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 [Soviet 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 a nonequilibrium stationary state 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,[1] 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 semiphenomenological 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.[2] 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 $x(t)=x$ 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 equilibuium 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 currents and for the probability functionals—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 thermostat) 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.[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

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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 $x(t)$ in thermodynamic equilibrium start to act on a system and alter the Hamiltonian of the system:

$$H(t)=H_0-x(t)Q=H_0-x_\alpha(t)Q_\alpha,$$

where Q_α 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 τ, 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 τ_0 (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 $I(t)=\dot{Q}(t)$, 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. This transition is actually effected only conceptually, and reduces to the assumption that certain integrals

$$\int_0^\infty \langle I_\alpha(t), I_\alpha(0)\rangle dt$$

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.

1. We denote by $P[I(\tau); x(\tau)]$ 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:[2)]

$$P[I(\tau);x(\tau)]\exp\left\{-\beta\int I(\tau)x(\tau)d\tau\right\} =P[-\varepsilon I(-\tau);\ \varepsilon x(-\tau)]. \quad (1)$$

Here $\varepsilon_k=\pm1$ depending on the temporal parity of the macrovariable Q_k, $\beta\equiv 1/T$, T is the temperature of the thermostat contained in the considered system. For the characteristic functional of the currents

$$B[u(\tau);x(\tau)]=\ln\left\langle \exp\left\{\int u(\tau)I(\tau)d\tau\right\}\right\rangle$$

$$=\sum_{n=1}^\infty \frac{1}{n!}\int\ldots\int \langle I(\tau_1),I(\tau_2),\ldots,I(\tau_n)\rangle_{x(\tau)}u(\tau_1)\ldots u(\tau_n)d\tau_1\ldots d\tau_n,$$

the formula equivalent to (1) has the form

$$B[u(\tau)-\beta x(\tau);\ x(\tau)]=B[-\varepsilon u(-\tau);\ \varepsilon x(-\tau)]. \quad (2)$$

The angular brackets with comma inside (Malakhov's cumulant brackets[5)]) denote the cumulant functions, for example, $\langle A,B\rangle\equiv\langle AB\rangle-\langle A\rangle\langle B\rangle$.

In an open system, certain macrovariables[3)] $Q(t)$ experience diffusion. We introduce the diffusion coefficients $D_n(x)$ at constant forces by means of the generating relation

$$\sum_{n=1}^\infty \frac{u^n}{n!}D_n(x)=\lim_{t\to\infty}\frac{1}{t}\ln\langle e^{u\Delta Q(t)}\rangle_x,$$

$$\Delta Q(t)=Q(t)-Q(0).$$

By properly choosing the trial function $u(\tau)$ in (2), we obtain the FDR

$$\sum_{n=1}^\infty \frac{(-\beta x)^n}{n!}D_n(x)=0,\qquad (-\varepsilon)^n D_n(\varepsilon x)=\sum_{k=0}^\infty \frac{(-\beta x)^k}{k!}D_{n+k}(x). \quad (3)$$

We note that in the multidimensional case the tensors $D_n(x)$ are completely symmetric (this follows from their definition) and that $D_1(x)\equiv\bar{I}(x)$ is the vector of the average values of the currents.

We consider the differential form of these relations. Transforming to tensor notation, we introduce

$$D_{\alpha\ldots\beta}^{\gamma\ldots\delta}\equiv\left(\frac{\partial}{\partial x_\gamma}\ldots\frac{\partial}{\partial x_\delta}D_{\alpha\ldots\beta}(x)\right)_{x=0}$$

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

$$D_\alpha^\beta\equiv\left(\frac{\partial \bar{I}_\alpha}{\partial x_\beta}\right)_{x=0}=\frac{1}{2T}D_{\alpha\beta}=\frac{1}{2T}\left\{\int_0^\infty\langle I_\alpha(t),I_\beta(0)\rangle_0 dt +\int_0^\infty\langle I_\alpha(0),I_\beta(t)\rangle_0 dt\right\}$$

at $\varepsilon_\alpha\varepsilon_\beta=+1$ and

$$D_{\alpha\beta}=0,\quad D_\alpha^\beta=-D_\beta^\alpha$$

at $\varepsilon_\alpha\varepsilon_\beta=-1$. This leads to the Onsager-Casimir relations

$$(\partial\bar{I}_\alpha/\partial x_\beta)_{x=0}=\varepsilon_\alpha\varepsilon_\beta(\partial\bar{I}_\beta/\partial x_\alpha)_{x=0}. \quad (4)$$

We write down the analogous three-index formulas: at $\varepsilon_\alpha\varepsilon_\beta\varepsilon_0=1$

$$D_{\alpha\beta\gamma}=0,\quad D_\alpha^{\beta\gamma}=\frac{1}{2T}(D_{\alpha\beta}^{\gamma}+D_{\alpha\gamma}^{\beta}),$$

and at $\varepsilon_\alpha\varepsilon_\beta\varepsilon_\gamma=-1$,

$$D_{\alpha\beta}^{\gamma}=\frac{1}{2T}D_{\alpha\beta\gamma},\quad D_\alpha^{\beta\gamma}+D_\beta^{\alpha\gamma}+D_\gamma^{\alpha\beta}=\frac{3}{2T}D_{\alpha\beta\gamma}.$$

We also give the four-index relations: at $\varepsilon_\alpha\varepsilon_\beta\varepsilon_\gamma\varepsilon_\delta=1$,

$$D_{\alpha\beta\gamma}{}^{\delta}=\frac{1}{2T}D_{\alpha\beta\gamma\delta},\quad D_{\alpha}{}^{\beta\gamma\delta}=\frac{1}{2T}(D_{\alpha\beta}{}^{\gamma\delta}+D_{\alpha\gamma}{}^{\beta\delta}+D_{\alpha\delta}{}^{\beta\gamma})-\frac{1}{4T^3}D_{\alpha\beta\gamma\delta},$$

and at $\varepsilon_\alpha\varepsilon_\beta\varepsilon_\gamma\varepsilon_\delta=-1$,

$$D_{\alpha\beta\gamma\delta}=0,\quad D_{\alpha\beta}{}^{\gamma\delta}=\frac{1}{2T}(D_{\alpha\beta\gamma}{}^{\delta}+D_{\alpha\beta\delta}{}^{\gamma}),$$

$$(D_{\alpha}{}^{\beta\gamma\delta}+D_{\beta}{}^{\alpha\gamma\delta}+D_{\gamma}{}^{\alpha\beta\delta}+D_{\delta}{}^{\alpha\beta\gamma})=\frac{1}{2T^2}(D_{\alpha\beta\gamma}{}^{\delta}+D_{\alpha\beta\delta}{}^{\gamma}+D_{\alpha\gamma\delta}{}^{\beta}+D_{\beta\gamma\delta}{}^{\alpha}).$$

The basic unfavorable consequence of the obtained FDR is that the nonlinear transport coefficients

$$D_{\alpha}^{\beta\ldots\gamma}=\left(\frac{\partial \bar I_\alpha}{\partial x_\beta\ldots\partial x_\gamma}\right)_{x=0}$$

are not connected by any universal symmetry formulas similar to (4), i.e., not containing the diffusion coefficients. Consequently, the nonlinear crossing transport processes do not have to be reciprocal. This fact was first established by Stratonovich,[2,3] in the phenomenological Markov model of fluctuations of $Q(t)$, in which the formulas (3) are obtained if we identify $D_n(x)$ with the kinetic-operator coefficients that depend on x and Q and are averaged over the equilibrium distribution of Q. The absence of universal nonlinear reciprocity relations of course does not exclude the possibility of such relations for specific systems possessing dynamical symmetries.

2. We consider those additional limitations imposed on the structure of the characteristic functional of the currents by the causality condition: independence of the correlators $\langle I(t_1), I(t_2), \ldots, I(t_n)\rangle$ of $x(t)$ at $t\geqslant\max\{t_i\}$. As a result of simple analysis (see Appendix 1), we obtain

$$B[u(\tau);x(\tau)]=\int_{-\infty}^{\infty}dt\,u(t)\int_{-\infty}^{t}d\theta\,\Gamma_{t,\theta}[u(\tau);x(\tau)]\{u(\theta)+\beta x(\theta)\},\tag{5}$$

where the tensor functional $\Gamma_{t,\theta}$ posseses the following properties:

1) it depends only on $u(\tau)$ and $x(\tau)$ at $t>\tau>\theta$;

2) $\Gamma_{t,\theta}[u(\tau)-\beta x(\tau);x(\tau)]=\varepsilon\Gamma^{+}_{-\theta,-t}[-\varepsilon u(\tau);\varepsilon x(-\tau)]\varepsilon$;

3) $\beta\int_{-\infty}^{t}d\theta\,\Gamma_{t,\theta}[0;x(\tau)]x(\theta)=\bar I^t[x(\tau)]$.

Here $\bar I^t$ is the mean value of the current vector at the time t.

The most important first property means that the functional $\Gamma_{t,\theta}$ is constructed from quasi-equilibrium correlators that correspond to the cut-off trajectory of the external forces $x(\tau)\eta(\tau-\theta)$, where $\eta(\tau)$ is the unit step function:

$$\Gamma_{t,\theta}[u(\tau);x(\tau)]=\langle I(t),I(\theta)\rangle_{x(\tau)\eta(\tau-\theta)}+\int_{\theta}^{t}dt'\langle I(t),I(t'),I(\theta)\rangle_{x(\tau)\eta(\tau-\theta)}u(t')+\ldots.$$

From this formula and (5) we obtain the relations between the real and the quasi-equilibrium cumulant functions[4)]:

$$\bar I^t[x(\tau)]=\beta\int_{-\infty}^{t}\langle I(t),I(\theta)\rangle_{x(\tau)\eta(\tau-\theta)}x(\theta)\,d\theta,$$

$$\langle I(t_1),\ldots,I(t_n),I(t)\rangle_{x(\tau)}=\langle I(t_1),\ldots,I(t_n),I(t)\rangle_{x(\tau)\eta(\tau-t)}+\beta\int_{-\infty}^{t}\langle I(t_1),\ldots,I(t_n),I(t),I(\theta)\rangle_{x(\tau)\eta(\tau-\theta)}x(\theta)\,d\theta\quad(t_i\geqslant t).\tag{6}$$

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

$$\bar I_\alpha(x)=\sum_\beta\Gamma_{\alpha\beta}(x)x_\beta,\quad \Gamma_{\alpha\beta}(x)=\beta\int_0^\infty\langle I_\alpha(t),I_\beta(0)\rangle_{x\eta(\tau)}dt.\tag{7}$$

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

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

$$\Gamma_{\alpha\beta}(x)=\Gamma_{\alpha\beta}'(x)+\Gamma_{\alpha\beta}''(x),$$
$$\Gamma_{\alpha\beta}'(x)=-\varepsilon_\alpha\varepsilon_\beta\Gamma_{\alpha\beta}'(\varepsilon x),\quad \Gamma_{\alpha\beta}''(x)=\varepsilon_\alpha\varepsilon_\beta\Gamma_{\alpha\beta}''(\varepsilon x).\tag{8}$$

The (irreversible) transport coefficients $\Gamma''_{\alpha\beta}$ proper can be expressed, by using again the property 2) and formula (3), in terms of the even diffusion coefficients D_{2n}:

$$N''(x)=\sum_{\alpha,\beta}\Gamma_{\alpha\beta}''(x)x_\alpha x_\beta=\sum_{n=1}^{\infty}C_n\left(\frac{\beta}{2}\right)^{2n-1}\frac{1}{2}\{D_{2n}(x)x^{2n}+D_{2n}(\varepsilon x)(\varepsilon x)^{2n}\},\tag{9}$$

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

$$\operatorname{th} z=\sum_{n=1}^{\infty}C_n z^{2n-1}.$$

As for the inverse coefficients $\Gamma'_{\alpha\beta}$ that are not connected through the FDR with the fluctuation characteristics 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.[5)] Since the $\Gamma'_{\alpha\beta}$ describe the redistribution of the energy among the macroscopic degrees of freedom, but not the energy dissipation, the corresponding power $N'(x)\equiv\Sigma\Gamma'_{\alpha\beta}(x)x_\alpha x_\beta$ should be equal to zero. Then the power absorbed by the system from the external source in the SNS is given by the expression (9). If the SNS has come about in the evolution of the system from an initial equilibrium state, then, in correspondence with the results of Ref. 1, this power is non-negative: $N''(x)\geqslant 0$. Consideration of the more general case (in which the system is in nonequilibrium state prior to the switching-on of the forces $x(t)$ goes beyond the framework of this paper.

3. We derive below the formula that connects the probability distribution functions of the currents at equilibrium, $W_0(I)$, and in the non-equilibrium stationary state, $W_x(I)$. For this purpose, we set $x(\tau)=x\eta(\tau)$ in (1) (strictly speaking, we should take $x(\tau)=e^{-\mu\tau}x\eta(\tau)$, and then take to the limit as $\mu\to 0$), multiply (1) by $\delta[I(0)-I]$, and integrate over the trajectories (we shall not define strictly this operation, which is symbolic in our case, since formula (1) can always be replaced by well-defined expressions for the averages). As a result, we obtain

$$W_0(I)\left\langle\exp\left\{-\beta\int_0^\infty xI(t)\,dt\right\}\right\rangle^{I}_{x\eta(\tau)}=W_{\varepsilon x}(-\varepsilon I),$$
$$W_0(-\varepsilon I)=W_0(I),\tag{10}$$

where $\langle\ldots\rangle^{I}_{x\eta(\tau)}$ denotes the conditional mean value (under the condition that at the time $t=0$ we have $I(0)=I$ and prior to this moment the system had been in equilibrium). This formula expresses the distribution of the currents in SNS in terms of the equilibrium distribution and the cumulant power function $N(t)\equiv x(t)I(t)$ following a stepwise (instantaneous) switching-on of the external forces. Although the energy absorbed by the system

$$E(t)=\int_0^t N(\tau)d\tau$$

has a diffusion behavior, the mean value of the exponential in (10) is finite, since the contributions from $\langle N(\tau)\rangle$ (which increase with t) to the upper cumulant power functions cancel mutually. The explicit finite expression for this exponential can be obtained by transforming it with the help of the FDR and going over to the characteristic function

$$\theta_x(u)=\int e^{uI}W_x(I)dI.$$

The result has the form

$$\theta_x(u)=\theta_0(u)\exp\left\{\beta\sum_{k=1}^{\infty}\frac{1}{k!}u^k\int_0^{\infty}dt\langle I^{(k)}(t),I(0)\rangle_{x\eta(\tau)}x\right\},$$

where

$$\langle A^{(k)},B\rangle\equiv\langle\underbrace{A,A,\ldots,A}_{k},B\rangle.$$

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

It is evident from what has been said that the nonequilibrium statistical characteristics, which describe the response to the stepwise external action, play a special role in the theory. We have called these quasi-equilibrium in the foregoing. They form a set that is closed relative to the FDR and all the remaining characteristics are expressed in their terms by means of the FDR.

In order to give the formula (10) a more "physical" interpretation, we note that it can be rewritten in the form

$$W_{\varepsilon x}(-\varepsilon I)=W_0(I)\exp\{-\beta\Delta E(I;\ x)\},\tag{11}$$

where $\Delta E(I,x)$ has the sense of the average work which is performed by the external forces against the fluctuation currents and is produced by the deviation of $I=I(0)$ from $\bar{I}(x)$. If τ_0 is the characteristic time of damping of the fluctuation currents in the SNS, then

$$\Delta E(I,x)\approx\tau_0 N(I,x),$$

where $N(I,x)$ is determined by the power dissipated in the SNS. In order to verify this, we assume that (approximately) the second and higher conditional cumulant functions of the current in (10) do not depend on the initial condition[6] $I(0)=I$. We then have (see Appendix, Sec. 2)

$$W_{\varepsilon x}(-\varepsilon I)\sim W_0(I)\exp\left\{-\beta\int_0^{\infty} x[\langle I(t)\rangle^{I}_{x\eta(\tau)}-\bar{I}(x)]\right\}.\tag{12}$$

Here the argument of the exponential contains the energy transferred from the force source to the thermostat in the process of damping of the macroscopic fluctuations. If the damping law is approximately exponential, i.e., if $\dot{I}(t)=-\lambda[I-\bar{I}(x)]$, $\lambda=\lambda(x)$ then we obtain from (12)

$$W_x(I)\sim W_0(I)\exp\{\beta x\varepsilon\lambda^{-1}\varepsilon I\}.\tag{13}$$

In the general case, $\Delta E(I,x)$ contains also contributions from the higher cumulants in (10), which are connected with $\langle I(t)\rangle$ by virtue of the nonlinearity.[7]

3. FLUCTUATION-DISSIPATION RELATIONS IN THE MARKOV THEORY OF FLUCTUATIONS IN OPEN SYSTEMS

1. The Markov assumption is sufficient for the construction of a closed theory (i.e., a theory which allows us to calculate all the statistical characteristics of the processes). This assumption is equivalent to the generalized Onsager hypothesis, since in the Markov theory one and the same kinetic operator describes both the local properties of the fluctuations and the global irreversible behavior.

If the system considered is an open one, then as the Markov set should be chosen to be the finite macrovariables and the currents corresponding to the diffusion macrovariable, i.e., variables which experience stationary fluctuations under fixed external conditions. For simplicity, we consider only the case in which only the diffusion variables are important (the mixed case adds nothing new to this). We shall also assume, as has already been done implicitly above, that the diffusion is homogeneous [only the increases $\Delta Q(t)$ are important here and not the absolute values of $Q(t)$]. The various generalizations are made in elementary fashion.

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

$$L\equiv L\left(x,I,\frac{\partial}{\partial I}\right)=\sum_{n=1}^{\infty}\frac{1}{n!}\left(-\frac{\partial}{\partial I}\right)^n K_n(x,I),$$

$$L^+\equiv L^+\left(x,I,\frac{\partial}{\partial I}\right)=\sum_{n=1}^{\infty}\frac{1}{n!}K_n(x,I)\left(\frac{\partial}{\partial I}\right)^n\quad(x=x(t)).$$

The operator equation

$$L\left(x,I,\frac{\partial}{\partial I}\right)W_0(I)=W_0(I)\left\{L^+\left(\varepsilon x,-\varepsilon I,-\varepsilon\frac{\partial}{\partial I}\right)+\beta xI\right\}\tag{14}$$

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

$$W_0(I)\varepsilon^n K_n(\varepsilon x,-\varepsilon I)=\sum_{m=0}^{\infty}\frac{1}{m!}\left(-\frac{\partial}{\partial I}\right)^m K_{n+m}(x,I)W_0(I)\quad(n\geq1),$$
$$LW_0(I)=\beta xIW_0(I).\tag{15}$$

Here W_0 is the equilibrium distribution of the currents at $x=0$. These relations enable us to eliminate "half" the parameters of the kinetic operator—to express the coefficients K_n in terms of their symmetrized combinations:

$$K_n(x,I)=K_n'(x,I)+\frac{1}{W_0(I)}$$
$$\times\sum_{m=1}^{\infty}C_m\left(\frac{1}{2}\frac{\partial}{\partial I}\right)^{2m-1}K'_{n+2m-1}(x,I)W_0(I),\tag{16}$$
$$K_n'(x,I)\equiv{}^1/_2[K_n(x,I)+\varepsilon^n K_n(\varepsilon x,-\varepsilon I)]\quad(n\geq0),$$
$$K_0(x,I)\equiv-{}^1/_2\beta xI.$$

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

$$L\left(x,I,\frac{\partial}{\partial I}\right)=\frac{1}{2}\beta xI+\sum_{n+m\geq 1}\left(\frac{\partial}{\partial I}\right)^n S_{nm}$$
$$\times K'_{n+m}(x,I)W_0(I)\left(\frac{\partial}{\partial I}\right)^m\frac{1}{W_0(I)},\qquad(17)$$

where the number S_{nm} (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:

$$\sum_{n,m=0}^{\infty}S_{nm}u^n v^m=2(e^u+e_v)^{-1}.$$

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 $n\geq 3$ and the kinetic equation transforms into the Fokker-Planck equation. Formulas (15)–(17) reduce to the form

$$K_1(x,I)=K_1'(x,I)+\frac{1}{2W_0(I)}\frac{\partial}{\partial I}K_2'(x,I)W_0(I),\qquad(18)$$
$$K_2(x,I)=K_2'(x,I);$$

$$\beta xI+\frac{1}{W_0(I)}\frac{\partial}{\partial I}K_1'(x,I)W_0(I)=0;\qquad(19)$$

$$L=-\frac{\partial}{\partial I}K_1'(x,I)+\frac{1}{2}\frac{\partial}{\partial I}K_2'(x,I)W_0(I)\frac{\partial}{\partial I}\frac{1}{W_0(I)}.\qquad(19')$$

The functions K_1' 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 (describing the interaction of the macrovariables with the thermostat) terms. In the same fashion, the operator (17) splits into parts. The dependence of K'_n on x at $n\geq 2$ indicates that the external forces, generally speaking, influence the state of the thermostat. However, the FDR (15) admit of such a possibility when this influence is absent and $K_n(x, I)=K_n(0, I)\equiv K_n(I)$, $n\geq 2$. In this simplest case, formula (19) follows from (17) as well as the equality

$$L=-\frac{\partial}{\partial I}[K_1'(x,I)-K_1'(0,I)]+L_0,$$
$$L_0=L_0\left(I,\frac{\partial}{\partial I}\right)=L\left(0,I,\frac{\partial}{\partial I}\right).\qquad(20)$$

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

$$K_1'(x,I)=K_1'(0,I)+A(I)x.$$

Here

$$L=-\frac{\partial}{\partial I}A(I)x+L_0,\quad \beta I_\alpha+\sum_\gamma\frac{1}{W_0(I)}\frac{\partial}{\partial I_\gamma}A_{\gamma\alpha}(I)W_0(I)=0.\qquad(20')$$

2. 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 $L\equiv L(t)$ depends on time through the force. If the perturbation acts over a finite interval, then

$$\bar I(t)=\langle I(t)\rangle=\int dI\,I\,\overleftarrow{\exp}\left\{\int_{-\infty}L(\tau)d\tau\right\}W_x(I),$$

where $\overleftarrow{\exp}$ is the chronologically ordered exponential. We then find for the linear response in the case (20′)

$$\frac{\delta I_\alpha}{\delta x_\gamma(0)}=\int dI\,I_\alpha e^{tL}\frac{\partial L}{\partial x_\gamma}W_x(I)$$
$$=\beta\langle I_\alpha(t),I_\gamma\rangle_x-\sum_{\alpha'}\left\langle I_\alpha(t),A_{\alpha'\gamma}(I)\frac{\partial}{\partial I_{\alpha'}}\ln V(x,I)\right\rangle_x,\qquad(21)$$
$$I=I(t=0),\quad W_x(I)=W_0(I)V(x,I).$$

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

$$A^+(I)\frac{\partial}{\partial I}\ln V(x,I)=\text{const}+b(x)I,\qquad(22)$$

we get from (21) a modernized FDT

$$\frac{\delta}{\delta x_\gamma(0)}I_\alpha(t)=(\beta-b(x))\langle I_\alpha(t),I_\gamma\rangle_x\qquad(23)$$

with an effective temperature that depends on the external forces. The equation for the stationary distribution function $LW_x(I)=0$ reduces then to the form

$$L_0W_x(I)+(\beta-b(x))(Ix-N(x))W_x(I)=0,\quad N(x)=\bar I(x)x.\qquad(24)$$

It can then be concluded that (for the proof see the Appendix, Sec. 4)

$$[\beta-b(x)]N(x)=\sum_{n=1}^{\infty}\frac{1}{n!}D_n(0)[\beta-b(x)]^n x^n,\qquad(24')$$

i.e., the power dissipated in the SNS is expressed in terms of the equilibrium diffusion coefficients and the effective temperature $T(x)\equiv[\beta-b(x)]^{-1}$. It follows from (23) that

$$\frac{\partial}{\partial x_\gamma}\bar I_\alpha(x)=\frac{1}{T(x)}\int_0^\infty\langle I_\alpha(t),I_\gamma\rangle_x dt,\quad \frac{\partial}{\partial x_\gamma}\bar I_\alpha+\frac{\partial}{\partial x_\alpha}\bar I_\gamma=\frac{1}{T(x)}D_{\alpha\gamma}(x).$$

Consequently, the temperature $T(x)$ of the macrovariables in the nonequilibrium system with properties (22), (23) can be determined, in analogy with 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

$$W_x(I)\sim\frac{1}{A(I)}\exp\left\{-\frac{1}{T(x)}\left[\int_0^I\frac{y\,dy}{A(y)}-\bar I(x)\int_0^I\frac{dy}{A(y)}\right]\right\}.\qquad(25)$$

Definite conditions follow then from this and from (24) on the kinetic operator L_0. In particular, if L_0 is the Fokker-Planck operator, then it is completely determined by the function $A(I)$. For the case $\varepsilon=1$, we find, with account taken of the FDR.

$$L=\frac{\partial}{\partial I}\lambda(I)(I-gx)+Tg\left(\frac{\partial}{\partial I}\lambda(I)\right)^2,$$
$$A(I)=A(-I)=g\lambda(I),\quad T(x)=T,\quad \bar I(x)=gx,\quad g=\text{const}.\qquad(26)$$

In the case $\varepsilon=-1$ (I is an even variable in time)

$$L=\frac{\partial}{\partial I}\lambda(I)\left\{(1-hx)I-gx+Tg\frac{\partial}{\partial I}\lambda(I)\left(1+\frac{h}{g}I\right)\right\},$$
$$A(I)=\lambda(I)(g+hI),\quad T(x)=\frac{T}{1-hx},\quad I(x)=\frac{gx}{1-hx},\quad h=\text{const.} \tag{27}$$

Here $[\lambda(I)]^{-1}$ 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 (29) 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_α by the relations $\partial I_\gamma/\partial P_\alpha=A_{\gamma\alpha}(I)$, which is always possible if (22) is satisfied. Then the non-equilibrium distribution $W_x(P)$ and the kinetic operator for the variables P have the form

$$W_x(P)\sim W_0(P)\exp\{\beta P_\alpha I_\alpha(x)\},$$
$$W_0(P)\sim\exp\left\{-\beta\int^P I dP\right\},\quad L=-x_\alpha\frac{\partial}{\partial P_\alpha}+L_0. \tag{27'}$$

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 $P(I)$ 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 formula for the variables Q in a closed equilibrium system[1]:

$$W_x(Q)\sim W_0(Q)\exp\{\beta Q_\alpha x_\alpha\}.$$

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

Thus, 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 non-equilibrium 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

$$D(u-x/T;\,x)=D(-u;\,x),$$
$$D(u;x)=\lim_{t\to\infty}\frac{1}{t}\langle\exp\{u[Q(t)-Q(0)]\}\rangle. \tag{28}$$

Obviously, $\exp\{tD(u;\,x)\}$ as a function of u is a characteristic function of an infinitely divisible distribution (see, for example, Ref. 7). Consequently, $D(u;\,x)$ can be represented in the form[8]

$$D(u;x)=u\bar{I}(x)+D(x)\int_{-\infty}^{\infty}\frac{e^{au}-1-au}{a^2}p(a,x)\,da, \tag{29}$$

where $p(a;\,x)$ is a nonnegative function normalized to unity; $\bar{I}=D_1$ and $D=D_2$ 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 dimensionalities 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 $[D(x)/a]p(a,x)\,da$. The following FDR result from (28) and (29):

$$p(-a;x)=e^{-ax/T}p(a;x),\quad \bar{I}(x)=\frac{D(x)}{2}\int_{-\infty}^{\infty}(1-e^{-ax/T})\frac{p(a,x)}{a}da;$$
$$\frac{x}{T}\bar{I}(x)=D(x)\int_{-\infty}^{\infty}\left(e^{-ax/T}-1+\frac{a}{T}x\right)\frac{p(a;x)}{a^2}da, \tag{30}$$

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 fluctuation characteristics of the transport process. Naturally, the description of this process in terms of only some of the diffusion coefficients $D_n(x)$ 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

$$p(a;\,x)=p(x)\delta(a-q)+(1-p(x))\delta(a+q).$$

We then find from (29), (30) (setting $z\equiv x/T$)

$$p(x)=(1-e^{-qz})^{-1},\quad \bar{I}=\frac{D(x)}{q}\frac{1-e^{-qz}}{1+e^{-qz}},$$
$$D(u;\,x)=n_1(x)(e^{qu}-1)+n_2(x)(e^{-qu}-1),$$
$$n_1(x)=\frac{D(x)}{q^2(1+e^{-qz})},\quad n_2(x)=e^{-qz}n_1(x). \tag{31}$$

Here n_1 and n_2 are the mean values of the charges traveling in the forward and reverse directions.

If $n_2(x)=n$ does not depend on x, as is the case for a semiconducting diode, we obtain an exponential volt-ampere characteristic (VAC):

$$\bar{I}(x)=qn(e^{xq/T}-1), \quad D(x)=q^2n(e^{xq/T}+1). \qquad (32)$$

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 ($p-n$ 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 t_k between direct transitions are independent, but are distributed not exponentially as in the Poisson case, but with a probability density

$$v_1(t)=\frac{\nu_1}{\gamma!}(\nu_1 t)^\gamma e^{-\nu_1 t} \quad (-1<\gamma<\infty). \qquad (33)$$

The average number of transitions per unit time is equal to $n_1\equiv\nu_1/(\gamma+1)$. We consider the distribution of the charge $Q_1(t)$ that passes in the forward direction. We introduce the function

$$\Delta_1(u)=\lim_{t\to\infty}\frac{1}{t}\ln\langle e^{u(Q_1(t)-Q_1(0))}\rangle.$$

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

$$s-\Delta_1(u)=0, \quad 1-e^{uq}v_1(s)=1-e^{uq}\int_0^\infty v_1(t)e^{-st}dt=0, \qquad (34)$$

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

$$\int_0^\infty \langle e^{u(Q(t)-Q(0))}\rangle e^{-st}\,dt.$$

Since in our case,

$$v_1(s)=(\nu/(\nu+s))^{\gamma+1},$$

we get from (34)

$$\Delta_1(u)=\nu_1(e^{qu/(\gamma+1)}-1)=n_1(\gamma+1)(e^{qu/(\gamma+1)}-1).$$

For the reverse current $Q_2(t)$, with the same statistics (33) but with the parameters ν_2 in place of ν_1 and $-q$ in place of q, we obtain

$$\Delta_2(u)=n_2(\gamma+1)(e^{-qu/(\gamma+1)}-1).$$

It then follows from the FDR (28) (if we introduce the dependence of n_1 and n_2 on x) that

$$D(u;x)=n_1(x)(\gamma+1)(e^{qu/(\gamma+1)}-1)+n_2(x)(\gamma+1)(e^{-qu/(\gamma+1)}-1),$$
$$n_2(x)=e^{-qx/(\gamma+1)}n_1(x), \qquad (35)$$
$$\bar{I}(x)=qn_1(x)(e^{qx/T(\gamma+1)}-1), \quad D(x)=q^2\frac{n_2(x)}{\gamma+1}(e^{qx/T(\gamma+1)}+1).$$

If n_2 does not depend on x, then we obtain a VAC which differs from (32) by the correction factor $1/(\gamma+1)$ in the argument of the exponential. This factor is actually present and varies in the range from $\frac{1}{2}$ to 1.[8] 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

$$D=2T(d\bar{I}/dx)_{x=0},$$

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

$$\exp\{qx/T(\gamma+1)\}\gg 1,$$

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

$$D(x)=(\gamma+1)^{-1}q\bar{I}(x).$$

Consequently, $1/(\gamma+1)$ plays simultaneously the role of the depression coefficient 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 become 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 $u(\tau)$, $x(\tau)$ can be uniquely represented in the form

$$B[u(\tau);x(\tau)]=\int_{-\infty}^{\infty}a(t)u(t)dt+\int_{-\infty}^{\infty}b(t)x(t)dt$$
$$+\int_{-\infty}^{\infty}dt\int_{-\infty}^{t}d\theta\{u(t)\Gamma^1_{t,\theta}[u(\tau);x(\tau)]u(\theta)+u(t)\Gamma^2_{t,\theta}[u(\tau);x(\tau)]x(\theta)$$
$$+x(t)\Gamma^3_{t,\theta}[u(\tau);x(\tau)]u(\theta)+x(t)\Gamma^4_{t,\theta}[u(\tau);x(\tau)]x(\theta)\}, \quad \text{(A.1)}$$

where the matrix-functionals $\Gamma^i_{t,\theta}$ depend only on $u(\tau)$, and $x(\tau)$ at $\theta<\tau<t$, i.e., on the end segments of the trajectories. Now let (A.1) be the characteristic functional of the currents. By virtue of its definition, we have the equality

$$B[0;x(\tau)]=0,$$

from which, in view of the arbitrariness of $x(\tau)$, it follows that

$$b(t)=0, \quad \Gamma^4_{t,\theta}[0;x(\tau)]=0. \qquad \text{(A.2)}$$

The causality condition formulated above means that

$$\left[\frac{\delta}{\delta x(\tau)}\frac{\delta}{\delta u(\tau_1)}\cdots\frac{\delta}{\delta u(\tau_n)}B\right]_{u=0}=0 \quad \text{at} \quad \tau>\max\{\tau_i\}.$$

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

$$\Gamma^3_{t,\theta}[u(\tau);x(\tau)]u(\theta)+\Gamma^4_{t,\theta}[u(\tau);x(\tau)]x(\theta)=0.$$

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

$$a(t)=\frac{d}{dt}\langle Q(t)\rangle_0=0.$$

Thus Eq. (A.1) takes the form

$$B[u(\tau);x(\tau)]=\int_{-\infty}^{\infty}dt\int_{-\infty}^{t}d\theta\,u(t)\{\Gamma^1_{t,\theta}[u(\tau);x(\tau)]u(\theta)+\Gamma^2_{t,\theta}[u(\tau);x(\tau)]x(\theta)\}. \quad (A.3)$$

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

$$\Gamma^2_{t,\theta}[u(\tau);x(\tau)]=\beta\Gamma^1_{t,\theta}[u(\tau);x(\tau)]\equiv\beta\Gamma_{t,\theta}[u(\tau);x(\tau)],$$

$$\Gamma_{t,\theta}[u(\tau)-\beta x(\tau);x(\tau)]=\varepsilon\Gamma^{+}_{-\theta,-t}[-\varepsilon u(-\tau);\varepsilon x(-\tau)],$$

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,

$$\left\langle\exp\left\{-\beta\int_0^\infty xI(t)\,dt\right\}\right\rangle_{x\eta(\tau)}=\exp\left\{\sum_{n=1}^{\infty}\frac{(-\beta)^n}{n!}\langle E^{(n)}\rangle\right\}=1, \quad (A.4)$$

$$E=\int_0^\infty xI(t)\,dt.$$

Dividing (10) by this relation, we obtain

$$W_{\varepsilon x}(-\varepsilon I)=W_0(I)\exp\left\{\sum_{n=1}^{\infty}\frac{(-\beta)^n}{n!}(\langle E^{(n)}\rangle^I-\langle E^{(n)}\rangle)\right\}. \quad (A.5)$$

Assuming that the higher cumulants $\langle E^{(n)}\rangle^I$, $n\geq 2$, which characterize the fluctuations of the work, are determined largely by the thermostat and depend weakly on the initial condition, we can write approximately

$$W_{\varepsilon x}(-\varepsilon I)\sim W_0(I)\exp\{-\beta(\langle E\rangle^I-\langle E\rangle)\},$$

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 ψ (for example, ψ can represent the set of microscopic variables of some subsystem):

$$W_{\varepsilon x}(\varepsilon^\psi\psi)=W_0(\psi)\langle\exp\{-\beta E\}\rangle^{\psi}_{x\eta(\tau)},$$

where E is equal to (A.4). 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 $x(\tau)=0$ in (1) at $\tau>t$ and integrate (1) over all trajectories $I(\tau)$ with fixed currents $I(0)\equiv I_0$, $I(t)\equiv I_1$. Denoting by $V_t(I_1|I_0;x(\tau))$ the probability density of the transition from I_0 to I_1, we obtain

$$V_t(I_1|I_0;x(\tau))W_0(I_0)\left\langle\exp\left\{-\beta\int_0^t x(\tau)I(\tau)d\tau\right\}\right\rangle^{I_1,I_0}=V_t(-\varepsilon I_0|-\varepsilon I_1;\varepsilon x(t-\tau))W_0(I_1), \quad (A.6)$$

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 $x(\tau)=0$ at $\tau<0$, then the formula

$$\left\langle\exp\left\{-\beta\int_t^\infty x(\tau)I(\tau)d\tau\right\}\right\rangle^{I_1}V_t(I_1|I_0;x(\tau))W_0(I_0)\times\left\langle\exp\left\{-\beta\int_0^t x(\tau)I(\tau)d\tau\right\}\right\rangle^{I_1,I_0}=V_t(-\varepsilon I_0|-\varepsilon I_1;\varepsilon x(t-\tau))W(-\varepsilon I_1;\varepsilon x(t-\tau)) \quad (A.7)$$

can be derived from (1) in analogy with (A.6). Here, however, in contrast to (A.6), as is seen, we have used the assumption of Markov currents, and $W(I;\varepsilon x(t-\tau))$ denotes (for the process with time reversal) the nonequilibrium current distribution $I(t=0)=I$ after the action of the forces $\varepsilon x(t-\tau)$. From (A.7) and the formula (10) (generalized to variable forces), we obtain

$$V_t(-\varepsilon I_0|-\varepsilon I_1;\varepsilon x(t-\tau))=V_t(I_1|I_0;x(\tau))\times\left\langle\exp\left\{-\beta\int_0^t x(\tau)I(\tau)d\tau\right\}\right\rangle^{I_1,I_0}.$$

Evidently, the right-hand side of this equation does not depend on $x(\tau)$ at $\tau>t$. Consequently, the left-hand side does not depend on the reversed trajectory of the forces $\varepsilon x(t-\tau)$ at $\tau<0$ and the transition probability density V_t 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 consequences of the time symmetry for 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 $t\to 0$, $x(\tau)\approx x$:

$$e^{-\beta x I_0 t}W_0(I_0)\exp\{tL(x;I_1;\partial/\partial I_1)\}\delta(I_1-I_0)=W_0(I_1)\exp\{tL(\varepsilon x;-\varepsilon I_0;-\varepsilon\,\partial/\partial I_0)\}\delta(I_1-I_0).$$

$$W_0(I_0)\{L(x;I_1;\partial/\partial I_1)-\beta xI_0\}\delta(I_1-I_0)=W_0(I_1)L(\varepsilon x;-\varepsilon I_0;-\varepsilon\,\partial/\partial I_0)\delta(I_1-I_0). \quad (A.8)$$

Multiplying (A.8) by an arbitrary function $f(I_1)$ and integrating over I_1, we obtain, at $I_0\equiv I$,

$$L(\varepsilon x;-\varepsilon I;-\varepsilon\,\partial/\partial I)W_0(I)f(I)=W_0(I)\{L^+(x;I;\partial/\partial I)-\beta xI\}f(I).$$

This equation is equivalent to the operator equation (14).

4. We shall show that formula (24′) is a consequence of the assumption [(22), (23)] of the existence in the given system of the analog of an equilibrium FDR. We introduce the generating function for the diffusion coefficients:

$$D(u;x)=\sum_{n=1}^{\infty}\frac{u^n}{n!}D_n(x).$$

In the Markov theory it can also be expressed as

$$D(u;x)=\lim_{t\to\infty}\frac{1}{t}\ln\left\{\int dI\exp[t(L+uI)]W_x(I)\right\}. \quad (A.9)$$

It is seen then that $D(u;x)$ is identical with the largest eigenvalue of the operator $L+uI$.

We now consider the formula (24), rewriting it in the form

$$\exp\{t[L_0+\beta(x)(Ix-N(x))]\}W_x(I)=W_x(I)$$

$$\beta(x)N(x)=\lim_{t\to\infty}\frac{1}{t}\ln\left\{\int dI\exp[t(L_0+\beta(x)xI)]\right\}W_x(I). \quad (A.10)$$

Comparing (A.10) with (A.9) (for the case in which the forces in (A.9) are equal to zero and the operator L_0 replaces L), in the limit $t\to\infty$, we arrive at (24′)

$$\beta(x)N(x)=D(\beta(x);0)=\sum_{n=1}^{\infty}\frac{1}{n!}D_n(0)(\beta(x)x)^n,$$

where the $D_n(0)$ are equilibrium diffusion coefficients and $D_1(0)=\langle I\rangle_0=0$.

[1]Of course, SNS of the indicated type—excited by constant external forces—do not exhaust all the diversity of SNS, and the Markov FDR derived below have a more limited circle of applications than the initial relations. We shall not touch upon cases in which the constant forces do not destroy the equilibrium but the SNS can arise under the action of periodic forces (resonantly interacting with the system). Moreover, an SNS can occur even in the absence of dynamic perturbations as a result of nonequilibrium boundary conditions (thermal perturbations). True, in this case one can sometimes replace the nonequilibrium conditions by certain effective forces and use dynamical FDR.

[2]As in Ref. 1, we use a scalar notation for vectors and tensors; only in case of necessity do we transform to the full description.

[3]The term "macrovariable" can apply also to an individual particle (or an ensemble of noninteracting particles).

[4]These relations yield a generalization of the Kubo formula to the nonlinear case.

[5]It is clear that the coefficients Γ' take into account the nondissipative contribution of the thermostat to the motion of the macrovariables, for example, the renormalization of their eigenfrequencies as a result of the interaction with the thermostat. The same can also be said about the Markov kinetic coefficients K_1' introduced below.

[6]This is certainly true for a linear system.

[7]We note that formulas (11)-(13) confirm certain results of the phenomenological approach,[6] in which the condition of the maximum of informational entropy for a given value of dissipation (or, in our terminology, at given mean values of the currents) is used to construct the distribution functions in the SNS.

[8]As before, we use the real variable u instead of iu, since we are interested only in the formal structure of the relations.

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Two-dimensional electronic phenomena in germanium bicrystals at helium temperatures

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The two-dimensional conductivity, Hall effect, and Shubnikov–de Haas oscillations on the intergrowth surfaces of germanium bicrystals with inclination angles $6^\circ \leq \theta \leq 30^\circ$ were investigated at helium temperatures in magnetic fields up to 150 kOe. A transition from metallic to thermally activated conductivity was observed at an angle $\theta \approx (8–9)^\circ$. The minimal metallic conductivity is found to be $\sigma_{\min} \approx e^2/2\pi\hbar$, in agreement with the simplest theoretical estimates. Shubnikov–de Haas oscillations are observed in the metallic conductivity region at $\theta \gtrsim 20^\circ$. They are shown to be due to the contribution of the light holes. It is established that the conductivity decreases sharply with decreasing angle θ, owing to onset of one-dimensional conduction conditions. The anisotropy of the conductivity is investigated. A model is proposed to explain the observed phenomena.

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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.[1] A more reliable model of a two-dimensional system, in our opinion, consists of highly conducting layers adjacent to the cleavage planes of germanium crystals. 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.[2,3]

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 $\theta/2$ to the [100] axis,

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