SOVIET PHYSICS JETP VOLUME 28, NUMBER 6 JUNE, 1969

A FLUCTUATION DISSIPATION THEOREM FOR NONLINEAR MEDIA

G. F. EFREMOV

Gor'kiĭ State University

Submitted June 26, 1968

Zh. Eksp. Teor. Fiz. 55, 2322–2333 (December, 1968)

A theorem is derived for quantities of arbitrary physical nature, expressing the third moments of equilibrium fluctuations and the spectral fluctuations of first order with respect to the external force, in terms of the second order nonlinear susceptibilities.

INTRODUCTION

At the present time fluctuation theory has investigated most extensively the spectral properties of equilibrium fluctuations. A decisive step in the theory of equilibrium fluctuations was the derivation by Callen and Welton[1,2] of a fluctuation-dissipation theorem, relating the spectral density of equilibrium fluctuations to the dynamical behavior of the system in an external field (the linear susceptibility)[3-7]. Under special assumptions about the Markovian or Gaussian character of the processes involved, the complete theory of equilibrium fluctuations reduces to spectral theory[8,9].

It is of general physical interest to establish for equilibrium fluctuations relations among the amounts of higher order involving nonlinear susceptibilities and the quantities which determine the nonequilibrium fluctuations.

In many phenomena a fundamental role is played by additional fluctuations which appear in the system under the action of the external excitation (Mandel'shtam-Brillouin and Raman scattering, parametric fluctuations). It is therefore interesting to find relations between the qualities which determine the additional fluctuations and the quantities which determine the dynamical behavior (i.e., the nonlinear susceptibilities).

In the first part of this paper the third moments of the equilibrium fluctuations and the additional fluctuations of first order with respect to the external force are expressed in terms of the quantities determining the dynamical behavior of the system (nonlinear susceptibilities of the second order in the force).

In the second part the general equations are applied to electromagnetic fluctuations. In particular, the parametric fluctuations are considered which were experimentally discovered by Harris, Oshman and Byer[10] and Akhmanov, Fadeev, Khokhlov and Chunaev[11].

In the third part general relations are found between the nonlinear susceptibilities of third order in the external force and the additional fluctuations of second order.

All possible particular cases are considered for the general formulas, as well as symmetry relations. In the case when there is a resonance only for the difference frequency, the general relations go over into the formula obtained by Faĭn and Yashchin[12].

The fourth moments of equilibrium fluctuations are expressed in terms of quantities which determine the additional fluctuations of second order in the external force.

1. BASIC DEFINITIONS

We consider an arbitrary system described by quantum mechanics, subjected to an external perturbation of the form

$$V(t) = -X_a f_a(t), \tag{1.1}$$

where X_a is a physical quantity describing the system and $f_a(t)$ is a given external field.

The expectation value of X_a at the instant t under the action of the perturbation (1.1) will be denoted by $\langle X_a(t)\rangle$. The brackets $\langle\ldots\rangle$ denote averaging over an equilibrium state, described by the density matrix of a canonical ensemble[1)]:

$$\rho_0 = \exp\left\{-\frac{\beta}{\hbar}(\mathcal{H}_0 - F)\right\} \tag{1.2}$$

where $\mathcal{H}_0$ is the Hamiltonian of the unperturbed system; $\beta = \hbar/kT$; k is the Boltzmann constant and $\hbar$ is Planck's constant. $X_a^H(t)$ is an operator in the Heisenberg picture

$$X_a^H(t) = S^{-1}(t)X_a(t)S(t), \tag{1.3}$$

where S(t) is the time evolution operator, related to the external perturbation (1.1) by means of the formula[2)]

$$S(t) = T\exp\left\{\frac{i}{\hbar}\int_{-\infty}^{t} dt'\, X_{a'}(t')f_{a'}(t')\right\}, \tag{1.4}$$

$X_a(t)$ is the operator X_a in the interaction picture.

Assume $\langle X_a^H(t)\rangle$ to be expanded in powers of the external field $f_a(t)$. Then in first order in the field we obtain the linear response or reaction function $\varphi_{ab}(t, t_1)$, in second order we obtain the nonlinear response of second order $\varphi_{abc}(t, t_1, t_2)$, etc.

$$\langle X_a^H(t)\rangle = \int_{-\infty}^{\infty} dt_1\, \varphi_{ab}(t, t_1)f_b(t_1) + \int_{-\infty}^{\infty} dt_1 \int_{-\infty}^{\infty} dt_2\, \varphi_{abc}(t, t_1, t_2)f_b(t_1)f_c(t_2) + \ldots \tag{1.5}$$

---

1)We assume that the external field $f_a(t)$ is switched on adiabatically. For $t = -\infty$ the quantity $f_a(-\infty) = 0$ and the system is in the equilibrium state (1.2).

2)T is the time-ordering operator which arranges the operators belonging to earlier times to the right of operators defined at a later time (cf., e. g., [13]).

similarly we obtain a more detailed description of the system if we define the function

$$\Psi_{ab}{}^{H}(t, t_1) = {}^1/_2\langle [X_a{}^{H}(t),\ X_b{}^{H}(t_1)]_+\rangle. \quad (1.6)$$

The function (1.6) determines the correlation properties of nonlinear fluctuations.

We assume that (1.6) may be expanded in powers of the external field:

$$\Psi_{ab}{}^{H}(t, t_1) = \Psi_{ab}(t, t_1) + \int_{-\infty}^{\infty} dt_2\, \Psi_{abc}(t, t_1, t_2) f_c(t_2) + \int_{-\infty}^{\infty} dt_2 \int_{-\infty}^{\infty} dt_3\, \Psi_{abcd}(t, t_1, t_2, t_3) f_c(t_2) f_d(t_3) + \ldots \quad (1.7)$$

The first term in (1.7) determines the correlation properties of equilibrium fluctuations, $\Psi_{abc}(t, t_1, t_2)$ determines the nonequilibrium fluctuations to first order in the field, etc. By analogy with (1.6) one may define the moments of higher-order fluctuations. In the present paper however we restrict ourselves to the consideration of correlation properties of the nonequilibrium fluctuations.

We now go over from a temporal description to a spectral one. We define the Fourier transforms of the reaction functions $\varphi_{ab}(t, t_1)$, $\varphi_{abc}(t, t_1, t_2), \ldots$ and of the functions $\Psi_{ab}(t, t_1)$, $\Psi_{abc}(t, t_1, t_2), \ldots$ by means of the relations[3)]:

$$\chi_{ab}(\omega) = \int_{-\infty}^{\infty} d\tau\, e^{i\omega\tau} \varphi_{ab}(\tau), \quad (1.8)$$

$$\chi_{abc}(\omega_1, \omega_2) = \int_{-\infty}^{\infty} d\tau_1 \int_{-\infty}^{\infty} d\tau_2\, e^{i\omega_1\tau_1 + i\omega_2\tau_2} \varphi_{abc}(\tau_1, \tau_2), \quad (1.8')$$

. . . .

$$\Phi_{ab}(\omega) = \int_{-\infty}^{\infty} d\tau\, e^{i\omega\tau}\, \Psi_{ab}(\tau), \quad (1.9)$$

$$\Phi_{abc}(\omega_1; \omega_2) = \int d\tau_1 \int d\tau_2\, e^{i\omega_1\tau_1 + i\omega_2\tau_2}\, \Psi_{abc}(\tau_1, \tau_2), \quad (1.9')$$

. . . .

Here $\chi_{ab}(\omega)$ is the linear susceptibility, $\chi_{abc}(\omega_1, \omega_2)$ is the nonlinear susceptibility, or cross-susceptibility of second order, etc., $\Phi_{ab}(\omega)$ determines the spectral intensity (density) of the equilibrium fluctuations, $\Phi_{abc}(\omega_1, \omega_2)$ describes the spectral properties of the second order nonequilibrium fluctuations, etc.

The inverse Fourier transform, e.g., of (1.9′), has the form:

$$\Psi_{abc}(\tau_1, \tau_2) = \int_{-\infty}^{\infty} \frac{d\omega_1}{2\pi} \int_{-\infty}^{\infty} \frac{d\omega_2}{2\pi} e^{-i\omega_1\tau_1 - i\omega_2\tau_2}\, \Phi_{abc}(\omega_1; \omega_2). \quad (1.10)$$

2. THE CAUSAL NONEQUILIBRIUM GREEN'S FUNCTION

Further we must write the reaction and fluctuation functions in such a form that use can be made of the information following from the thermodynamic equilibrium condition, and from the symmetry of the equations of motion with respect to time reversal.

[3)]Owing to the temporal homogeneity of an equilibrium system the reaction and fluctuation functions depend only on time differences $\tau_i = t - t_i (i = 1, 2, \ldots)$. We shall call the functions $\Psi_{abc}(t, t_1, t_2')$ etc. fluctuation functions.

We show that the dynamic behavior of the system and the fluctuations defined by (1.6) are described by the causal Green's function:

$$G_{ab}{}^{H}(t, t_1) = -\frac{i}{\hbar} \langle T[X_a{}^{H}(t),\ X_b{}^{H}(t_1)]\rangle$$
$$= -\frac{i}{\hbar} \langle X_a{}^{H}(t) X_b{}^{H}(t_1)\rangle\, \eta(t - t_1) + \frac{i}{\hbar} \langle X_b{}^{H}(t_1) X_a{}^{H}(t)\rangle\, \eta(t_1 - t)$$
$$= -\frac{i}{2\hbar} \langle [X_a{}^{H}(t), X_b{}^{H}(t_1)]_-\rangle\, \mathrm{sign}(t - t_1) + \frac{i}{2\hbar} \langle [X_a{}^{H}(t), X_b{}^{H}(t_1)]_+\rangle, \quad (2.1)$$

where

$$\eta(t - t_1) = \begin{cases} 1, & t - t_1 > 0 \\ 0, & t - t_1 < 0 \end{cases} \quad (2.2)$$

is the Heaviside step function;

$$\mathrm{sign}(\tau) = \eta(\tau) - \eta(-\tau), \quad (2.2')$$

and

$$\eta(\tau) + \eta(-\tau) = 1, \qquad \mathrm{sign}(\tau) \cdot \mathrm{sign}(\tau) = 1. \quad (2.3)$$

We consider the change of $\langle X_a^H(t)\rangle$ under an infinitesimal change of the external field $f_a(t)$, cf.[14]. We set

$$f_a'(t) = f_a(t) + \delta f_a(t). \quad (2.4)$$

The variation of (1.5) for a small increment (2.4) has the form

$$\delta\langle X_a{}^{H}(t)\rangle = \int_{-\infty}^{\infty} dt_1\, \varphi_{ab}(t, t_1)\, \delta f_b(t_1) + 2 \int dt_1 \int dt_2\, \varphi_{abc}(t, t_1, t_2) f_c(t_2)\, \delta f_b(t_1) + \ldots \quad (2.5)$$

By definition the coefficient of $\delta f_b(t_1)$ is a functional derivative. Thus

$$\varphi_{ab}(t, t_1) = \frac{\delta\langle X_a{}^{H}(t)\rangle}{\delta f_b(t_1)}\bigg|_{f=0},$$
$$\varphi_{abc}(t, t_1, t_2) = \frac{1}{2!} \frac{\delta^2\langle X_a{}^{H}(t)\rangle}{\delta f_b(t_1)\, \delta f_c(t_2)}\bigg|_{f=0} \quad (2.6)$$

or

$$\varphi_{ab}(t, t_1) = \varphi_{ab}{}^{\Gamma}(t, t_1)\,|_{f=0},$$
$$\varphi_{abc}(t, t_1, t_2) = \frac{1}{2!} \frac{\delta\varphi_{ab}{}^{H}(t, t_1)}{\delta f_c(t_2)}\bigg|_{f=0},$$

where

$$\varphi_{ab}{}^{H}(t,\ t_1) = \delta\langle X_a{}^{H}(t)\rangle / \delta f_b(t_1). \quad (2.7)$$

Let us find the explicit form of the function (2.7). For this purpose we compute the variation δS of the S matrix (1.4) for an infinitesimal variation of $f(t)$, (2.4). We have

$$\delta S(t) = T\left\{\delta\left[\exp\frac{i}{\hbar}\int_{-\infty}^{t} dt'\, X_{a'}(t') f_{a'}(t')\right]\right\}$$
$$= \frac{i}{\hbar}\int_{-\infty}^{t} dt'\, \delta f_{a'}(t') S(t, t') X_{a'}(t') S(t')$$
$$= \frac{i}{\hbar}\int_{-\infty}^{t} dt'\, \delta f_{a'}(t')\, T[S(t) X_{a'}(t')]. \quad (2.8)$$

This leads to (cf. Eq. (2.18))

$$\frac{\delta S(t)}{\delta f_b(t_1)} = \frac{i}{\hbar} T[S(t) X_b(t_1)]\, \eta(t - t_1) = \frac{i}{\hbar} S(t) X_b{}^{H}(t_1)\, \eta(t - t_1). \quad (2.9)$$

The functional derivative of the inverse S matrix can be determined from the unitarity condition

$$S^{-1}(t)S(t) = 1; \tag{2.10}$$

we obtain

$$\frac{\delta(S^{-1}(t))}{\delta f_b(t_1)} = -S^{-1}(t)\frac{\delta S(t)}{\delta f_b(t_1)}S^{-1}(t) = -\frac{i}{\hbar}X_b^{\mathrm{H}}(t_1)S^{-1}(t)\eta(t-t_1). \tag{2.11}$$

Using (2.9) and (2.11) we obtain the following expression for (2.7):

$$\varphi_{ab}^{\mathrm{H}}(t,t_1) = \frac{\delta\langle S^{-1}(t)X_a(t)S(t)\rangle}{\delta f_b(t_1)} = \frac{i}{\hbar}\langle[X_a^{\mathrm{H}}(t),X_b^{\mathrm{H}}(t_1)]_-\rangle\eta(t-t_1). \tag{2.12}$$

From (2.12), (2.3) and the commutation properties it follows that the function $\varphi_{ab}^{\mathrm{H}}(t, t_1)$ has the obvious properties:

$$\varphi_{ab}^{\mathrm{H}}(t,t_1) - \varphi_{ba}^{\mathrm{H}}(t_1,t) = \frac{i}{\hbar}\langle[X_a^{\mathrm{H}}(t),X_b^{\mathrm{H}}(t_1)]_-\rangle, \tag{2.13}$$

$$\varphi_{ab}^{\mathrm{H}}(t,t_1) + \varphi_{ba}^{\mathrm{H}}(t_1,t) = \frac{i}{\hbar}\langle[X_a^{\mathrm{H}}(t),X_b^{\mathrm{H}}(t_1)]_-\rangle\mathrm{sign}(t-t_1), \tag{2.14}$$

$$\varphi_{ab}^{\mathrm{H}}(t,t_1) - \varphi_{ba}^{\mathrm{H}}(t_1,t) = [\varphi_{ab}^{\mathrm{H}}(t,t_1) + \varphi_{ba}^{\mathrm{H}}(t_1,t)]\,\mathrm{sign}(t-t_1). \tag{2.15}$$

Comparing now (2.1) with (1.6) and (2.14) we obtain

$$G_{ab}^{\mathrm{H}}(t,t_1) = \frac{1}{2}[\varphi_{ab}^{\mathrm{H}}(t,t_1) + \varphi_{ba}^{\mathrm{H}}(t_1,t)] + \frac{i}{\hbar}\Psi_{ab}^{\mathrm{H}}(t,t_1). \tag{2.16}$$

Thus the real part of the Green's function (2.1) determines the dynamical behavior of the system and the imaginary part determines the nonequilibrium fluctuations (1.6).

We transform (2.1) to the interaction picture according to (1.3) making use of the properties of the operator S (cf.[13])

$$S(\infty,t)S(t,-\infty) = S(\infty,-\infty), \tag{2.17}$$

or

$$S^{-1}(t) = S^{-1}(\infty)S(\infty,t), \tag{2.17'}$$

$$S(t,t_1)S(t_1,-\infty) = S(t,-\infty), \quad t > t_1,$$

$$S(t,t_1) = S(t)S^{-1}(t_1). \tag{2.18}$$

For $t > t_1$ we obtain

$$G_{ab}^{\mathrm{H}}(t,t_1) = \frac{i}{\hbar}\langle S^{-1}(t)X_a(t)S(t)S^{-1}(t_1)X_b(t_1)S(t_1)\rangle = \frac{i}{\hbar}\langle S^{-1}(\infty)T[S(\infty)X_a(t)X_b(t_1)]\rangle. \tag{2.19}$$

It is obvious that the result obtained in this manner does not depend on the ordering of the times t and t_1. Using equations (2.9) and (2.11) for $t = \infty$ it is easy to find the expansion coefficients (or functional derivatives) of the Green's function (2.19) with respect to the powers of the external field. Thus

$$\left.\frac{\delta G_{ab}^{\mathrm{H}}(t,t_1)}{\delta f_c(t_2)}\right|_{f=0} = \left(\frac{i}{\hbar}\right)^2\Big\{\langle T[X_a(t)X_b(t_1)X_c(t_2)]\rangle - \langle X_c(t_2)T[X_a(t)X_b(t_1)]\rangle\Big\}, \tag{2.20}$$

$$\left.\frac{\delta^2 G_{ab}^{\mathrm{H}}(t,t_1)}{\delta f_c(t_2)\delta f_d(t_3)}\right|_{f=0} = \left(\frac{i}{\hbar}\right)^3\Big\{\langle T[X_a(t)X_b(t_1)X_c(t_2)X_d(t_3)]\rangle - \langle X_c(t_2)T[X_a(t)X_b(t_1)X_d(t_3)]\rangle - \langle X_d(t_3)T[X_a(t)X_b(t_1)X_c(t_2)]\rangle - \langle T[X_c(t_2)X_d(t_3)]T[X_a(t)X_b(t_1)]\rangle + \langle[X_c(t_2),X_d(t_3)]_+T[X_a(t)X_b(t_1)]\rangle\Big\}. \tag{2.21}$$

For the purpose of abbreviation we introduce the notations

$$\frac{1}{i^2}\langle T[X_a(t)X_b(t_1)X_c(t_2)]\rangle = G(t,t_1,t_2),$$

$$\langle X_c(t_2)T[X_a(t)X_b(t_1)]\rangle = B_2(t_2;t,t_1),$$

$$-\frac{1}{i}\langle T[X_a(t)X_b(t_1)X_c(t_2)X_d(t_3)]\rangle = G(t,t_1,t_2,t_3), \tag{2.22}$$

$$i\langle X_c(t_2)T[X_a(t)X_b(t_1)X_d(t_3)]\rangle = A_2(t_2;t,t_1,t_3),$$

$$i\langle T[X_c(t_2)X_d(t_3)]T[X_a(t)X_b(t_1)]\rangle - i\langle[X_c(t_2),X_d(t_3)]_+T[X_a(t)X_b(t_1)]\rangle = \alpha_{23}(t_2,t_3;t,t_1).$$

Finally, equating the functional derivatives of the left- and right-hand sides of (2.16) and utilizing the notations (2.22) we obtain the equations

$$\varphi_{abc}(t,t_1,t_2) + \varphi_{bac}(t_1,t,t_2) + \frac{i}{\hbar}\Psi_{abc}(t,t_1,t_2) = \frac{1}{\hbar^2}\{G(t,t_1,t_2) + B_2(t_2;t,t_1)\}, \tag{2.23}$$

$$3[\varphi_{abcd}(t,t_1,t_2,t_3) + \varphi_{bacd}(t_1,t,t_2,t_3)] + 2\frac{i}{\hbar}\Psi_{abcd}(t,t_1;t_2,t_3) = \frac{1}{\hbar^3}\{G(t,t_1,t_2,t_3) + A_2(t_2,t,t_1,t_3) + A_3(t_3,t,t_1,t_2) + \alpha_{23}(t_2,t_3;t,t_1)\}. \tag{2.24}$$

For the functions occurring in the right-hand sides of the equations (2.23), (2.24) it is easy to write down relations following from the conditions of thermodynamic equilibrium and the symmetry with respect to time reversal. Time-reversal invariance of the equations and the hermitean character of the operators X_a imply the following properties of the functions (2.22) (cf.[4,15]),

$$G(t,t_1) = \pm G(-t,-t_1), \qquad G(t,t_1,t_2) = \pm G(-t,-t_1,-t_2), \tag{2.25}$$

$$B_2(-t_2,-t,-t_1) = \pm\langle T[X_a(t),X_b(t_1)]X_c(t_2)\rangle, \tag{2.26}$$

$$A_2(-t_2,-t,-t_1,-t_3) = \pm i\langle T[X_a(t),X_b(t_1),X_d(t_3)]X_c(t_2)\rangle, \tag{2.27}$$

$$\alpha_{23}(-t_2,-t_3,-t,-t_1) = \pm i\{\langle T[X_a(t),X_b(t_1)]T[X_c(t_2),X_d(t_3)]\rangle - \langle T[X_a(t),X_b(t_1)][X_c(t_2),X_d(t_3)]_+\rangle\}. \tag{2.28}$$

The sign + corresponds to the case when the product $\epsilon^a\epsilon^b\ldots$ equals +1. The value of ϵ^a is +1 if X_a does not change sign under time reversal, and is −1 if X_a does change sign under time reversal.

It should be noted that from the symmetry of the Green's function $G(\tau)$, (2.25) and of the equations (2.16), (2.15) for $f = 0$ follow the Onsager reciprocity relations

$$\varphi_{ab}(\tau) = \varepsilon^a\varepsilon^b\varphi_{ba}(\tau), \qquad \Psi_{ab}(\tau) = \varepsilon^a\varepsilon^b\Psi_{ba}(\tau). \tag{2.29}$$

For definiteness we select in Eqs. (2.25)—(2.27) the sign +. Then the causal Green's functions (2.25) will be symmetric with respect to time-reversal invariance. The symmetric and antisymmetric parts of function $B_2(t_2; t_1, t_1)$ have, respectively, the form

$$B_2^s(t_2;t,t_1) = {}^1\!/_2\langle[X_c(t_2),T[X_a(t),X_b(t_1)]]_+\rangle,$$

$$B_2^a(t_2;t,t_1) = {}^1\!/_2\langle[X_c(t_2),T[X_a(t),X_b(t_1)]]_-\rangle. \tag{2.30}$$

In a thermodynamic equilibrium state the commutator and anticommutator are related by the equation[4)]:

4)The equation (2.31) can be easily obtained from the formal resemblance of the evolution operator of the system, $\exp(-i\mathcal{H}_0 t/\hbar)$ to the density matrix $\exp(-\beta\mathcal{H}_0/\hbar)$ (cf. /[16, 19]/).

$$\langle [X_a, X_b(\omega)]_+\rangle = \operatorname{cth}\left(\frac{\beta\omega}{2}\right)\langle [X_a, X_b(\omega)]_-\rangle, \tag{2.31}$$

where $X_b(\omega)$ is the Fourier transform of $X_b(-\tau)$, defined by

$$X_b(\omega) = \int_{-\infty}^{\infty} d\tau\, e^{i\omega\tau} X_b(-\tau). \tag{2.32}$$

In analogy to Eq. (2.31) we have

$$B_2{}^s(\omega_2;\omega_1) = -\operatorname{cth}\left(\frac{\beta\omega_2}{2}\right) B_2{}^a(\omega_2;\omega_1), \tag{2.33}$$

$$A_2{}^s(\omega_2;\omega_1,\omega_3) = -\operatorname{cth}\left(\frac{\beta\omega_2}{2}\right) A_2{}^a(\omega_2;\omega_1,\omega_3), \tag{2.34}$$

$$\alpha_{23}{}^s(\omega_2,\omega_3;\omega_1) = -\operatorname{cth}\left(\frac{\beta(\omega_2+\omega_3)}{2}\right)\alpha_{23}{}^a(\omega_2,\omega_3;\omega_1), \tag{2.35}$$

where $A_2^S(\omega_2;\omega_1,\omega_3)$, $\alpha_{23}^S(\omega_2,\omega_3,\omega_1)$ are the Fourier transforms of the symmetric parts of the corresponding functions. The proof of equations (2.33)–(2.35) runs along the same lines as that of (2.31).

Writing down Eqs. (1.6) and (2.13) for f = 0 in the spectral form

$$\Phi_{ab}(\omega) = {}^1/_2\langle [X_a, X_b(\omega)]_+\rangle,$$
$$\chi_{ab}(\omega) - \chi_{ba}{}^*(\omega) = \frac{i}{\hbar}\langle [X_a, X_b(\omega)]_-\rangle \tag{2.36}$$

and utilizing the relation (2.31) we obtain the fundamental theorem of Callen-Welton

$$\Phi_{ab}(\omega) = \operatorname{cth}\left(\frac{\beta\omega}{2}\right)\frac{\hbar}{2i}[\chi_{ab}(\omega) - \chi_{ba}{}^*(\omega)]. \tag{2.37}$$

Utilizing the Onsager reciprocity for $\epsilon^a\epsilon^b = 1$, we obtain

$$\Phi_{ab}(\omega) = \operatorname{cth}(\beta\omega/2)\hbar\chi''_{ab}(\omega). \tag{2.37'}$$

The Callen-Welton theorem relates the spectral intensity of equilibrium fluctuations and the linear susceptibility which determines the dynamic behavior of the system in a weak external field.

3. A FLUCTUATION-DISSIPATION THEOREM FOR MEDIA WITH QUADRATIC NONLINEARITIES

We rewrite the symmetric and antisymmetric parts of the equation (2.23) in the spectral form[5]:

$$\chi'_{abc}(\omega_1,\omega_2) + \chi'_{bac}(\omega_0,\omega_2) + \frac{i}{\hbar}\Phi'_{abc}(\omega_1;\omega_2)$$
$$= \frac{1}{\hbar^2}[G(\omega_1,\omega_2) + B_2{}^s(\omega_2;\omega_1)], \tag{3.1}$$

$$i\chi''_{abc}(\omega_1,\omega_2) + i\chi''_{bac}(\omega_0,\omega_2)$$
$$-\frac{1}{\hbar}\Phi''_{abc}(\omega_1;\omega_2) = \frac{1}{\hbar^2}B_2{}^a(\omega_2;\omega_1). \tag{3.2}$$

Separating here real and imaginary parts we have

$$\chi'_{abc}(\omega_1,\omega_2) + \chi'_{bac}(\omega_0,\omega_2) = \frac{1}{\hbar^2}G'(\omega_1;\omega_2)$$
$$+\frac{1}{\hbar^2}B_2{}'^s(\omega_2;\omega_1), \tag{3.3}$$

$$-\Phi''_{abc}(\omega_1;\omega_2) = \frac{1}{\hbar}B_2{}'^a(\omega_2;\omega_1), \tag{3.4}$$

$$\chi''_{abc}(\omega_1,\omega_2) + \chi''_{bac}(\omega_0,\omega_2) = \frac{1}{\hbar^2}B_2''^a(\omega_2;\omega_1), \tag{3.5}$$

$$\Phi'_{abc}(\omega_1;\omega_2) = \frac{1}{\hbar}G''(\omega_1,\omega_2) + \frac{1}{\hbar}B_2''^s(\omega_2;\omega_1). \tag{3.6}$$

The remaining equations are obtained from (3.3)–(3.6) by cyclic permutation of the indices a, b, c and simultaneously of the frequencies ω_0, ω_1, ω_2.

Due to the homogeneity in time of the system we have

$$\omega_0 + \omega_1 + \omega_2 = 0. \tag{3.7}$$

The spectral decomposition of the functions B and G (cf. Appendix imply the relations

$$B_0{}'^a(\omega_0;\omega_2) + B_1{}'^a(\omega_1;\omega_0) + B_2{}'^a(\omega_2;\omega_1) = 0, \tag{3.8}$$

$$B_0''^s(\omega_0;\omega_2) + B_1''^s(\omega_1;\omega_0) + B_2''^s(\omega_2;\omega_1) + G''(\omega_1,\omega_2) = 0. \tag{3.9}$$

Utilizing (2.33) we can rewrite the relation (3.8) in the form

$$\operatorname{th}\left(\frac{\beta\omega_0}{2}\right)B_0{}'^s(\omega_0;\omega_2) + \operatorname{th}\left(\frac{\beta\omega_1}{2}\right)B_1{}'^s(\omega_1;\omega_0) + \operatorname{th}\left(\frac{\beta\omega_2}{2}\right)B_2{}'^s(\omega_2;\omega_1) = 0, \tag{3.10}$$

where

$$B_0{}'^s(\omega_0;\omega_2) = {}^1/_4\langle [X_a(\omega_0), [X_b, X_c(\omega_2)]_+]_+\rangle,$$
$$B_1{}'^s(\omega_1;\omega_0) = {}^1/_4\langle [X_b(\omega_1), [X_c, X_a(\omega_0)]_+]_+\rangle \tag{3.11}$$

are the symmetrized third moments which determine the equilibrium fluctuations. More details on this subject can be found in the paper by Bernard and Callen[17].

Let us find a solution of the system (3.3)–(3.6). Eliminating between (3.3) and its cyclic permutations the function $G'(\omega_1,\omega_2)$ and substituting $B_2'^S(\omega_2;\omega_1)$, $B_1'^S(\omega_1;\omega_0)$ into the relation (3.10), we obtain

$$\frac{1}{4}\langle [X_a, [X_b(\omega_1), X_c(\omega_2)]_+]_+\rangle$$
$$= \hbar^2\operatorname{cth}\left(\frac{\beta\omega_0}{2}\right)P_{12}[\chi'_{abc}(\omega_1,\omega_2) - \chi'_{bac}(\omega_0,\omega_2)]\operatorname{cth}\left(\frac{\beta\omega_2}{2}\right) \tag{3.12}$$

where P_{12} is the permutation operator of $b\omega_1$, $c\omega_2$. The expectation value of the cube of X_a is given by the integral

$$\langle X_a{}^3\rangle = 2\hbar^2\int_{-\infty}^{\infty}\frac{d\omega_1}{2\pi}\int_{-\infty}^{\infty}\frac{d\omega_2}{2\pi}[\chi'_{aaa}(\omega_0,\omega_2)$$
$$-\chi'_{aaa}(\omega_1,\omega_2)]\operatorname{cth}\left(\frac{\beta\omega_2}{2}\right)\operatorname{cth}\left(\frac{\beta(\omega_1+\omega_2)}{2}\right). \tag{3.13}$$

Thus, the third moments of the equilibrium fluctuations are determined by the nonlinear susceptibilities.

We establish relations between the nonlinear susceptibilities and the functions of fluctuations. Substituting (3.12) into the equation

$$\Phi''_{bca}(\omega_2;\omega_0) = -\frac{1}{\hbar}B_0{}'^a(\omega_1;\omega_2) = \frac{1}{\hbar}\operatorname{th}\left(\frac{\beta\omega_0}{2}\right)B_0{}'^s(\omega_1;\omega_2), \tag{3.14}$$

we have

$$\Phi''_{bca}(\omega_2;\omega_0) = \hbar P_{12}[\chi'_{abc}(\omega_1,\omega_2) - \chi'_{bca}(\omega_2,\omega_0)]\operatorname{cth}\left(\frac{\beta\omega_2}{2}\right). \tag{3.15}$$

The equations (3.5), (3.6), (3.9) also imply

$$\Phi'_{bca}(\omega_2;\omega_0) = \hbar P_{12}[\chi''_{abc}(\omega_1,\omega_2) + \chi''_{bca}(\omega_2,\omega_0)]\operatorname{cth}(\beta\omega_2/2). \tag{3.16}$$

Together the formulas (3.15) and (3.16) become

$$\Phi_{bca}(\omega_2;\omega_0) = -i\hbar P_{12}[\chi_{bca}(\omega_2,\omega_0) - \chi^*_{abc}(\omega_1,\omega_2)]\operatorname{cth}(\beta\omega_2/2). \tag{3.17}$$

Two other relations are obtained from (3.17) by means of cyclic permutations of $\omega_0 a$, $\omega_1 b$, $\omega_2 c$. It is easy to invert the relations (3.17) and to express the

[5]Since the functions $\varphi_{abc}(\tau_1,\tau_2)$ and $\Psi_{abc}(\tau_1,\tau_2)$ are real, the Fourier transforms of the symmetric parts are real and those of the antisymmetric parts are purely imaginary.

nonlinear susceptibilities in terms of the fluctuation functions.

If the quantities X_a change sign under time reversal, (3.17) is replaced by

$$\Phi_{bca}(\omega_2;\omega_0) = -i\hbar P_{12}[\chi_{bca}(\omega_2,\omega_0) + \overset{*}{\chi}_{abc}(\omega_1,\omega_2)]\,\mathrm{cth}(\beta\omega_2/2). \quad (3.18)$$

In the special case $\omega_0 = 0$ $(\omega_1 = -\omega_2 = \omega)$ (3.17) becomes

$$\Phi_{bca}(\omega;0) = \hbar\,\mathrm{cth}(\beta\omega/2)\,2\chi''_{bca}(\omega;0). \quad (3.19)$$

Defining the susceptibilities and fluctuations in the presence of a constant external force $f_a(0)$, we find that (3.19) coincides with the Callen-Welton formula for a system in a constant external field[6)] (cf.[18]):

$$\Phi_{bc}(\omega;f^0) = \hbar\,\mathrm{cth}\,(\beta\omega/2)\chi''_{bc}(\omega;f^0). \quad (3.19')$$

In the limiting case $kT \gg \hbar\omega$ we have $\coth(\beta\omega/2) \approx 2kT/\hbar\omega$ and the equation (3.12) takes the form

$$\langle X_aX_b(\omega_1)X_c(\omega_2)\rangle = (2kT)^2P_{12}\frac{\chi'_{bac}(\omega_0,\omega_2) - \chi'_{abc}(\omega_1,\omega_2)}{\omega_2(\omega_1+\omega_2)}. \quad (3.20)$$

In agreement with the classical nature of the fluctuations Planck's constant cancels out in (3.20) and the correlation function does not depend on the ordering of X_a, X_b, X_c.

When none of the frequencies ω_0, ω_1, ω_2 vanishes, Eq. (3.20) can be rewritten in the form

$$\langle X_aX_b(\omega_1), X_c(\omega_2)\rangle = -(2kT)^2\Big[\frac{\chi'_{abc}(\omega_1,\omega_2)}{\omega_1\omega_2} + \frac{\chi'_{bca}(\omega_2,\omega_0)}{\omega_2\omega_0} + \frac{\chi_{cab}(\omega_0,\omega_1)}{\omega_0\omega_1}\Big]. \quad (3.21)$$

If the condition $kT \gg \hbar\omega$ is valid for all relevant frequencies (i.e., when $\chi'_{abc}(\omega_1,\omega_2)$ is significantly different from zero) one may go to the classical limit in Eq. (3.13):

$$\langle X_a^3\rangle = -2(kT)^2\frac{1}{\pi^2}\int_{-\infty}^{\infty}\frac{P}{\omega_1}d\omega_1\int_{-\infty}^{\infty}\frac{P}{\omega_2}d\omega_2\,\chi'_{aaa}(\omega_1,\omega_2)$$
$$-(kT)^2\frac{1}{\pi^2}\int_{-\infty}^{\infty}d\omega_1\int_{-\infty}^{\infty}d\omega_2\frac{P}{\omega_0}\Big(\frac{P}{\omega_1}+\frac{P}{\omega_2}\Big)\chi'_{aaa}(\omega_1,\omega_2). \quad (3.22)$$

Taking into account the operator identity[7)]

$$-\frac{P}{\omega_0}\Big(\frac{P}{\omega_1}+\frac{P}{\omega_2}\Big) = \frac{P}{\omega_1}\frac{P}{\omega_2} + \pi^2\delta(\omega_1)\delta(\omega_2) \quad (3.23)$$

and utilizing the dispersion relation[12]

$$-\frac{1}{\pi^2}\int\frac{P}{\omega_1}d\omega_1\int\frac{P}{\omega_2}d\omega_2\chi'_{aaa}(\omega_1,\omega_2) = \overset{st}{\chi}_{aaa}(0,0), \quad (3.24)$$

we find

$$\langle X_a^3\rangle = 2(kT)^2\overset{st}{\chi}_{aaa}(0,0). \quad (3.25)$$

This result should follow from classical considerations.

Let X_a be a classical quantity (cf.[19]). The density matrix of the canonical ensemble in the presence of a time-independent perturbation $V = -X_af_a$ has the form

$$\rho = \exp\Big\{-\frac{1}{kT}(\mathscr{H}_0 - f_aX_a - F)\Big\}. \quad (3.26)$$

From the definition of the static susceptibilities

$$\langle X_a\rangle = \mathrm{Sp}(\rho X_a) = \overset{st}{\chi}_{aa}(0)f_a + \overset{st}{\chi}_{aaa}(0,0)f_af_a + \cdots \quad (3.27)$$

we obtain a result in agreement with equation (3.25):

$$2\overset{st}{\chi}_{aaa}(0,0) = \frac{d^2[\mathrm{Sp}(\rho X_a)]}{df_a^2}\Big|_{f_a=0} = \frac{\langle X_a^3\rangle_0}{(kT)^2},$$

or

$$\langle X_a^3\rangle_0 = 2(kT)^2\overset{st}{\chi}_{aaa}(0,0).$$

The formulas obtained above can be reformulated in the form of the following theorem.

The moments of the equilibrium fluctuations, $\frac{1}{4}\langle[X_a,[X_b(\omega_1),X_c(\omega_2)]_+]_+\rangle$ and the fluctuation functions $\Phi_{bca}(\omega_2;\omega_0)$ are determined by the nonlinear susceptibilities $\chi_{abc}(\omega_1,\omega_2)$, i.e., the dynamical behavior of the system. This relation is given by Eqs. (3.12), (3.17) and (3.18). The expectation value of the cube of the quantity X_a is given by the integral (3.13). Conversely, the nonlinear susceptibilities $\chi_{abc}(\omega_1,\omega_2)$ are determined by the fluctuation functions $\Phi_{abc}(\omega_1;\omega_2)$.

In a subsequent paper we apply the given theorem to electromagnetic fluctuations. It turns out that in the general case (and not only for a transparent medium) the parametric fluctuations are determined by the nonlinear susceptibilities.

In conclusion, I use this occasion to express my deep gratitude to V. M. Faĭn for proposing to study this problem, and also to V. B. Tsaregradskiĭ and M. A. Novikov for discussions and their interest in this work.

APPENDIX

The spectral representations for the functions B and G follow from the definition of the T-ordering operator, in terms of the step functions (2.1), (2.2) and their Fourier representation (cf.[18]):

$$\mathrm{sign}(\tau) = \frac{i}{\pi}\int_{-\infty}^{\infty}d\Omega'\frac{P}{\Omega'}e^{-i\Omega'\tau}. \quad (A.1)$$

For instance, for (2.30) we find

$$B_2^s(\omega_2;\omega_1) = \frac{1}{4}\langle[X_c(\omega_2),[X_a,X_b(\omega_1)]_+]_+\rangle$$
$$+\frac{i}{4\pi}\int_{-\infty}^{\infty}d\Omega_1\frac{P}{\omega_1-\Omega_1}\langle[X_c(\omega_2),[X_a,X_b(\Omega_1)]_-]_+\rangle. \quad (A.2)$$

Whence

$$B_2'^s(\omega_2;\omega_1) = \tfrac{1}{4}\langle[X_c(\omega_2),[X_a,X_b(\omega_1)]_+]_+\rangle, \quad (A.3)$$

$$B_2''^s(\omega_2;\omega_1) = \frac{1}{4\pi}\int_{-\infty}^{\infty}d\Omega_1\frac{P}{\omega_1-\Omega_1}\langle[X_c(\omega_2)[X_aX_b(\Omega_1)]_-]_+\rangle \quad (A.4)$$

(The Fourier transforms of the correlation functions are real for $\epsilon^a\epsilon^b\epsilon^c = 1$, cf.[18].)

---

6)Equation (3.19′) agrees with the derivation of the fluctuation-dissipation theorem for stationary states in the paper of Grafov and Levich /[18]/.

7)The identity (3.23) is a consequence of the relation

$$\eta(\tau_1-\tau_2)\eta(\tau_2)+\eta(\tau_2-\tau_1)\eta(\tau_1) = \eta(\tau_1)\eta(\tau_2)$$

or in spectrally decomposed form

$$\frac{1}{-\omega_0+i\varepsilon}\frac{1}{\omega_1+i\varepsilon}+\frac{1}{-\omega_0+i\varepsilon}\frac{1}{\omega_2+i\varepsilon} = \frac{1}{\omega_1+i\varepsilon}\frac{1}{\omega_2+i\varepsilon}. \quad (*)$$

Utilizing the equality $1/(\omega+i\epsilon) = (P/\omega) - i\pi\delta(\omega)$, the real part of (*) yields (3.23).

Considering that

$$B_2'^{a}(\omega_2;\,\omega_1) = {}^1\!/_4\langle[X_c(\omega_2),\,[X_a,\,X_b(\omega_1)]_-]_+\rangle, \tag{A.5}$$

one checks directly that (3.8) is valid.

The proof of (3.9) follows from (A.4) and the spectral representation for $G''(\omega_1, \omega_2)$:

$$-G''(\omega_1,\omega_2) = P_{012}^{\mathrm{cycl}}\frac{1}{4\pi}\int_{-\infty}^{\infty} d\Omega_1 \frac{\mathrm{P}}{\omega_1-\Omega_1}\langle[X_c(\omega_2)[X_aX_b(\Omega_1)]_-]_+\rangle =$$

$$= P_{012}^{\mathrm{cycl}}B_2''^{s}(\omega_2;\,\omega_1), \tag{A.6}$$

where P_{012}^{cycl} is the operator of cyclic permutations 0a, 1b, 2c.

[1] H. B. Callen and T. A. Welton, Phys. Rev. 83, 34 (1951).

[2] H. B. Callen and R. F. Green, Phys. Rev. 86, 702 (1952).

[3] R. Kubo, J. Phys. Soc. Japan 12, 570 (1952).

[4] L. D. Landau and E. M. Lifshitz, Élektrodinamika sploshnykh sred (Electrodynamics of Continuous Media), Gostekhizdat, 1957, [Addison-Wesley, 1965].

[5] S. M. Rytov, Vvedenie v statitsticheskuyu radiofiziku (Introduction to Statistical Radiophysics), Nauka, 1966.

[6] M. L. Levin and S. M. Rytov, Teoriya ravnovesnykh teplovykh fluktuatsiĭ v elektrodinamike (Theory of Equilibrium Thermal Fluctuations in Electrodynamics) Nauka, 1967.

[7] S. M. Rytov, Dokl. Akad. Nauk SSSR 110, 371 (1956) [Sov. Phys. Dokl. 1, 555 (1957)]

[8] M. Lax, Rev. Mod. Phys. 32, 25 (1960).

[9] V. V. Karavaev, Zh. Eksp. Teor. Fiz. 47, 1877 (1964) [Sov. Phys. JETP 20, 1263 (1965)].

[10] S. E. Harris, M. K. Oshman and R. L. Byer Phys. Rev. Lett. 18, 732 (1967).

[11] S. A. Akhmanov, V. V. Fadeev, R. V. Khokhlov and O. N. Chunaev, Report at Symposium on Modern Optics, March 1967, USA; ZhETF Pis. Red. 6, 575 (1967) [JETP Lett. 6, 85 (1967)].

[12] V. M. Faĭn and E. G. Yashchin, Zh. Eksp. Teor. Fiz. 46, 695 (1964) [Sov. Phys. JETP 19, 474 (1964)].

[13] A. A. Abrikosov, L. P. Gor'kov and I. E. Dzyaloshinskiĭ, Metody kvantovoĭ teorii polya v statisticheskoi fizike (Methods of Quantum Field Theory in Statistical Physics) Fizmatgiz, 1962 (Engl. Transl. Prentice-Hall 1963).

[14] L. Kadanoff and G. Baym, Quantum Statistical Mechanics, Benjamin, New York 1962 (Russ. Transl. Mir, 1964).

[15] G. F. Efremov, Zh. Eksp. Teor. Fiz. 51, 156 (1966) [Sov. Phys. JETP 24, 105 (1967)].

[16] P. C. Martin and J. Schwinger, Phys. Rev. 115, 1342 (1959).

[17] W. Bernard and H. B. Callen, Rev. Mod. Phys. 31, 1017 (1959).

[18] B. M. Grafov and V. G. Levich, Zh. Eksp. Teor. Fiz. 54, 951 (1968) [Sov. Phys. JETP 27,

[19] L. D. Landau and E. M. Lifshitz, Statisticheskaya Fizika (Statistical Physics) Fizmatizdat, 1964 (Engl. Transl., Addison Wesley, 1958).

Translated by M. E. Mayer
260