transport and nonlinear dissipation.

3. We now consider the somewhat more complicated stochastic model of current through a semiconducting diode \( (x = n \) junction). We assume that non-Poisson statistics of electron (and hole) transport, i.e., that the separate transitions correlate with one another. More accurately, we assume that the successive time intervals \( \tau \) between direct transitions are independent, but are distributed not exponentially as in the Poisson case, but with a probability density

\[ n(t) = \frac{1}{T} e^{-\frac{t}{T}} \]

The average number of transitions per unit time is equal to \( \lambda = n \). We consider the distribution of the charge \( Q(t) \) that passes in the forward direction. We introduce the function

\[ \Lambda(x) = \lim_{T \to \infty} \frac{1}{T} \ln[e^{-\lambda ft} - \lambda ft] \]

It can be shown that this function is found from the set of equations

\[ x - \Delta_x(x) = 0 \]

which constitute two forms of the equation for the pole of the function

\[ \int e^{-\lambda ft} \ln[e^{-\lambda ft} - \lambda ft] \]

Since in our case,

\[ \lambda(x) = \mu e^{\frac{m^2}{2}} \]

we get from (34)

\[ \Lambda(x) = \mu e^{\frac{m^2}{2} - 1} \]

For the reverse current \( Q(t) \), with the same statistics (33) but with the parameters \( v \) in place of \( \mu \) and \( -m \) in place of \( -m \), we obtain

\[ \Lambda_{v}(x) = \mu e^{\frac{m^2}{2} - 1} \]

It then follows from the FDR (28) (if we introduce the dependence of \( n \) and \( \Lambda \) on \( x \)) that

\[ D(x) = \Delta_x(x) \]

\[ I(x) = g(x)(e^{m^2} - 1) \]

If \( \Lambda \) does not depend on \( x \), then we obtain a VAC which differs from (32) by the correction factor \( 1/(v + 1) \) in the argument of the exponential. This factor is actually present in the range from \( 1 \) to \( 1 \). Its origin can thus be connected with the correlations of the elementary acts of charge transport.

We now consider the relation between the average current \( \bar{I} \) and the spectral density of the current fluctuations (on the zeroth part) of \( D \). As \( x \to 0 \), we have

\[ D = \gamma \delta(t) \]

i.e., the equilibrium fluctuation-dissipation relation. At

\[ \gamma \delta(t) \]

i.e., in the range of shot noise, we have

\[ D = \gamma \delta(t) \]

Consequently, \( \gamma (1/2 - 1) \gamma \delta(t) \) plays simultaneously the role of the depression factor of the shot noise.

What is the nature of the considered correlation effect? We note that the correlation is negative (at \( \gamma > 0 \)): it decreases the current noise and increases the electrical resistance (however, it does not, of course, disturb the equilibrium FDR). One should obviously connect such a negative correlation with the Pauli principle and with the Fermi statistics of the charges. Then the parameter \( \gamma > 0 \) is smaller the larger the number of free levels onto which the particle moves. With decrease in the number of free levels, the Poisson-statistics model becomes unsuitable (see, for example, Ref. 9, where this case corresponds to a high injection level). Positive correlation \( \gamma < 0 \), which is characteristic for Bose particles, would have led to opposite effects—increase of the noise intensity and decrease of the resistance.

**APPENDIX**

1. Any functional of two trajectories \( u(t) \), \( v(t) \) can be uniquely represented in the form

\[ \langle u(t); v(t) \rangle = \int \langle u(t); v(t) \rangle \delta(t) \delta(t) \]

where the matrix-functionals \( \Gamma_x^{(2)} \) depend only on \( u(t) \) and \( v(t) \) at \( \delta < t < t \), i.e., on the end segments of the trajectories. Now let \( \Gamma_x^{(2)} \) be the characteristic functional of the currents. By virtue of its definition, we have the equality

\[ B(0, x(0)) = 0 \]

from which, in view of the arbitrariness of \( v(t) \), it follows that

\[ B(0, x(0)) = 0 \]

(\( A.2 \))

The causality condition formulated above means that

\[ B(0, x(0)) = 0 \]

From this and from (A.2) we conclude that

\[ \Gamma_x^{(2)}(u(t), v(t)) = 0 \]

Furthermore, \( a(t) \) is the mean value of the currents at equilibrium; therefore,

\[ a(t) = \frac{1}{2} \langle Q(t) \rangle = 0 \]

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Applying the FDR (2) to (A.3) we obtain, after elementary transformations,

\[ \Gamma_{\alpha}(t; \tau) \equiv \Gamma_{\alpha}^{(\alpha)}(t; t) = -\frac{1}{2} \int d\Sigma \left( \Gamma_{\alpha}^{(\alpha)}(t; \tau) \right) f(t; \tau) \]

\[ \Rightarrow \Gamma_{\alpha}^{(\alpha)}(t; t) = -\frac{1}{2} \int d\Sigma \left( \Gamma_{\alpha}^{(\alpha)}(t; \tau) \right) f(t; \tau) \]

\[ \Gamma_{\alpha}(t; \tau) = -\frac{1}{2} \int d\Sigma \left( \Gamma_{\alpha}^{(\alpha)}(t; \tau) \right) f(t; \tau) \]

whence follows the representation (5).

2. We now consider the derivation of Eq. (12). Integrating (10) with respect to \( I \), we obtain, as a consequence of the normalization condition,

\[ \left\{ \exp \left[ -\frac{1}{2} \int d\Sigma \left( \Gamma_{\alpha}^{(\alpha)}(t; \tau) \right) f(t; \tau) \right] \right\}_{\alpha AN} \rightarrow \exp \left\{ \sum_{\alpha} \frac{(-1)^{\alpha}}{\alpha!} \left( E^{(\alpha)} - E^{(\alpha)} \right) \right\} \]

\[ = \exp \left[ \sum_{\alpha} \frac{(-1)^{\alpha}}{\alpha!} \left( E^{(\alpha)} - E^{(\alpha)} \right) \right] \]

Dividing (10) by this relation, we obtain

\[ \exp \left[ -\frac{1}{2} \int d\Sigma \left( \Gamma_{\alpha}^{(\alpha)}(t; \tau) \right) f(t; \tau) \right] \rightarrow \exp \left[ \sum_{\alpha} \frac{(-1)^{\alpha}}{\alpha!} \left( E^{(\alpha)} - E^{(\alpha)} \right) \right] \]

which is identical with (12).

We note that the formulas similar to (10) follow automatically from (1), not only for currents, but also for any set of variables \( \phi \) (for example, \( \phi \) can represent the set of microscopic variables of some subsystem):

\[ \exp \left[ \sum_{\alpha} \frac{(-1)^{\alpha}}{\alpha!} \left( E^{(\alpha)} - E^{(\alpha)} \right) \right] \]

where \( E \) is equal to (A.3). It is not difficult to generalize this formula and (10) to the case of arbitrarily varying forces.

3. We consider the symmetry formulas (14) for the kinetic operator of the currents. We set \( x(t) = 0 \) in (1) at \( T > t \) and integrate (1) over all trajectories \( (T) \) with fixed currents \( I, I, I \) with the probability density of the transition from \( I \) to \( I \), we obtain

\[ V_{\alpha}(I; I) \equiv V_{\alpha}^{(I)}(I; I) \rightarrow \exp \left[ -\frac{1}{2} \int d\Sigma \left( \Gamma_{\alpha}^{(\alpha)}(t; \tau) \right) f(t; \tau) \right] V_{\alpha}(I; I) \]

\[ \Rightarrow V_{\alpha}(I; I) = \exp \left[ -\frac{1}{2} \int d\Sigma \left( \Gamma_{\alpha}^{(\alpha)}(t; \tau) \right) f(t; \tau) \right] V_{\alpha}(I; I) \]

where the angle brackets have the meaning of the conventional mean value under the condition that \( I(0) \) and \( I(t) \) are given.

If the external forces satisfy only the condition \( x(t) = 0 \) at \( t < 0 \), then the formula

\[ \exp \left[ -\frac{1}{2} \int d\Sigma \left( \Gamma_{\alpha}^{(\alpha)}(t; \tau) \right) f(t; \tau) \right] V_{\alpha}(I; I) \]

\[ \Rightarrow V_{\alpha}(I; I) = \exp \left[ -\frac{1}{2} \int d\Sigma \left( \Gamma_{\alpha}^{(\alpha)}(t; \tau) \right) f(t; \tau) \right] V_{\alpha}(I; I) \]

\[ \Rightarrow V_{\alpha}(I; I) = \exp \left[ -\frac{1}{2} \int d\Sigma \left( \Gamma_{\alpha}^{(\alpha)}(t; \tau) \right) f(t; \tau) \right] V_{\alpha}(I; I) \]

can be derived from (1) in analogy with (A.6). Here, however, compared to (A.6), as is seen, we have used the assumption of Markov currents, and \( W_{\alpha}(z; \alpha; \tau) \) denotes (for the process with time reversal) the non-equilibrium current distribution \( I(0) = 0 \) after the action of the forces \( \alpha(0 - \tau) \). From (A.7) and the formula (10) (generalized to variable forces), we obtain

\[ V_{\alpha}(I; I) = \exp \left[ -\frac{1}{2} \int d\Sigma \left( \Gamma_{\alpha}^{(\alpha)}(t; \tau) \right) f(t; \tau) \right] V_{\alpha}(I; I) \]

Evidently, the right-hand side of this equation does not depend on \( x(\tau) \) at \( t > \tau \). Consequently, the left-hand side does not depend on the reversed trajectory of the forces \( \alpha(t - \tau) \) at \( t < 0 \) and the transition probability \( V_{\alpha}(I; I) \) is determined only the value of \( x(\tau) \) at \( t > \tau > 0 \). This means that the kinetic current operator has an instantaneous dependence on the forces.

Therefore, the time symmetry of the transition probability can be considered with the help of the relation (A.6). For the transition to the kinetic operator, we need to take the infinitesimal form of (A.6) at \( t = 0 \), \( x(\tau) = x \):

\[ e^{-\tau \tau} W_{\alpha}(I; I) \rightarrow \exp \left[ -\frac{1}{2} \int d\Sigma \left( \Gamma_{\alpha}^{(\alpha)}(t; \tau) \right) f(t; \tau) \right] W_{\alpha}(I; I) \]

Multiplying (A.8) by an arbitrary function \( f(t) \) and integrating over \( I \), we obtain, at \( I = I \),

\[ L] = \tau \tau = \tau \tau \rightarrow \exp \left[ -\frac{1}{2} \int d\Sigma \left( \Gamma_{\alpha}^{(\alpha)}(t; \tau) \right) f(t; \tau) \right] W_{\alpha}(I; I) \]

This equation is equivalent to the operator equation (14).
Interest in the so-called one-dimensional and two-dimensional systems has increased of late in connection with searches for high-temperature superconductivity and superfluidity. Electronic phenomena in systems that are close to two-dimensional were investigated in inversion layers of silicon in metal-insulator-semiconductor structures. In these structures it is easy to control the carrier density by an external field, but it is difficult to obtain identical oxide layers, and this introduces an uncertainty in the interpretation of the obtained data that characterize a two-dimensional system. A more reliable model of a two-dimensional system, in our opinion, consists of highly conducting layers adjacent to the cleavage planes of germanium bicrystals. They are formed at a junction of single crystals and are characterized by a sufficiently well-ordered structure, as confirmed by the small scatter of the carrier densities and mobilities in these layers, as obtained in various laboratories of the world.  

1. PREPARATION OF BICRYSTALS  

The germanium bicrystals were grown by the Czochralski method on a double seed crystal by a method similar to that described in Ref. 2. The double seed was prepared by cutting a single-crystal ingot into two at a specified inclination angle 6/2 to the [100] axis,