The noise of a nonequilibrium medium and nonlinear, fluctuation–dissipation reciprocity relations

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1. The magnitude and spectral composition of thermal noise in an arbitrary system in thermodynamic equilibrium can be found using the fluctuation–dissipation theorem[1] (FDT) if the temperature of the system and its linear dissipative characteristics are known. The converse use of the FDT and the allied relations (Planck's formula, Kirchhoff's law) is also possible – to measure the dissipative characteristics and/or the temperature of a medium from its intrinsic noise and thermal electromagnetic emission. Thus, the method developed in Refs. 2 and 3 (see also Ref. 4) permits a differential determination of the temperature of internal human organs from their emission in the UHF range.

The accuracy of such measurements is limited by the fact (along with other factors[2,3]) that the system being studied is practically always in a thermodynamically nonequilibrium (although steady) state, and the FDT is invalid for such a state. Continuously occurring transport processes, accompanied by irreversible entropy production and energy dissipation, are a feature of a nonequilibrium steady state (NSS). The fluctuation emission of a nonequilibrium medium differs in its intensity and spectrum, generally speaking, from the emission of an equilibrium medium with the same complex conductivity and the same internal temperature T. The latter is the quantity which a thermometer placed inside the medium would read; in other words, T is the thermostatic temperature, i.e., most of the microscopic degrees of freedom remain in quasi-equilibrium. It is convenient to characterize this difference by the effective emission temperature T', distinct for each frequency (T' is introduced by analogy with the Nyquist or Kirchhoff formulas for equilibrium noise).

The difference between T' and T is the cause of a certain error if the aim is the remote determination of the internal temperature T of the medium. On the other hand, the difference between T' and T (or the difference between T' at different frequencies) can serve as a source of information about the nonequilibrium of the medium. In the first case one would like to know how significant the error is; in the second case one would like to know what kind (in a fundamental respect) of information about the properties of the nonequilibrium medium is contained in T'.

2. In this paper it is shown that a fully common answer can be given to these two interconnected questions.

That is, a certain nonequilibrium process occurring steadily in a medium affects the fluctuation emission of frequency ω only in the case when an external electromagnetic field of frequency ω is capable of initiating the given transport process in the medium, the effect being greater the greater this capability. Both effects are determined by the same parameters of the medium, although the "elementary" kinetic processes responsible for them are not identical – they are mutually reversed in time. Thus, here we encounter a natural reciprocity relation. In contrast to the Onsager linear relations, however, it connects two nonlinear effects, one of them "dissipative" and the other "fluctuational" (the nonlinearity is already seen from the fact that disturbances of the system at different frequencies are connected).

A well known example of this kind is the photogalvanic effect; the corresponding fluctuational effect is the nonthermal (unconnected with Joule heating) change in the emission of a conductor when a direct current is present in it (in the region linear in the current this effect is described by Efremov's exact quadratic FDT[5]). As an example of a similar effect we can consider the chemiluminescence[6] of chemically nonequilibrium media – in particular, the increased ultraviolet radiation of living plant leaves. The corresponding dissipative effect is the initiation of a chemical reaction by radiation.

We emphasize that it is precisely the initiation of transport processes by radiation that is of fundamental importance, i.e., the capacity of the latter to alter the ratio of the amounts of the mutually opposite "elementary" acts of transport (e.g., to disrupt chemical equilibrium). The fluctuational effect is absent if the radiation simply increases or decreases the number (per unit time) of certain particular acts without changing their proportion.

The reciprocity relation obtained below is expressed quantitatively (in the simple, "discrete" variant) by the formula

$$T'/T = 1 - \eta, \tag{1}$$

where η is that fraction of the total absorbed power of weak external radiation which is absorbed (dissipated) through the participation of the radiation in nonequilibrium transport processes taking place in the medium (e.g., through the participation of the radiation in a chemical reaction). In the equilibrium state, $\eta = 0$; the quantities η and T' pertain to the same frequency and degree of nonequilibrium. The case T' > T corresponds to $\eta < 0$; here the nonequilibrium is the source of a negative contribution to absorption. Formula (1) is derived below for the classical region $\omega \ll T/\hbar$, although it is also valid, as can be shown, for higher frequencies ($\omega \gtrsim T/\hbar$) in the form $S'/S = 1 - \eta$, where S' is the intensity of nonequilibrium emission and S is the intensity which the emission of an equilibrium medium with the same absorption would possess.

This result allows us to state that in the case of a medium such as the tissues of a living organism, the error in a radio measurement of T as a result of nonequilibrium should be negligibly small. The quantity η characterizes the relative role of the nonthermal action of the radiation being absorbed. Although a living medium is essentially nonequilibrium in a chemical respect, here the dissipation of UHF radiation is due mainly to the mobility of the entire mass of ions and polar molecules or groups, i.e., to mechanisms that act independently of reaction processes and are essentially equilibrium mechanisms. Therefore, the quantity η is small. Accordingly, almost all the self-emission owes its origin to the equilibrium thermostat – the thermal motion of ions and polar molecules. Indeed, experiments[2] give no indications of a difference between T' and T that is not connected with nonequilibrium.

Clearly, however, the "energetic" smallness of η does not at all mean that the biological action of a UHF field is weak. Therefore, it would be interesting to attempt to measure η or estimate it on the basis of a concrete biophysical model. On the other hand, in other frequency ranges η may be large. In such a case the measured difference between T' and T can be treated, in accordance with (1), as a characterization of the potential (biological) activity of this kind of radiation.

3. In contrast to the FDT itself, the fundamental physical principles lying at the foundation of the FDT are valid regardless of the degree of proximity of the system to equilibrium. These are, first, the principle of conservation of microscopic phase volume (the Liouville theorem); secondly, the symmetry of the laws of microscopic dynamics with respect to time reversal; thirdly, the causality principle. In Refs. 7-10 it was shown that they lead to universal fluctuation–dissipation relations (FDR) generalizing the FDT.

In their structure the FDR differ considerably from the usual FDT: Besides the quadratic-correlation characteristics of noise, they include more-complicated and subtle indices of noise – cumulant functions, or correlators, of higher orders. In combination the latter carry information about the probabilities of various fluctuation processes. Thus, the connection between dissipation and the quadratic-correlation parameters of nonequilibrium noise (intensity, spectral density) depends on the statistical structure of the noise.

The FDT fully reveal the form of this dependence,[7,8] thereby permitting the constructive use of information or hypotheses about the type of statistics inherent to a given transport process (the very widespread Poisson statistics plays an important role, as is well known) and serving as a basis for the model analysis of the nonequilibrium noise. The consequences of the FDR have a thermodynamic character, since they do not depend on the concrete microscopic mechanisms of the transport.

In brief notation a set of FDR is reduced to the simple formula[7,9]

$$P(\Pi_+)\exp\{-\Delta S(\Pi_+)\} = P(\Pi_-). \tag{2}$$

Here $P(\Pi_+)$ is the probability of a difference in a certain fluctuation process Π_+ in the nonequilibrium system; $P(\Pi_-)$ is the probability that the process Π_- obtained from Π_+ by time reversal is realized;

$$\Delta S(\Pi_+) = \frac{1}{T}\int_0^t j_\alpha(t')x_\alpha(t')dt' + z_\alpha\int_0^t J_\alpha(t')dt' \tag{3}$$

is the change in entropy as a result of the process Π_+; $\Delta S(\Pi_-) = -\Delta S(\Pi_+)$; $x_\alpha(t)$ are the external dynamic forces (fields); z_α are the internal thermal forces[7,8]; $j_\alpha(t)$ and $J_\alpha(t)$ are the fluxes conjugate to these forces; T is the temperature of the thermal part of the system.

4. Let a system be internally nonequilibrium [this is described by the inequality $z \neq 0$ (in the case of chemical nonequilibrium $z = A/T$, where A is the rate of some particular reaction)] and be subjected to an external alternating disturbance x(t) of frequency ω. We shall assume that the forces z are time-symmetric[7,9] and there is no external permanent magnetic field. Then from (2) and (3) we get the equality (the analog of the formulas of Refs. 7 and 9)

$$\left\langle \exp\left[-\frac{1}{T}\int_0^t m(t')dt'\right]\right\rangle = \left\langle \exp\left[-z\int_0^t J(t')dt'\right]\right\rangle, \tag{4}$$

where $m(t) = j(t)x(t)$ is the absorbed power of the alternating disturbance. As applied to an NSS we introduce the function

$$D(z) = -\lim_{t\to\infty}\frac{1}{t}\ln\left\langle \exp\left[-z\int_0^t J(t')dt'\right]\right\rangle. \tag{5}$$

Taking the alternating disturbance as (infinitely) weak, from (4) we have

$$\frac{S_m}{2TM} = 1 - \frac{T}{M}D(z), \tag{6}$$

where $M \equiv \langle m(t)\rangle$; S_m is the spectral density of the fluctuations m(t) (at zero frequency); $M \to 0$. We note that as $x(t) \to 0$ and $M \to 0$ the higher cumulants of m(t) are proportional to M^2, M^3,... and therefore do not appear in (6). The ratio $S_m/M = S_j(\omega)/g(\omega)$, where $S_j(\omega)$ is the spectral density of the thermal fluctuations of the current j(t) at the frequency ω, while $g(\omega)$ is the real part of the differential (linear) conductivity at the same frequency (both these quantities depend on z).

If the system is internally equilibrium ($z = 0$), then $D(z) \equiv 0$ and (6) is reduced to the FDT. As is seen from (4) and (5), $D(z) = 0$ for $z \neq 0$ too if there is no external alternating disturbance ($M = 0$). In such a case the equality $D(z) = 0$ expresses conservation of the microscopic phase volume.[7,9]

For definiteness we shall discuss below an electromagnetic field and chemical nonequilibrium ($z \neq 0$). In the presence of a field the variation of the phase volume in the course of the reaction (in the chemical "channel") will no longer equal zero; in a time t it decreases by a factor of $\exp[tD(z)]$. This decrease is compensated for by an equal and opposite phase-volume flux created in the electrical channel. Thus, the quantity D(z) characterizes the interconnection between the electrical and chemical processes, i.e., the intensity of the cross processes. It equals the increment (per unit time) of the logarithm of the phase volume, i.e., the entropy increment caused in the electrical channel by the cross processes [in the absence of the latter, $D(z) = 0$ and the electrical noise is described by the usual FDT even in the nonequilibrium state]. Hence it follows that $TD(z)$ is the field power taking part in the cross processes. Designating $\eta = TD(z)M^{-1}$ and introducing the effective temperature $T' = S_m/2M$, we arrive at formula (1).

Let us illustrate this conclusion. In a weakly nonequilibrium state ($z \ll 1$) from (5) and (6) we have $D(z) \approx z\langle J\rangle$ and in the linear approximation in z

$$\frac{T'}{T} = 1 - \frac{zT\langle J\rangle}{M}, \qquad (7)$$

where $\langle J\rangle \propto M$ is the average flux arising (for $z = 0$) as a result of "rectification" of the variable field; $Tz\langle J\rangle$ is the work of the force against this flux. An expression analogous to (7) for the "electrical" case, when $z = U/T$ and U is the constant emf on the nonequilibrium conductor, was obtained in Ref. 5 as a consequence of the quadratic FDT (Refs. 5 and 11). We note that this expression was confirmed (within the limits of the fulfillment of the condition of weak nonequilibrium) experimentally in Ref. 12 for the example of the noise of electrolytic cells.

Let us consider the case of strong nonequilibrium ($z \gg 1$), taking simple Poisson statistics for the reaction; then

$$D(z) = -[n_+(e^{-z} - 1) + n_-(e^{z} - 1)],$$

where n_+ and n_- are the numbers of forward and reverse reaction acts per unit time. As a consequence of (2), $n_- \, n_+ = \exp(-\Delta s)$; Δs is the entropy increment in a forward act. We can write $\Delta s = z + \varepsilon/T$, where ε is the field energy dissipated in a forward act. When $M \to 0$ we have $\varepsilon \to 0$ and $D(z) = (\varepsilon/T)n_+(1 - e^{-z}) = \eta(M/T)$, since $\varepsilon n_+(1 - e^{-z})$ is the power "oonsumed' in the reaction. Thus, we again arrive at (1). And since $n_+(1 - e^{-z}) = \langle J\rangle$, where $\langle J\rangle$ is the reaction rate, we have $\eta = \langle J\rangle\varepsilon/M$.

The relations obtained can be translated to the langugage of distributed systems without fundamental changes.

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A spherical explosion with intense emission in a confined gas cloud

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When shock waves are used as powerful radiators one of the main problems consists in liberating the radiation created in the shock-compressed plasma to large distances from the front. When a relatively low-amplitude wave propagates in an unbounded medium, only quanta with an energy ε less than the transparency limit, the value ε_* of which is close to the value I_1 of the first ionization potential of the working gas, escape to large distances. As the velocity of the wave and the gas temperature behind its front increase, an ever greater fraction of the radiation belongs to quanta with $\varepsilon > I_1$. Such radiation is absorbed immediately ahead of the front, forming a heated layer. The heated gas in this layer starts to absorb radiation emitted by the front in the long-wavelength part of the spectrum. This limits the brightness of strong shock waves and the magnitude of the flux of radiation emitted by them.[1]

In order to remove these limitations, in Ref. 2 it was proposed to limit the mass of that volume of gas through which the shock wave propagates. Then the escaping radiation flux increases sharply at the instant the heated layer emerges at the boundary between the gas "cloud" and the vacuum. From simple relations at the shock front we can show that the limiting flux density q_r of the emitted radiation flux for a quasi-steady shock wave equals the hydrodynamic energy-flux density $q_h = {}^1/_2\rho_0 D u_s^2$, where D is the wave velocity and u_s is the gas velocity at the front. In such a wave the compression $\sigma = \infty$, the velocity of all the particles $u = u_s$ and $D = u_s$, and the internal energy behind the front $e = 0$ (with the exception of a gas layer of infinitesimal mass immediately at the front, where $e = e_s$).

Let us consider a point explosion, i.e., a problem analogous to that of Ref. 3 but with a very strongly radiating shock wave and infinite compression behind the front. We shall assume that at the initial time ($t = 0$) all the energy E_0 is the kinetic energy E_k of the cool and dense explosion products flying apart with high velocity (the inertial stage of dispersion). In addition, we assume that at $t = 0$ these products consist of an infinitely dense

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