Quantum fluctuations of physical quantities in stationary states

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An admittance-fluctuation theorem is formulated. The Kubo formula is generalized to the case of a nonlinear response. The admittance-fluctuation theorem and the generalized Kubo formula are also considered at nonzero temperatures.

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1. INTRODUCTION

There is a category of physical quantities for which the external action is described in the Hamiltonian by an operator of the type given by Eq. (123.1) in Ref. 1:

$$V=-fx, \tag{1}$$

where f is a generalized force and x is the operator of a given physical quantity. It is then possible to establish a relationship between fluctuations and the behavior of the investigated system under the influence of external actions (see, for example, §§ 123 and 124 in Ref. 1).

An operator of the (1) type may appear in the Hamiltonian also when there is no external action. In this case the Hamiltonian of the system can also be described in the form

$$H=H_0-fx. \tag{2}$$

For example, the Dirac Hamiltonian for a free electron can be represented in the form of Eq. (2) where $H_0=\rho_3 m_0 c^2$, $f=-p_z$, $x=c\alpha_z$ (c is the velocity of light, p_z is the projection of momentum along the z axis, m_0 is the rest mass of an electron, α_z and ρ_3 are the Dirac matrices). We can also assume that

$$H_0=c\alpha_z p_z,\quad f=-m_0c^2,\quad x=\rho_3.$$

In the Hamiltonian of a hydrogen-like atom, we can assume that

$$H_0=-\hbar^2\nabla^2/2m,\quad f=Ze^2,\quad x=1/r$$

(or, $H_0=-Ze^2/r$, $f=\hbar^2/2m$, $x=\nabla^2$). In the polar theory Hamiltonian, we can assume

$$H_0=\sum_{k,\sigma}\varepsilon_k n_{k\sigma}$$

(ε_k is the kinetic energy, $n_{k\sigma}$ is the occupation number of a state with a wave vector k and a spin projection σ), $f=-s$, $x=A$ (s is the number of pairs, A is an integral representing the intra-atomic repulsion of electrons in a pair), etc. These examples show that the relationships established in the investigation of a response to an external action may be important in the Hamiltonian (2) even when discussing the internal properties of the system (i.e., the properties in the absence of an external action).

We can also have cases when the Hamiltonian depends nonlinearly on the generalized force:

$$H=H(f). \tag{3}$$

In such cases we can investigate the influence of f irrespective of whether f is due to external or internal action.

It is shown in Ref. 2 that in the case of the Hamiltonian (2) in which H_0, f, and x are independent of time, and additionally H_0 and x are independent of f, the rms fluctuation x in a stationary state with a wavefunction ψ_n is given by the expression

$$(\Delta x)_n^2=\overline{(x-\bar{x})_n^2}=\langle\psi_n|[xH]_-|\partial\psi_n/\partial f\rangle, \tag{4}$$

where

$$[xH]_-=xH+Hx.$$

In statistical physics the relationship between fluctuations and the response to an external action is usually "revealed only after these quantities are expressed in terms of the temperature of a given body" (Ref. 3). This is illustrated by Eq. (124.14) in Ref. 1:

$$(\Delta x)^2_{f\to 0}=\overline{x^2}_{f\to 0}=kT\left.\frac{\partial\bar{x}}{\partial f}\right|_{f\to 0}=kT\alpha(0), \tag{5}$$

where k is the Boltzmann constant [Eq. (5) is derived allowing for the fact that $\bar{x}_{f\to 0}=0$]. In contrast to Eq. (4), the relationship (5) is obtained in the temperature limit ($kT\gg\hbar\omega$) and also assuming that $f\to 0$.

The following expression for the real part of the static susceptibility is obtained from Eq. (5):

$$\alpha(0)=\left.\frac{\partial\bar{x}}{\partial f}\right|_{f\to 0}=\frac{1}{kT}(\Delta x)^2_{f\to 0}. \tag{6}$$

Fluctuations in this case may be thermal. For example, in the theory of magnetism an equation of the (6) type is obtained for the differential paramagnetic susceptibility χ in the limit $h=0$ (h is the intensity of a static and homogeneous magnetic field) when the Hamiltonian is

$$H=H_0-\mu_z h \tag{7}$$

provided $[\mu_z H_0]_-=[\mu_z H]_-=0$ (μ_z is the operator of the magnetic moment projection). In this case [see, for example, Eq. (2.37) in Ref. 4], we have

$$\chi=\left.\frac{\partial\bar{\mu}_z}{\partial h}\right|_{h=0}=\frac{1}{kT}\left.\overline{(\mu_z-\bar{\mu}_z)^2}\right|_{h=0}, \tag{8}$$

where $\bar{\mu}_z|_{h=0}=0$. Since $[\mu_z H]_-=0$ in the case under discussion, it follows from Eq. (4) that quantum fluctuations of μ_z vanish. On the other hand, in the case of the Van Vleck paramagnetism when $[\mu_z H]_-\neq 0$, we can expect a contribution of quantum fluctuations to $\partial\bar{\mu}/\partial h$.

The question arises whether in the limit $T=0$ without assuming that f is small we can derive a relationship analogous in a sense to Eq. (6). Such a relationship would allow us, firstly, to apply to a stationary state when investigating the influence not only of external but also of internal generalized forces; secondly, it should make it possible to identify the role of quantum fluctuations and give the limiting value (related to these fluctuations) of the generalized differential susceptibility in the limit $T\to 0$.

An affirmative response to the above question [naturally after some modification of Eq. (6), particularly a after replacement of kT with the difference between the energies independent of temperature] is given by the admittance-fluctuation theorem formulated in the present paper.

2. ADMITTANCE-FLUCTUATION THEOREM

In the case of dissipative processes we have the fluctuation-dissipation theorem, which relates fluctuations of the physical quantities to the Fourier components of the imaginary part of the susceptibility [see, for example, Eq. (124.10) in Ref. 1]. We shall show that in a stationary state when there are no dissipative processes there is a relationship between the generalized differential susceptibility, i.e., the admittance $\partial x/\partial f$, and the Fourier components of the fluctuations $(\Delta x)_\omega^2$.

Differentiation with respect to f of the average value

$$\bar{L}_n=\langle n|L|n\rangle, \tag{9}$$

where L is the operator of a physical quantity, and allowance for the fact that, according to Eqs. (11.2) and (11.3″) in Ref. 5 and subject to the conditions $m\neq n$ and $E_m\neq E_n$,

$$\left\langle m\left|\frac{\partial n}{\partial f}\right.\right\rangle=\frac{\langle m|\partial H/\partial f|n\rangle}{E_n-E_m}, \tag{10}$$

gives

$$\frac{\partial}{\partial f}\langle n|L|n\rangle=\left\langle n\left|\frac{\partial L}{\partial f}\right|n\right\rangle+\sum_{m(\neq n)}\frac{\langle n|\partial H/\partial f|m\rangle\langle m|L|n\rangle}{E_n-E_m}+\text{c.c.} \tag{11}$$

If the dependence of H on f is linear, Eq. (11) reduces to Eq. (2.6) of Ref. 6.

For the Hamiltonian (2) we find that if $\partial H_0/\partial f=0$, $\partial x/\partial f=0$, and—consequently—$\partial H/\partial f=-x$, it follows from Eq. (11) that

$$\frac{\partial\bar{x}_n}{\partial f}=2\sum_{m(\neq n)}\frac{|\langle n|x|m\rangle|^2}{E_m-E_n}. \tag{12}$$

Equation (12) can be written in the form

$$\frac{\partial\bar{x}_n}{\partial f}=\frac{2}{\hbar}\int_{-\infty}^{\infty}\sum_{m(\neq n)}\frac{|\langle n|x|m\rangle|^2}{\omega}\delta(\omega-\omega_{mn})\,d\omega, \tag{13}$$

where $\omega_{mn}=(E_m-E_n)/\hbar$, and $2\pi\hbar$ is the Planck constant. The admittance-fluctuation theorem follows from Eq. (13):

$$\frac{\partial\bar{x}_n}{\partial f}=\frac{2}{\hbar}\int_{-\infty}^{\infty}\frac{(\Delta x)_{n\omega}^2}{\omega}d\omega, \tag{14}$$

where the Fourier components of the rms fluctuations

$$(\Delta x)_{n\omega}^2=\sum_{m(\neq n)}|\langle n|x|m\rangle|^2\delta(\omega-\omega_{mn}) \tag{15}$$

satisfy

$$\int_{-\infty}^{\infty}(\Delta x)_{n\omega}^2d\omega=\sum_{m(\neq n)}|\langle n|x|m\rangle|^2=(\Delta x)_n^2. \tag{16}$$

The admittance-fluctuation theorem and the fluctuation-dissipation theorem solve in a sense mutually inverse problems. As is known, according to the fluctuation-dissipation theorem, fluctuations are governed by the properties of a response to "an external action no matter how weak" (Ref. 1). However, according to the admittance-fluctuation theorem represented by Eq. (14), a differential response to an external action (which need not be weak) is governed by fluctuations in a stationary state.

Certain properties of the generalized differential susceptibility in a stationary state[7] follow from the admittance-fluctuation theorem expressed in the form (12) if the Hamiltonian is given by Eq. (2): 1) this susceptibility is positive in the ground state; 2) if the energy spectrum of the system is bounded, then the susceptibility is negative in the most excited state; 3) if the spectrum of the system consists of a finite number of levels, the sum of the generalized differential susceptibilities vanishes; 4) if x does not fluctuate and has a definite value in the n-th stationary state, the corresponding generalized differential susceptibility vanishes.

The admittance-fluctuation theorem in the form of Eq. (12) also yields the inequality

$$\partial\bar{x}_g/\partial f\leqslant 2(\Delta x)_g^2/(E_1-E_g), \tag{17}$$

where E_1-E_g is the minimum energy of the excitation of the system, representing the difference between the energies of the first excited (E_1) and ground (E_g) states. According to Eq. (17), the maximum possible value of the generalized differential susceptibility in the ground state, given by the right-hand side of Eq. (17), increase in the minimum excitation energy of the system.

3. GENERALIZATION OF THE KUBO FORMULA

If the fluctuation-dissipation theorem is considered in the inverse aspect, it yields the dependence of the imaginary part of the susceptibility on the correlation function [see, for example, Eq. (126.1) in Ref. 1]. A shortcoming of this dependence is that it gives only the imaginary part and not the total susceptibility. This shortcoming is removed by the Kubo formula, obtained for a linear response. In the static case this formula is [see, for example, Eq. (126.8) in Ref. 1]

$$\alpha_n(0)=\frac{i}{\hbar}\int_0^{\infty}\langle n^0|[x(t)x(0)]_-|n^0\rangle dt. \tag{18}$$

In the derivation of Eq. (18) the wave functions of the perturbed states are considered in the first approximation of perturbation theory. Therefore, the averaging in Eq. (18) is carried out over the n-th unperturbed [part $-fx$ of the Hamiltonian (2)] stationary state $|n^0\rangle$. The first perturbation-theory approximation for wave functions corresponds to the second approximation for energy. Therefore, the Kubo formula of Eq. (18) has the same boundedness as that expected of the second-order perturbation theory. Since the admittance-fluctuation theorem of Eqs. (12) and (14) is not derived on the assumption that f is small, we can expect the Kubo formula of Eq. (18) to be generalized also to the case of a nonlinear response. This generalization is indeed possible. We shall prove this bearing in mind that, in the same way as Eq. (126.4) is transformed into Eq. (126.8) in Ref. 1, the right-hand side of Eq. (12) can be modified to

$$2\sum_{m(\neq n)}\frac{|\langle n|x|m\rangle|^2}{E_m-E_n}=\frac{i}{\hbar}\int_0^\infty\langle n|[x(t)x(0)]_-|n\rangle dt. \tag{19}$$

Equations (12) and (19) yield the generalized Kubo formula

$$\frac{\partial\bar{x}_n}{\partial f}=\frac{i}{\hbar}\int_0^\infty\langle n|[x(t)x(0)]_-|n\rangle dt. \tag{20}$$

The right-hand side of this formula has the same formal structure as the right-hand side of the Kubo formula of Eq. (18). However, in contrast to the Kubo formula (18), $|n\rangle$ in Eq. (20) is the exact eigenfunction of the Hamiltonian (2) so that Eq. (20) includes all the perturbation theory orders and can be applied also in the case of a nonlinear response. The formula (20) is valid also when the average value $\bar{x}_n$ differs from zero for $f=0$ [the Kubo formula (18) is derived on the assumption that $\bar{x}_n|_{f=0}=0$; see for example, §§ 123 and 126 in Ref. 1]. It follows that the admittance-fluctuation theorem is equivalent to the Kubo formula generalized to the case of a nonlinear response.

4. ADMITTANCE-FLUCTUATION THEOREM AND THE GENERALIZED KUBO FORMULA AT $T\neq 0$

Applying the rule for the differentiation of an exponential function of an operator [see, for example, Eq. 12.14) in Ref. 8], we find that in the case of the Hamiltonian (2),

$$\frac{\partial\langle x\rangle}{\partial f}=\frac{1}{\theta}\int_0^1[\langle xx(\tau)\rangle-\langle x\rangle\langle x(\tau)\rangle]d\tau, \tag{21}$$

where

$$x(\tau)=e^{-\tau H/\theta}xe^{\tau H/\theta}, \tag{22}$$

$\theta=kT$; $\langle\ldots\rangle$ is the averaging over a Gibbs distribution. The admittance-fluctuation theorem follows from Eqs. (21) and (22):

$$\frac{\partial\langle x\rangle}{\partial f}=\sum_n W_n\sum_m\frac{|\langle n|x|m\rangle|^2}{E_n-E_m}\left(\exp\left\{\frac{E_n-E_m}{\theta}\right\}-1\right)-\frac{\langle x\rangle^2}{\theta}, \tag{23}$$

where

$$W_n=\exp\left(-\frac{E_n}{\theta}\right)\Big/\sum_n\exp\left(-\frac{E_n}{\theta}\right). \tag{24}$$

Bearing in mind that

$$\frac{i}{\hbar}\sum_n W_n\int_0^\infty dt\langle n|[x(t)x(0)]_-|n\rangle$$
$$=\sum_m\sum_n W_n\frac{|\langle n|x|m\rangle|^2}{E_n-E_m}\left(1-\exp\left\{\frac{E_n-E_m}{\theta}\right\}\right), \tag{25}$$

we obtain from Eq. (23) the generalized Kubo formula

$$\frac{\partial\langle x\rangle}{\partial f}=\frac{i}{\hbar}\int_0^\infty\langle[x(t)x(0)]_-\rangle dt-\frac{\langle x\rangle^2}{\theta}. \tag{26}$$

The admittance-fluctuation theorem described by Eq. (23) can be rewritten in the form

$$\frac{\partial\langle x\rangle}{\partial f}=\frac{1}{\hbar}\int_{-\infty}^{\infty}\left[1-\exp\left(-\frac{\hbar\omega}{\theta}\right)\right]\sum_{m,n}W_n\frac{|\langle n|x|m\rangle|^2}{\omega}\times\delta(\omega+\omega_{nm})d\omega-\frac{\langle x\rangle^2}{\theta}. \tag{27}$$

Bearing in mind that according to § 124 in Ref. 1

$$(x^2)_\omega=\pi(1+e^{-\hbar\omega/\theta})\sum_{m,n}W_n|\langle n|x|m\rangle|^2\delta(\omega+\omega_{nm}), \tag{28}$$

we find from Eq. (27) that

$$\frac{\partial\langle x\rangle}{\partial f}=\frac{1}{\pi\hbar}\int_{-\infty}\frac{(x^2)_\omega}{\omega}\operatorname{th}\frac{\hbar\omega}{2\theta}d\omega-\frac{\langle x\rangle^2}{\theta}. \tag{29}$$

If the inequality $kT\gg\hbar\omega$ is satisfied by all the important frequencies, then in the high-temperature limit we obtain from Eq. (29), using Eq. (122.6) of Ref. 1, a dependence of the (8) type:

$$\partial\langle x\rangle/\partial f=(\langle x^2\rangle-\langle x\rangle^2)/\theta, \tag{30}$$

i.e., the quantum constant $\hbar$ drops out and $\partial\langle x\rangle/\partial f$ is governed by classical fluctuations. As is known, the same expression is obtained from the Gibbs distribution in classical statistics (see, for example, Ref. 1).

If in the case $f\to 0$, we find that $\langle x\rangle\to 0$, Eq. (29) gives

$$\alpha(0)=\lim_{f\to 0}\frac{\partial\langle x\rangle}{\partial f}=\int_{-\infty}^{\infty}\alpha_\omega(0)d\omega, \tag{31}$$

where

$$\alpha_\omega(0)=\frac{1}{\pi\hbar}\frac{(x^2)_\omega}{\omega}\operatorname{th}\frac{\hbar\omega}{2\theta}. \tag{32}$$

It follows from Eq. (32) above and from Eq. (122.6) of Ref. 1 that

$$\langle x^2\rangle_{f\to 0}=\frac{1}{2}\hbar\int_{-\infty}^{\infty}\omega\alpha_\omega(0)\operatorname{cth}\frac{\hbar\omega}{2\theta}d\omega. \tag{33}$$

This expression, obtained from the admittance-fluctuation theorem (29) in the limit $f\to 0$ and on condition $\langle x\rangle_{f\to 0}=0$, can be related to the fluctuation-dissipation theorem. In fact, in the limit $f\to 0$, when the response is linear, we can use the Kramers–Kronig relationships [see, for example, Eq. (123.15) in Ref. 1], which yields

$$\alpha_\omega(0)=\alpha_\omega''/\pi\omega, \tag{34}$$

where α_ω' is the imaginary part of the susceptibility. It follows from Eqs. (33) and (34) that

$$\langle x^2\rangle_{f\to 0}=\frac{\hbar}{2\pi}\int_{-\infty}^{\infty}\alpha_\omega'\operatorname{cth}\frac{\hbar\omega}{2\theta}d\omega, \tag{35}$$

which is identical with the fluctuation-dissipation theorem given by Eq. (124.10) in Ref. 1. Thus, in the limiting case considered here, we find that the fluctuation-dissipation theorem follows from the admittance-fluctuation theorem expressed in the form of Eq. (29) and from the Kramers–Kronig relationships.

In considering the limit $\theta\to 0$, it is convenient to rewrite Eq. (23) in the form

$$\frac{\partial\langle x\rangle}{\partial f}=\frac{1}{\theta}\left(\sum_n W_n|\langle n|x|n\rangle|^2-\langle x\rangle^2\right)+(x,x), \tag{36}$$

where (x,x) is a scalar product (see, for example, Ref. 9):

$$(x,x)=\sum_m\sum_n\frac{|\langle m|x|n\rangle|^2}{E_n-E_m}(W_m-W_n)\quad (m\neq n). \tag{37}$$

In the limit $\theta\to 0$, the first term on the right-hand side of Eq. (36) tends to zero. Therefore,

$$\lim_{\theta\to 0}\frac{\partial\langle x\rangle}{\partial f}=\lim_{\theta\to 0}(x,x). \tag{38}$$

According to Eq. (37),

$$\lim_{\theta\to 0}(x,x)=2\sum_{m(\neq g)}\frac{|\langle m|x|g\rangle|^2}{E_m-E_g}. \tag{39}$$

Equations (38) and (39) yield

$$\lim_{\theta\to 0}\frac{\partial\langle x\rangle}{\partial f}=\frac{\partial\bar{x}_g}{\partial f}, \tag{40}$$

where $\partial\bar{x}_g/\partial f$ is determined by the admittance-fluctuation theorem of Eq. (12). It therefore follows from Eq. (36) that the generalized differential susceptibility consists of two parts, one of which is "thermal" and tends to zero in the limit $\theta\to 0$, whereas the other is related to quantum fluctuations and reduces in the limit $\theta\to 0$ to the admittance-fluctuation theorem (12) for the ground stationary state.

It should be noted that in the case of a two-level system and a negative temperature, the first part of the admittance-fluctuation theorem (36) also tends to zero in the limit $\theta\to -0$, in the second part reduces to the admittance-fluctuation theorem (12) for an excited stationary state.

According to Eq. (11), in the case of the Hamiltonian (2) in which $\partial H_0/\partial f=0$ and $\partial x/\partial f=0$, the admittance-fluctuation theorem for a physical quantity y different from x and satisfying the condition $\partial y/\partial f=0$, has the following form at $T=0$:

$$\frac{\partial\langle n|y|n\rangle}{\partial f}=\sum_{m(\neq n)}\frac{\langle n|x|m\rangle\langle m|y|n\rangle}{E_m-E_n}+\text{c.c.} \tag{41}$$

In this case the rule for the differentiation of an exponential function of an operator gives not Eq. (21) but

$$\frac{\partial\langle y\rangle}{\partial f}=\frac{1}{\theta}\int_0^1[\langle yx(\tau)\rangle-\langle y\rangle\langle x(\tau)\rangle]d\tau. \tag{42}$$

From Eqs. (42) and (22), we now have the admittance-fluctuation theorem in the form different from Eq. (23):

$$\frac{\partial\langle y\rangle}{\partial f}=\sum_{n,m}W_n\frac{\langle n|y|m\rangle\langle m|x|n\rangle}{E_n-E_m}\left[\exp\left(\frac{E_n-E_m}{\theta}\right)-1\right]-\frac{\langle y\rangle\langle x\rangle}{\theta}. \tag{43}$$

This equation can be written in the form

$$\frac{\partial\langle y\rangle}{\partial f}=\frac{1}{\theta}\left[\sum_n W_n\langle n|y|n\rangle\langle n|x|n\rangle-\langle y\rangle\langle x\rangle\right]+(y,x), \tag{44}$$

where the scalar product is

$$(y,x)=\sum_m\sum_n\frac{\langle n|y|m\rangle\langle m|x|n\rangle}{E_n-E_m}(W_m-W_n)\quad (m\neq n). \tag{45}$$

The scalar products (x,x), (y,x), and (y,y) satisfy the Cauchy-Bunyakovskii-Schwarz inequality:

$$(x,x)(y,y)\geqslant|(x,y)|^2. \tag{46}$$

Equations (46), (45), and (36) yield the corresponding inequality for the generalized differential susceptibility.

The Cauchy-Bunyakovskii-Schwarz inequality gives also (see, for example, Ref. 10)

$$\sum_n W_n|\langle n|x|n\rangle|^2\geqslant\langle x\rangle^2. \tag{47}$$

According to Eqs. (47) and (36) when $\theta>0$, then

$$\partial\langle x\rangle/\partial f\geqslant(x,x). \tag{48}$$

If we add and subtract $\langle x^2\rangle/\theta$, to and from Eq. (26), we obtain

$$\frac{\partial\langle x\rangle}{\partial f}=\frac{1}{\theta}(\langle x^2\rangle-\langle x\rangle^2)-\frac{1}{\theta}\sum_n W_n(\Delta x)_n^2+(x,x). \tag{49}$$

The last two terms on the right-hand side of Eq. (49) represent the deviation of the generalized differential susceptibility from the expression with the structure of Eq. (30) or from the Kirkwood formula in the form of Eq. (8). Equation (49) and the inequality (48) lead to

$$\langle x^2\rangle-\langle x\rangle^2\geqslant\langle(\Delta x)_n^2\rangle, \tag{50}$$

which also follows directly from Eq. (47).

Applications of the results obtained above will be published elsewhere.

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