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The Fluctuation-Dissipation Theorem fails for fast superdiffusion

I. V. L. Costa, R. Morgado, M. V. B. T. Lima and F. A. Oliveira

Institute of Physics and International Center of Condensed Matter Physics
University of Brasília, CP 04513, 70919-970, Brasília-DF, Brazil

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Abstract. – We study anomalous diffusion for one-dimensional systems described by a generalized Langevin equation. We show that superdiffusive systems can be divided into two classes: normal and fast. For fast superdiffusion we prove that the Fluctuation-Dissipation Theorem does not hold. As a result, the system acquires an effective temperature. This effective temperature is a signature of metastability found in many complex systems such as spin-glass and granular material.

Introduction. – Since its formulation, the Fluctuation-Dissipation Theorem (FDT) has played a central role [1, 2] in non-equilibrium statistical mechanics in the linear-response regime. It reaches such an importance that a full formulation of statistical physics is given [2] based on it. In the last 30 years, fundamental concepts and methods have been developed [1–6] and a large number of connections have been established. A necessary requirement for the FDT is that the time-dependent dynamical variables are well defined at equilibrium. The presence of nonlinear effects or far-from-equilibrium dynamics may lead to situations where the FDT does not hold, the aging process in spin-glass systems being a good example [7–13].

In this letter, we present a straightforward proof of the inconsistency of the FDT for a certain class of superdiffusive processes described by a generalized Langevin equation (GLE). The surprising result here is the fact that the violation happens in the linear regime, *i.e.* where we expect the validity of the FDT. The use of the FDT allows us to classify two classes of superdiffusion. The first class, which we shall call normal superdiffusion, does obey the FDT; the second class, which we shall call fast superdiffusion, does not obey the FDT. The proof is simple and we discuss as well how the diffusive process leads to an equilibrium.

Diffusion is one of the simplest processes by which a system reaches equilibrium. For normal diffusion, the process is so well known that it may be described by an equilibrium-type distribution for the velocity and position of a particle. However, the strange kinetics of anomalous diffusion, intensively investigated in the last years [14–19], shows surprising results. Consequently, studying anomalous diffusion seems to be a good way to obtain the conditions of validity for the FDT.

Violation of the FDT. – We shall start writing the GLE for an operator A in the form [1,3,5]

$$\frac{\mathrm{d}A(t)}{\mathrm{d}t} = -\int_0^t \Gamma(t-t')A(t')\mathrm{d}t' + F(t), \tag{1}$$

where $F(t)$ is a stochastic noise subject to the conditions $\langle F(t)\rangle = 0$, $\langle F(t)A(0)\rangle = 0$ and

$$C_F(t) = \langle F(t)F(0)\rangle = \langle A^2\rangle_{\mathrm{eq}}\Gamma(t). \tag{2}$$

Here $C_F(t)$ is the correlation function for $F(t)$ and the brackets $\langle\,\rangle$ indicate ensemble average. Equation (2) is the famous Kubo FDT and is quite general [1]. In principle, the presence of the kernel $\Gamma(t)$ allows us to study a large number of correlated processes. The main quantity is the correlation function $C_A(t) = \langle A(t)A(0)\rangle$, from which we can describe most of the process of interest.

We may naively expect that, by eq. (1) and eq. (2), a system will be driven to an equilibrium, *i.e.*

$$\lim_{t\to\infty}\langle A^2(t)\rangle = \langle A^2\rangle_{\mathrm{eq}}. \tag{3}$$

We shall see, however, that this is not always the case for superdiffusive dynamics. Let us define the variable

$$y(t) = \int_0^t A(t')\mathrm{d}t', \tag{4}$$

with asymptotic behavior

$$\lim_{t\to\infty}\langle y^2(t)\rangle \sim t^{\alpha}. \tag{5}$$

For normal diffusion $\alpha = 1$, we have subdiffusion for $\alpha < 1$ and superdiffusion for $\alpha > 1$. Notice that if $A(t)$ is the momentum of a particle with unit mass, $y(t)$ is its position. Using Kubo's definition of the diffusion constant, Morgado *et al.* [19] obtained a general classification for anomalous diffusion; *i.e.* using

$$D = \lim_{z\to 0}\widetilde{C}_A(z) = \lim_{z\to 0}\frac{\langle A^2\rangle_{\mathrm{eq}}}{z+\tilde{\Gamma}(z)}, \tag{6}$$

where $\tilde{\Gamma}(z)$ is the Laplace transform of $\Gamma(t)$, they obtained: For a finite value of $\tilde{\Gamma}(0) \neq 0$, normal diffusion; for $\lim_{z\to 0}\tilde{\Gamma}(z) = 0$, superdiffusion; and for $\lim_{z\to 0}\tilde{\Gamma}(z) = \infty$, subdiffusion. Anomalous diffusion means a time dependence on the diffusion constant [19,20].

Now the Laplace transform of eq. (1) suggests a solution of the form

$$A(t) = \int_0^t R(t-t')F(t')\mathrm{d}t', \tag{7}$$

where we have set $A(0) = 0$, and

$$\tilde{R}(z) = \frac{1}{z+\tilde{\Gamma}(z)}. \tag{8}$$

Squaring eq. (7) and taking the ensemble average we obtain

$$\langle A^2(t)\rangle = 2\int_0^t R(t')\int_0^{t'} C_F(t'-t'')R(t'')\mathrm{d}t''\,\mathrm{d}t'. \tag{9}$$

At this point, it is quite usual to perform numerical calculation [17]. However, we shall show here that very important results can be obtained analytically. From eq. (8), we can get a self-consistent equation for $R(t)$ as

$$\frac{\mathrm{d}R(t)}{\mathrm{d}t} = -\int_0^t \Gamma(t-t')R(t')\mathrm{d}t'. \tag{10}$$

Notice from eq. (6) and eq. (8) that $R(t)$ is the normalized correlation function, *i.e.* $R(t) = C_A(t)/C_A(0)$. By using the FDT equation (2) and eq. (10) we can exactly integrate eq. (9) and obtain

$$\langle A^2(t)\rangle = \langle A^2\rangle_{\mathrm{eq}}\lambda(t), \tag{11}$$

where

$$\lambda(t) = 1 - R^2(t). \tag{12}$$

Equation (3) is satisfied if and only if

$$\lim_{t\to\infty}\lambda(t) = \lambda^* = 1, \tag{13}$$

or, equivalently,

$$\lim_{t\to\infty} R(t) = \lim_{z\to 0} z\tilde{R}(z) = \lim_{z\to 0}\frac{z}{z+\tilde{\Gamma}(z)} = 0, \tag{14}$$

where in the first step we used the final value theorem [21]. Again we shall comment here that if eq. (14) does not hold, its equivalent eq. (3) does not hold as well, and it violates the FDT equation (2). Recently, Lee [6], using his recurrence relation formalism [5], obtained a condition similar to eq. (14) for the ergodicity hypothesis to work. *I.e.* if the system keeps some memory after an infinity time, the ergodicity does not hold. We show here that it is the same condition for the validity of the FDT.

Equation (14) is satisfied for normal and subdiffusion. For superdiffusion $\lim_{z\to 0}\tilde{\Gamma}(z) = 0$, and eq. (14) becomes

$$\lim_{t\to\infty} R(t) = \left(1 + \lim_{z\to 0}\frac{\partial\tilde{\Gamma}(z)}{\partial z}\right)^{-1}, \tag{15}$$

There are two distinct limits for eq. (15), which define two classes of superdiffusion. For the first class, $\lim_{z\to 0}\frac{\partial\tilde{\Gamma}(z)}{\partial z} = \infty$ and the system obeys the FDT. The second class has $\lim_{z\to 0}\frac{\partial\tilde{\Gamma}(z)}{\partial z} \neq \infty$ and it does violate the FDT. The first class we shall call normal superdiffusion (NSD) and the second class fast superdiffusion (FSD).

Consider now the asymptotic behavior for $\tilde{\Gamma}(z)$ as

$$\tilde{\Gamma}(z \longrightarrow 0) = az^{\nu}. \tag{16}$$

It is easy to see that for $\nu < 0$ we have subdiffusion, for $\nu = 0$ normal diffusion, and for $\nu > 0$ superdiffusion. From the above equations we have for $0 < \nu < 1$ NSD and, finally, for $\nu \geq 1$ we have FSD. There is an obvious connection between ν and α, defined in eq. (5), that classify the diffusion. Morgado *et al.* [19] show that $\nu = \alpha - 1$ and, consequently, the FSD starts at $\alpha \geq 2$, *i.e.*, the FDT does not work for the ballistic motion and beyond. Rather, the formalism of GLE + FDT works for $0 < \alpha < 2$. We shall discuss the lower limit $\alpha = 0$ later.

Ballistic motion. – The ballistic motion is at the limit of the validity of the FDT; to go beyond it is too dangerous, so we shall keep working within $\alpha = 2$. Before we give some example, let us make a very important association. The force $F(t)$ in eq. (1) can be obtained from a thermal bath composed of harmonic oscillations, consequently, according to eq. (2), the memory can be put as

$$\Gamma(t) = \int \rho(\omega) \cos(\omega t) \mathrm{d}\omega, \tag{17}$$

where $\rho(\omega)$ is the noise density of states. The same argument used before for the Laplace transform can be used for the Fourier transform, with the simplifying consequence $\tilde{\Gamma}(\omega) = \rho(\omega)$. This is a great advantage, since noise density of states exists not only for systems governed by GLE, but for most of the physical systems. Morgado *et al.* [19] proposed, MOBH conjecture, that: *If a disordered unidimensional system has* $\rho(\omega) \sim \omega^\nu$ *as* $\omega \to 0$, *then the diffusion exponent is*

$$\alpha = \nu + 1. \tag{18}$$

The MOBH conjecture has been observed for the quantum disordered Heisenberg ferromagnetic chain [22], and is under discussion for energy propagation on the harmonic disordered chain [23]. We can choose the density of states to produce the diffusion we want. Consider now

$$\rho(\omega) = \begin{cases} \text{const}, & \omega_1 < \omega < \omega_2, \\ 0, & \text{otherwise}; \end{cases} \tag{19}$$

for $\omega_1 = 0$ we have the Debye density of states for a thermal noise made out of acoustic phonons. Thus, for $\omega_1 = 0$ we have normal diffusion and for any $\omega_1 \neq 0$ we have superdiffusion. This density yields

$$\Gamma(t) = \beta \left[\frac{\sin(\omega_2 t)}{t} - \frac{\sin(\omega_1 t)}{t} \right]. \tag{20}$$

The Laplace transform of eq. (20) gives $\tilde{\Gamma}(z) \sim z$ as $z \to 0$; consequently, $\nu = 1$ and $\alpha = 2$, which is the ballistic limit. If we let $\beta = \omega_2/2$, we get λ^* as

$$\lambda^* = 1 - \left(\frac{2\omega_1\omega_2}{\omega_1 + \omega_2} \right)^2. \tag{21}$$

Values of $\lambda^* \neq 1$ show the inconsistency of the FDT because we start supposing the existence of an equilibrium value $\langle A^2 \rangle_{\text{eq}}$ and, after an infinite time, we end up with $\langle A^2 \rangle_{\text{eq}} \lambda^*$. Equation (21) has a parameter control ω_1, which measures the "hole" in the density of states, and how far we are from the result predicted by the FDT.

Now we select $A(t) = v(t)$, the particle's velocity, so that $\langle v^2(t) \rangle = \langle v^2 \rangle_{\text{eq}} \lambda(t)$. We simulate the GLE for a set of 10000 particles starting at rest at the origin and using the memory in eq. (20) with $\omega_2 = 0.5$ and different values of ω_1. The results of these simulations are shown in fig. 1, where we plot $\langle v^2(t) \rangle$. We used the normalization $\langle v^2 \rangle_{\text{eq}} = 1$, so that $\langle v^2(t) \rangle = \lambda(t)$. Notice that $\lambda(t)$ does not reach a stationary value, rather it oscillates around a final average value λ_s. This value of λ_s should be compared with λ^* obtained from eq. (21).

In fig. 2 we plot λ^* as a function of ω_1 as in eq. (21) with fixed $\omega_2 = 0.5$. We also plot the final average values λ_s obtained from simulations for different values of ω_1. Notice that as ω_1 increases λ^* decreases as expected. The agreement between simulations and eq. (21) shows that we can predict the average value λ_s, even when the FDT does not work.

Now we can define $\lambda^* = T^*/T$, where T^* is an effective temperature for the system. Effective temperatures different from the expected temperature T, or $\lambda^* \neq 1$, are found in

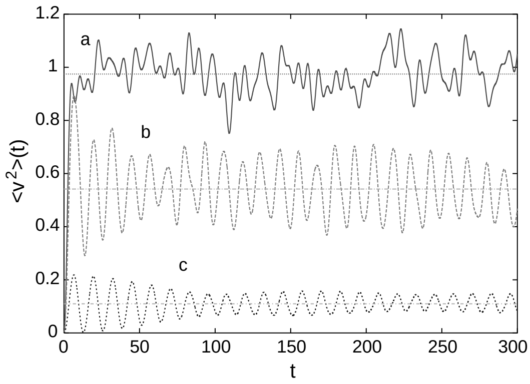

Fig. 1 – Normalized mean-square velocity as a function of time for the memory given by eq. (20). Here $\beta = \omega_2/2$ and $\omega_2 = 0.5$. Each curve corresponds to a different value of ω_1. a) $\omega_1 = 0$; b) $\omega_1 = 0.25$; c) $\omega_1 = 0.45$. The horizontal lines correspond to the final average value λ_s. In agreement with the theoretical prediction, λ_s decreases as ω_1 grows.

spin glasses where the FDT does not work [8–11]. The first observation of such phenomena was reported by Kauzmann [8]. He noticed that if the entropy of a supercooled liquid is extrapolated below the glass temperature T_g, it becomes equal to the crystal temperature $T_\mathrm{c} > 0$, and in some cases even $T_\mathrm{c} < 0$. To avoid this paradox, he suggested the existence of an effective spinodal temperature $T_\mathrm{sp} < T_\mathrm{c}$ in the supercooled liquid phase. In a recent work, Rubi *et al.* [9] investigated the violation of the FDT, using a Fokker-Planck approach. They found temperatures T^* which are greater and smaller than T. Ricci-Tersenghi *et al.* [10] and Cavagna *et al.* [11] performed single-spin-flip Monte Carlo simulations in square lattices with frustration and they obtained effective temperatures $T^* \neq T$. Methods for measuring those effective temperatures are discussed in the literature as well [12].

An oscillatory behavior similar to that found in fig. 1 was observed by Srokowski [17, 18], in his simulations using GLE. However, the kind of motion he studied is a subdiffusive motion. Using his memory, we get [18] $\tilde{\Gamma}(z) = \beta[1 - \exp[\varepsilon z]/(\varepsilon z) + E_1(\varepsilon z)]$; here β is a constant, ε a

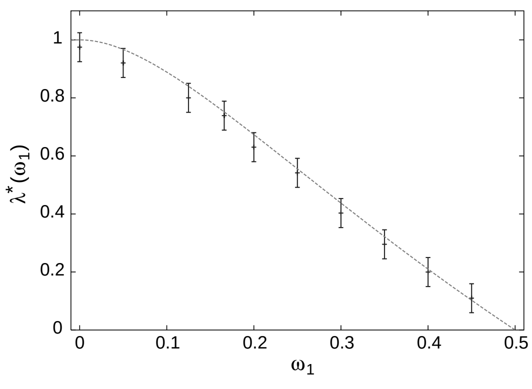

Fig. 2 – λ^* as a function of the parameter ω_1. Each dot corresponds to a value of λ_s obtained from simulations like those described in fig. 1. The line corresponds to the theoretical prediction given by eq. (21).

small number, and E_1 is the exponential integral. This result gives $\lim_{z\to 0}\tilde{\Gamma}(z) \sim z^{-1}$; from the above arguments we get $\nu = -1$ and $\alpha = 0$. We believe we can explain this strange result. Let us consider a constant memory of the form $\Gamma(t) = K$: if $A(t)$ in eq. (1) is the velocity, then the memory term yields $-Ky$, where y is the position. Consequently, the particle is bound to the origin by a harmonic spring and has no diffusive behavior. Indeed, the Laplace transform gives $\tilde{\Gamma}(z) = K/z$, with $\nu = -1$ and $\alpha = 0$, the same as the result we obtain from Srokowski memory. We shall notice that $\alpha = 0$ does not mean that the motion is a harmonic-type motion, rather it means it belongs to the same class; the diffusion behavior of Srokowski is not a power law as in eq. (4), probably it is slower than a power law, such as, for example, a logarithmic behavior.

For $\alpha > 2$, the FSD cannot be described by the methods we used here. For example, eq. (15) gives $R(t \to \infty) = 1$, and together with eq. (12) predicts a null dispersion for the dynamical variable, *i.e.* $\langle A^2(t \to \infty)\rangle = 0$. Moreover, the exponent α can be put as $\alpha = 2/D_{\mathrm{F}}$, where D_{F} is the fractal dimension [24]. Consequently $\alpha > 2$ leads to $D_{\mathrm{F}} < 1$, which is not a full curve, but a set of points such as the Cantor set, and cannot represent a classical trajectory.

Conclusion. – We discussed the stationary behavior for the mean-square value of a dynamical variable $A(t)$ and the mean-square displacement of the quantity $y(t) = \int_0^t A(t')\mathrm{d}t'$ as well. The asymptotic behavior of $\langle y^2(t)\rangle \sim t^\alpha$ as $t \to \infty$ can be explained for $0 < \alpha < 2$. We show that the superdiffusive motion must be classified in normal superdiffusive (NSD) for $1 < \alpha < 2$, and fast superdiffusive (FSD) for $\alpha \geq 2$. The FSD motion shows an inconsistency between the GLE and the FDT. This kind of superdiffusion with $\alpha \geq 2$ is common in hydrodynamical processes, where the use of fractional kinetics [25] and random walk approach [20] may become an alternative way to describe anomalous diffusion. It is not surprising that these processes will be far from equilibrium and violate the FDT. We pointed out here how it happens and precisely where the FDT breaks down. As we have already mentioned, spin glasses seem to be a rich field for studying these phenomena. Indeed, experimental [13] and theoretical works [7, 9, 10] have been reported in this area, confirming the violation of the FDT. Moreover, the effective temperature found in noncrystaline material is connected here with the FSD and the violation of the FDT. It would be very helpful if the exponent α for those diffusive processes could be measured. Other related phenomena are anomalous reaction rate [26] and chaos synchronization [27], which we expect to discuss soon. Although anomalous diffusion remains a surprising phenomenon, we hope that this work will help in the centennial effort to understand diffusion and the relation between fluctuation and dissipation. A generalization of the FDT to include the FSD is necessary, which will require a deeper understanding of systems far from equilibrium.

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