METHODOLOGICAL NOTES

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Why nature needs 1/f noise

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<u>Abstract.</u> While ubiquitous at all levels of organization in nature, including in nanotechnology, low-frequency 1/f noise is not yet understood. A possible reason is the unjustified application of probability theory concepts, primarily that of independence, to random physical phenomena. We show that in the framework of statistical mechanics, no medium can impart a definite diffusivity and mobility to a particle that performs random walk through it, which gives rise to flicker fluctuations in these properties. A universal source of 1/f noise in manyparticle systems in this example is a dependence of the time behavior of any particular relaxation or transport process on the details of the initial microstate of the system as a whole.

Keywords: 1/f noise, molecular random walk, Brownian motion, statistical mechanics of transport processes, dynamical foundations of kinetics, deterministic chaos in many-particle systems

1. Introduction

1.1 Root of the question and a popular hypothesis

For many years, the question of 1/f noise (flicker noise) has grown in urgency, extending and deepening together with physical experiments and new technologies and concerning

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Received 22 January 2015, revised 24 April 2015 Uspekhi Fizicheskikh Nauk **185** (7) 773–783 (2015) DOI: 10.3367/UFNr.0185.201507d.0773 Translated by Yu E Kuzovlev; edited by A M Semikhatov almost all things in the world, from cosmic phenomena down to molecular biology and nanoelectronics. However, there are no modern reviews proportional to the volume and significance of the question — perhaps, because investigators do not find inspirational ideas. True, one appropriate suggestion considered below was already made in [1–4], but it received no response. Meanwhile, today we see reports on more and more inventive and fine measurements of 1/f noise, for instance, in metal and alloy films [5] or atomic layers of graphene [6], but repeatedly without unambiguous indications of its origin. As was noted in [6], "... despite almost a century of research, 1/f noise remains a controversial phenomenon and numerous debates continue about its origin and mechanisms."

It can be added that the "debates," as our experience suggests, not infrequently take rather totalitarian forms. Maybe partly by this reason, from the author's standpoint, the present situation in general barely differs from what was outlined in [1] and a little later in [7]. It is to be compared with the situation in astronomy more than 300 years ago, before the publication of Isaac Newton's celebrated Princi*pia* [8]. We venture such a comparison not for the sake of witticism but in view of our intention to demonstrate in this article that just Newton's laws of mechanics may be the place where the solution to the 1/f noise problem is hidden. More precisely, 1/f noise is an immanent property of systems of many particles moving and interacting by these laws (in their classical or quantum formulation, also including fields). In order to recognize this, one has only to follow Newton's advice to avoid unnecessary hypotheses ("Hypotheses non fingo" [8]).

What hypotheses, then, are proposed by physicists as regards 1/f noise? We discuss this with the example of electric current noise in a conductor at a fixed voltage. The presence of 1/f noise there means that the current has no definite value, in the sense that its averaging (smoothing) over time produces an unpredictable result, randomly varying from one experiment to another, with a scatter that hardly decreases, or even increases, as the averaging interval grows in duration (because the related narrowing of the frequency band contributing to the scatter is almost compensated or even exceeded by the growth of the noise power spectral density inside that band).

As regards the cause of such a phenomenon, one usually first of all assumes that it lies in some specific fluctuation processes influencing the current (for instance, via the number of charge carriers or their mobility), while a feature of these processes is an extremely wide variety of their time scales (of memory, life, relaxation, correlation times, etc.) [5, 6, 9]. Just this is the basic hypothesis.

1.2 Concept of the answer and the strategy

The hypothesis just pointed in Section 1.1 is actually not necessary because the mechanical laws as they are in no way require certainty of the current, and do not therefore require any special reasons for its uncertainty. Indeed, no matter what concrete mechanism of conductivity takes place, if it is indifferent to the amount of charge transported previously through the conductor from one side of an outer electric circuit to the other and hence to a past value of timesmoothed current, then this mechanism continues to be indifferent to them, and on the whole it is unable to set conditions for certainty of the current. It therefore serves as a mechanism of 1/f noise. The mentioned indifference, in turn, is supported by the experimental conditions themselves, which state that fluctuations of the (time-smoothed) current do not induce back reaction of the outer circuit, which passively swallows them.

In this reasoning, no collections of large characteristic times are present; instead, there is a single time only, in practice usually a short one, after which the memory of the conductance mechanism expires. For instance, if it is less than several hours, then the transfer of charge carriers, under their collisions, scatterings, reflections, etc., that occurs now, at the present time interval, is indifferent to what amount of charge was transported yesterday, even if an experimental device was not switched off the preceding night. Correspondingly, at frequencies lower than 1 day⁻¹, one can find 1/f-noise.

Analogously, if somebody possesses unlimited resources for income and expenditures and does not keep count of them, then he himself cannot know how much his expenditures are on a time average, and they should be expected to be distributed over time like 1/f noise.

If, returning to the conductor, we short-circuit it, then the current 1/f noise disappears along with the direct current, but irregular charge displacements in opposite directions continue, again, indifferently to their past amount and therefore to their time-averaged intensity. The latter, therefore, is not aimed at a certain value, which results in 1/f fluctuations of the intensity (power spectral density) of thermodynamically equilibrium white (thermal) current noise. They are connected with the 1/f noise in a nonequilibrium current-carrying conductor by means of the 'generalized fluctuation-dissipation relations' [1, 2, 10–12].

If, on the contrary, one measures equilibrium thermal noise of the potential difference between sides of an open conductor, e.g., an electric junction, then 1/f fluctuations of the intensity of this noise can also be found, which testifies that the sum of numbers (per unit time) of random charge carrier transitions from one side to the other and back is not tracked and controlled by the system, in contrast to the difference of the forward and backward transition numbers [10]. Therefore, the characteristic time constant of the system (equivalent *RC* circuit) determines the upper time scale for fluctuations of the difference and the lower one for fluctuations in the sum (whose upper time scale does not exist because they do not change the system macrostate).

The aforesaid can easily be extended, under nonprincipal substitutions of particular terms and meanings, to other manifestations of 1/f noise in nature. Numerous examples were listed in [1, 2, 4, 10, 11, 13–15]. Our demonstration below is performed in terms of equilibrium 'molecular Brownian motion' [3, 7, 10, 12, 16–23].

Using the simplest tools, we show that the assumption of the certainty of a Brownian particle's diffusivity, or the rate of diffusion, is incompatible with exact equations of statistical mechanics, that is, with the dynamics underlying Brownian motion. Therefore, mechanics inevitably generates 1/f noise, or flicker fluctuations, of diffusivity and mobility of the particle. We then consider quantitative characteristics of this noise and in Section 4 we present their explanation in the language of the theory of deterministic chaos in many-particle systems.

2. Phenomenology of Brownian motion

2.1 Formulation of the problem

We imagine a small 'Brownian' particle in a three-dimensional statistically uniform, isotropic, and thermodynamically equilibrium medium. A very small particle of dust or flower pollen, whose movement in liquid was first observed through a microscope [24, 25] in the 19th century and theoretically analyzed [26–28] in the early 20th century, are suitable objects for us. But it is better to have in mind some 'nanoparticle' or merely a separate atom or molecule in a liquid or gas [29]. In principle, we can even speak about a free charge carrier or a point-like defect in a solid, but confine ourselves to a particle, a subject of the classical version of mechanics.

Let $\mathbf{R}(t)$ and $\mathbf{V}(t) = d\mathbf{R}(t)/dt$ denote vectors of the centerof-mass coordinate and velocity of our Brownian particle (BP) at a given time instant, and \mathbf{R} and \mathbf{V} be their possible values. We assume that initially at time t = 0, the BP was placed at some point of space known with certainty, although it is unimportant where exactly, owing to the spatial homogeneity of the medium and the thermodynamical equivalence of all BP positions. Therefore, it is convenient to choose the coordinate origin: $\mathbf{R}(0) = 0$. Then the instant current position of the BP, $\mathbf{R}(t)$, coincides with the vector of its total displacement, or path, during all the preceding observation time.

We now ask ourselves what the BP 'diffusion law' is, i.e., what the probability distribution of the BP path is. The density of this distribution is denoted by $W(t, \mathbf{R})$. It can be represented as

$$W(t, \mathbf{R}) = \left\langle \delta \left(\mathbf{R} - \mathbf{R}(t) \right) \right\rangle, \tag{1}$$

where the Dirac delta-function is involved, $\mathbf{R}(t)$ is understood as the result of all the preceding interaction between the BP and the medium, and the angular brackets denote averaging over the equilibrium statistical ensemble (Gibbs ensemble [30]) of initial states of the medium and initial values of the BP velocity. Undoubtedly, the plot of $W(t, \mathbf{R})$ as a function of **R** looks like a bell spreading with time. We are interested in what shapes may be taken by this bell in reality.

2.2 Conditional averaging and the continuity equation

In fact, Eqn (1) is a mere identity, but its time differentiation immediately brings us food for thought. From it, we have

$$\frac{\partial W(t, \mathbf{R})}{\partial t} = -\mathbf{\nabla} \langle \mathbf{V}(t) \,\delta\big(\mathbf{R} - \mathbf{R}(t)\big) \rangle$$

Invoking mathematical tools of probability theory [31], we can rewrite this equality as

$$\frac{\partial W(t, \mathbf{R})}{\partial t} = -\nabla \overline{\mathbf{V}}(t, \mathbf{R}) W(t, \mathbf{R}), \qquad (2)$$

where $\overline{\mathbf{V}}(t, \mathbf{R}) = \langle \mathbf{V}(t) \rangle_{\mathbf{R}}$ is the conditional mean of the BP instant velocity determined under the condition that its current position, and hence its previous path, is known (measured) to be equal to $\mathbf{R}(t) = \mathbf{R}$. Generally, the operation of conditional averaging $\langle \ldots \rangle_{\mathbf{R}}$ is defined by the formula

$$\langle \ldots \rangle_{\mathbf{R}} \equiv \frac{\langle \ldots \delta(\mathbf{R}(t) - \mathbf{R}) \rangle}{\langle \delta(\mathbf{R}(t) - \mathbf{R}) \rangle}.$$

Obviously, Eqn (2) is the 'continuity equation' for the probability density $W(t, \mathbf{R})$, and the 'velocity field of the probability flow' $\overline{V}(t, \mathbf{R})$ contains important information about solutions of this equation. Therefore, first of all, we consider the possible construction of the vector function $\overline{\mathbf{V}}(t, \mathbf{R})$.

2.3 Conditional mean velocity of a Brownian particle

We keep in mind that the duration t of our observations of a BP is much longer than the characteristic relaxation time τ of (fluctuations of) the BP velocity.

We then first apply a heuristic argument as follows. On the one hand, by the condition $\mathbf{R}(t) = \mathbf{R}$, the mean of the BP velocity in the past, at the time of its preceding observation, is equal to \mathbf{R}/t . On the other hand, because the BP makes a random walk and $t \ge \tau$, the same condition $\mathbf{R}(t) = \mathbf{R}$ tells us almost nothing about the BP velocity in the future; therefore, its mean at an equal next time interval can be expected to be zero. Hence, because the mean in question, $\overline{\mathbf{V}}(t, \mathbf{R})$, is related to the present time instant 'in the middle between the past and the future', it seems likely that it is equal to the half-sum of the mentioned quantities:

$$\overline{\mathbf{V}}(t,\mathbf{R}) = \frac{\mathbf{R}}{2t} \,. \tag{3}$$

We can confirm this conclusion in a more formally rigorous way, based on the main distinctive statistical property of Brownian motion [27]:

$$\langle \mathbf{R}^2(t) \rangle = \int \mathbf{R}^2 W(t, \mathbf{R}) \, \mathrm{d}\mathbf{R} = 6Dt$$
 (4)

for $t \ge \tau$, stating that the ensemble average of a squared BP displacement grows proportionally to the observation time.

For confirmation, it suffices to note that the continuity equation implies that

$$\frac{\partial}{\partial t} \int \mathbf{R}^2 W \, \mathrm{d}R = 2 \int \mathbf{R} \overline{\mathbf{V}} W \, \mathrm{d}R \,,$$

and that this requirement is naturally satisfied together with Eqn (4) (because $\overline{\mathbf{V}} \| \mathbf{R}$) when equality (3) is valid.

Incidentally, we note that the BP diffusivity, or the diffusion coefficient, D and the relaxation time τ can always be related as

$$D = V_0^2 \, \tau \equiv \frac{T}{M} \, \tau \,,$$

where T is the temperature of the medium, M is the mass of the BP, and $V_0 = \sqrt{T/M}$ is its characteristic thermal velocity.

2.4 General form of the probabilistic law of diffusion and uncertainty of diffusivity

After inserting function (3) into (2), we arrive at the partial differential equation

$$2t \frac{\partial W}{\partial t} = -3W - \mathbf{R} \nabla W, \qquad (5)$$

which clearly suggests a scale-invariant character of its solutions. The isotropic (spherically symmetric) solutions of interest have the form

$$W(t, \mathbf{R}) = (2Dt)^{-3/2} \Psi\left(\frac{\mathbf{R}^2}{2Dt}\right)$$
(6)

with some dimensionless function $\Psi(z)$ of the dimensionless argument $z = \mathbf{R}^2/(2Dt)$. In our context, representing the probability density, it must be nonnegative and satisfy the normalization condition $\int W d\mathbf{R} = 1$ together with equality (4), which can always be ensured. Equation (6) is then the most general law of *diffusional* random walk, when typical BP displacements are proportional to the square root of the observation time: $\mathbf{R}^2(t) \propto t$.

In particular, taking $\Psi(z) = (2\pi)^{-3/2} \exp(-z/2)$, we obtain the commonly known Gaussian diffusion law

$$W = W_D(t, \mathbf{R}) \equiv (4\pi Dt)^{-3/2} \exp\left(-\frac{\mathbf{R}^2}{4Dt}\right).$$
(7)

The corresponding walk is appealing because in a time scale sufficiently coarse in comparison with τ , its successive increments are mutually statistically independent. Owing to this, the only parameter of such random walk—its diffusion coefficient, or diffusivity, D—can be uniquely determined from observations of any of its particular realizations, by means of a long enough time averaging.

However, similar observations of non-Gaussian random walk corresponding to a distribution of general type (6) would produce different values of diffusivity every time [1–4, 7, 10]. Indeed, their coincidence — the convergence of all results of time averaging to the same value — would be impossible without the statistical independence of increments (at least those distant from one another in time), which, in turn, would mean, in accordance with the appropriate limit theorem in probability theory (the 'law of large numbers'), that the probability distribution of the total path tends to the Gaussian (normal) one at $t \ge \tau$ [32].

This becomes quite obvious if distribution (6) is represented by a linear combination of Gaussian bell-shape curves:

$$W(t, \mathbf{R}) = \int_0^\infty W_{\Delta}(t, \mathbf{R}) U\left(\frac{\Delta}{D}, \xi\right) \frac{\mathrm{d}\Delta}{D}$$

Such decompositions naturally arise in microscopic theory [12, 16–18]. Accordingly, instead of $\Psi(z)$ in (6), we can write

$$\Psi(z,\xi) = \int_0^\infty \frac{\exp\left(-z/2\zeta\right)}{\left(2\pi\zeta\right)^{3/2}} U(\zeta,\xi) \,\mathrm{d}\zeta \,. \tag{8}$$

The function $U(\zeta, \zeta)$ here plays the role of the probability distribution of $\zeta = \Delta/D$, which is the random diffusivity Δ expressed in units of the mean diffusivity D. The latter is formally defined by equality (4), while practically one can try to determine it by averaging over many experiments or many identical Brownian particles.

The additional argument ξ in this decomposition, if introduced, for example, as $\xi \equiv \tau/t$ under the convention that $\Psi(z,0) = \Psi(z)$, allows us to take violation of the ideal scale invariance of random walks at $\xi \neq 0$ into account, first of all at the far 'tails' of the diffusion law, where $\mathbf{R}^2 \gtrsim V_0^2 t^2$, that is, $z \gtrsim 1/\xi$. There, the rate of diffusion reaches values of the rate of free flight, $\Delta \sim V_0^2 t = D/\xi$.

Of course, a correction to the tails of the diffusion law may strongly influence its higher-order statistical moments and cumulants, even if $\xi \ll 1$. Be that as it may, nevertheless, the shape of the $W(t, \mathbf{R})$ bell mainly stays almost unchanged. Accordingly, a change in the function $\overline{\mathbf{V}}(t, \mathbf{R})$, required by Eqn (2) and condition (4), is as small as ξ is, and hence expression (3) remains valid.

We note that the very possibility of long-term violation of the scale invariance automatically presumes that the diffusion law is non-Gaussian, because Gaussian statistics merely reserve no place for the violation [which would contradict condition (4)]. This fact gives evidence that the Gaussian law is not a perfect reflection of reality, although in the minds of scientists it is firmly associated with diffusion of physical particles. At the same time, neither the general reasoning leading to (3) and (5) nor Eqn (5) by itself dictates the special Gaussian choice. Therefore, it is desirable to discuss other possibilities and search for criteria of choice among them in statistical mechanics.

3. Microscopic approach

3.1 Newton equation and Liouville equation

We pass from the kinematics of Brownian motion to its dynamics and directly consider a BP interaction with a medium using methods of statistical mechanics. For this, we can take a standard simple Hamiltonian for our system,

$$H = \frac{\mathbf{P}^2}{2M} + \Phi(\mathbf{R}, \Gamma) + H_{\rm th}(\Gamma), \qquad (9)$$

where $\mathbf{P} = M\mathbf{V}$ is the BP momentum, Γ is the full set of (canonical) variables of the medium, $\Phi(\mathbf{R}, \Gamma)$ is the energy of the BP-medium interaction, and $H_{\text{th}}(\Gamma)$ is the Hamiltonian of the medium itself (or, in other words, that of the 'heat bath'). If the BP has internal degrees of freedom, their variables are assumed to be included in the set Γ , thus being formally treated as pertaining to the medium.

Let $\mathcal{D} = \mathcal{D}(t, \mathbf{R}, \mathbf{P}, \Gamma)$ be (the density of) the full probability distribution of states of our system. Its evolution is described by the formally exact Liouville equation [30, 33]. We write the part of it that directly concerns the BP:

$$\frac{\partial \mathcal{D}}{\partial t} = -\mathbf{V} \,\nabla \mathcal{D} - \mathbf{F}(\mathbf{R}, \Gamma) \,\nabla_{\mathbf{P}} \mathcal{D} + \dots \tag{10}$$

Here, $\mathbf{F}(\mathbf{R}, \Gamma) = -\nabla \Phi(\mathbf{R}, \Gamma)$ is the force acting on the BP because of its interaction with the medium, and the ellipsis is for terms with Γ derivatives.

Considering the probability distribution of the BP displacement (coordinate),

$$W(t,\mathbf{R}) = \iint \mathcal{D}(t,\mathbf{R},\mathbf{P},\Gamma) \,\mathrm{d}\Gamma \,\mathrm{d}\mathbf{P}\,,$$

from Eqn (10) after its integration over Γ and **P**, we of course obtain continuity equation (2). The same integration after multiplying (10) by **V** produces the additional equation

$$\frac{\partial}{\partial t} \,\overline{\mathbf{V}}W = -\overline{\mathbf{V}} \,\overline{\mathbf{V}} \circ \overline{\mathbf{V}} \,W + M^{-1} \,\overline{\mathbf{F}}W. \tag{11}$$

Here and below, the symbol \circ denotes the tensor product of vectors, and the overline, as before, means the conditional average at a given $\mathbf{R}(t) = \mathbf{R}$; namely, in the first term in the right-hand side,

$$\overline{\mathbf{V} \circ \mathbf{V}}(t, \mathbf{R}) = \left\langle \mathbf{V}(t) \circ \mathbf{V}(t) \right\rangle_{\mathbf{R}} = \frac{\int \int \mathbf{V} \circ \mathbf{V} \,\mathcal{D} \,\mathrm{d}\Gamma \,\mathrm{d}\mathbf{P}}{W} \,,$$

and in the second term,

$$\overline{\mathbf{F}}(t,\mathbf{R}) = \left\langle \mathbf{F}\big(\mathbf{R}(t),\Gamma(t)\big)\right\rangle_{\mathbf{R}} = \frac{\int \int F(\mathbf{R},\Gamma) \,\mathcal{D} \,\mathrm{d}\Gamma \,\mathrm{d}\mathbf{P}}{W}$$

Equation (11) describes momentum exchange between the BP and the medium and, in essence, as can easily be verified, is merely the Newton equation $M \, dV/dt = F$ expressed in terms of the conditional averaging:

$$\left\langle M \, \frac{\mathrm{d} \mathbf{V}(t)}{\mathrm{d} t} - \mathbf{F} \big(\mathbf{R}(t), \Gamma(t) \big) \right\rangle_{\mathbf{R}} = 0 \, .$$

We transform it into a relation between functions $\overline{\mathbf{F}}(t, \mathbf{R})$ and $W(t, \mathbf{R})$, which can be useful in selecting admissible diffusion laws without going deeper into the Liouville equation.

3.2 Friction equation for a Brownian particle

Replacing the derivative $\partial W/\partial t$ in Eqn (11) with the righthand side of (2), after simple manipulations, we arrive at the equivalent exact equation

$$\frac{\mathrm{d}\overline{\mathbf{V}}}{\mathrm{d}t} + \frac{\mathbf{\nabla}\overline{\mathbf{\nabla}\circ\mathbf{\nabla}}W}{W} = \frac{\overline{\mathbf{F}}}{M}$$
(12)

with the 'material derivative' of the BP average velocity

$$\frac{\mathrm{d}\overline{\mathbf{V}}}{\mathrm{d}t} = \frac{\partial\overline{\mathbf{V}}}{\partial t} + (\overline{\mathbf{V}}\,\mathbf{\nabla})\,\overline{\mathbf{V}}$$

and with the double overline denoting the tensor (matrix) of conditional quadratic cumulants (second-order cumulants) of velocity:

$$\overline{\mathbf{V}\circ\mathbf{V}}\equiv\overline{\mathbf{V}\circ\mathbf{V}}-\overline{\mathbf{V}}\circ\overline{\mathbf{V}}\,.$$

We first consider this last expression.

Because we are speaking about thermodynamically equilibrium Brownian motion, we can state that the conditional cumulant matrix $\overline{\mathbf{V} \circ \mathbf{V}}(t, \mathbf{R})$ at $t \ge \tau$ coincides with the matrix of unconditional equilibrium quadratic statistical

moments of velocity, $\langle \mathbf{V}(t) \circ \mathbf{V}(t) \rangle$, that is, reduces to a number $V_0^2 = T/M$ that is independent of **R**. Indeed, if $t \ge \tau$, then at any **R**, the condition $\mathbf{R}(t) = \mathbf{R}$ fixes the BP position after many random steps and cycles of momentum and energy exchange between the BP and the medium in the framework of the detailed balance between them. Therefore, the value (variance) of the corresponding thermal randomness of the BP velocity is not affected by this condition (otherwise, the thermal kinetic energy of the BP, on average equal to $M\overline{\mathbf{VV}}/2$, would be dependent on where the BP is found).

The <u>above</u> can be confirmed by direct calculation of the matrix $\overline{\mathbf{V} \circ \mathbf{V}}(t, \mathbf{R})$ in the case of a Gaussian random walk subject to distribution (7), which yields

$$\overline{\overline{\mathbf{V}} \circ \mathbf{V}}(t, \mathbf{R}) = V_0^2 \left(1 - \frac{\xi}{2} \right) \to V_0^2$$
(13)

as $\xi \equiv \tau/t \to 0$. This result is also valid for a non-Gaussian walk obeying (6) and (8), because the difference between it and the Gaussian one starts at higher-order cumulants.

We next compare the two terms in the left-hand side of (12). For the first of them, inserting expression (3) gives

$$\frac{\mathrm{d}\overline{\mathbf{V}}}{\mathrm{d}t} = -\frac{\mathbf{R}}{4t^2} = -\frac{\xi}{2}\frac{T}{M}\frac{\mathbf{R}}{2Dt}$$

For the second term, after inserting (13) and (6), we have

$$\left(1-\frac{\xi}{2}\right)\frac{T}{M}\frac{2\,\mathrm{d}\ln\Psi(z,\xi)}{\mathrm{d}z}\frac{\mathbf{R}}{2Dt}\sim-\frac{T}{M}\frac{\mathbf{R}}{2Dt}\,,$$

with the same notation $z = \mathbf{R}^2/(2Dt)$ as before. The righthand expression here corresponds to the Gaussian diffusion law, for which d ln $\Psi/dz = -1/2$, but it is also valid by order of magnitude in general, at least at $z \ll 1/\xi$. This shows that the first term, being approximately $2t/\tau$ times smaller than the second, is negligibly small in the limit $\xi \to 0$.

Hence, consideration of long enough time intervals leads us from (12) to the shortened relation

$$-\left[\frac{T}{D}\left(-\frac{2\,\mathrm{d}\ln\Psi(z,\xi)}{\mathrm{d}z}\right)\right]\frac{\mathbf{R}}{2t} = \overline{\mathbf{F}}\,,\tag{14}$$

which resembles the equation of viscous friction, with $\mathbf{R}/(2t) = \overline{\mathbf{V}}$ playing the role of velocity of a body moving through a fluid, while the role of the friction coefficient is played by the contents of the square brackets.

One more simplification can be obtained, under the mentioned limit, by neglecting scale invariance violation and treating $\Psi(z, \xi)$ as a function of a single argument, $\Psi(z)$. In Sections 3.3–3.5, we first proceed this way.

But before that, we once again comment on the vanishing of the first term in (12). Writing its contribution to the mean force as

$$M \, \frac{\mathrm{d}\overline{\mathbf{V}}}{\mathrm{d}t} = -\nabla \, \frac{M\overline{\mathbf{V}}^2}{2} \, ,$$

we can say that this is the force of reaction of the medium to the addition of $M\overline{V}^2/2$ to the energy of the BP and the system as a whole, introduced by the very measurement of the BP path, and therefore this force is independent of the form of the diffusion law and of concrete peculiarities of the medium in particular. There is an evident analogy to the perturbing effects of measurements in quantum mechanics. Conversely, the part of the force remaining under the large-time limit, Eqn (14), is determined solely by the shape of the probability distribution of equilibrium Brownian displacements ('diffusion law'). Consequently, this force characterizes inherent BP-medium interaction unperturbed by observations, in particular, the levels of the interaction forces and energies necessary to realize one concrete diffusion law or another. We now examine Gaussian law (7) from this standpoint and make sure that it is unrealistic.

3.3 Statistical paradox of Brownian motion

For the Gaussian diffusion law, the content of the large round brackets in 'friction equation' (14) becomes equal to unity, and the equation becomes linear:

$$\overline{\mathbf{F}} \Rightarrow -\frac{T}{D} \frac{\mathbf{R}}{2t} = -\frac{\mathbf{R}}{|\mathbf{R}|} \sqrt{z} \frac{T}{\sqrt{2Dt}}, \qquad (15)$$

with the 'friction coefficient' in front of $\mathbf{R}/(2t) = \overline{\mathbf{V}}$ connected with the diffusivity via a relation similar to the widely known Einstein relation [27, 29]. Such a similarity, however, is not an advantage of equality (15), instead being a defect.

The problem is as follows. The friction force in the original Einstein relation represents the resistance of the medium to the directed motion of a particle. When the particle is displaced by a distance \mathbf{R} , this force makes the work

$$\sim |\mathbf{R}\,\overline{\mathbf{F}}| \sim \frac{T}{D}\frac{\mathbf{R}}{t}\,\mathbf{R} \sim zT,$$

thus producing heat [we recall that $z = \mathbf{R}^2/(2Dt)$]. This quantity, like the force itself, in principle can be arbitrarily large for a suitable initial value of the particle kinetic energy.

This is clear. But it is strange that equality (15) offers the same, also unbounded, characteristic values of the force and work. Such a picture categorically contradicts the sensible meaning.

Repeating the aforesaid, here the force that features in (15) represents a medium reaction to the particle displacement along a random trajectory of thermal motion, when the initial energy value is demonstrably only $\sim T$. Moreover, the medium creates obstructions to the inertial free flight of the BP but does not prevent the particle from traveling arbitrarily far from the beginning of its path. On the contrary, the particle travels arbitrarily far just due to the 'medium's will' and at the expense of its own equilibrium fluctuations.

Therefore, in reality, in contrast to (15), the mean force in (14) as a function of the traversed path **R** cannot take arbitrarily large absolute values, instead always being bounded. This is required by such a factual inherent property of Brownian motion as translational invariance, that is, indifference of the system to irretrievable departures of the BP to anywhere. Moreover, it is reasonable to expect on these grounds that the returning force vanishes at large $|\mathbf{R}|$.

Thus, we have to conclude that the Gaussian law is inadequate to the physical nature of real Brownian motion.

The inevitability of this conclusion is obvious if we notice that if equality (15) was true, then it would mean that the medium returns the BP to the starting point of its path with a force proportional to the separation from it, $\overline{\mathbf{F}} \propto -\mathbf{R}$, i.e., like an ideal spring with the potential energy $zT/2 \propto \mathbf{R}^2$. From the physical standpoint, this looks absurd, because an arbitrary far excursion by the BP is permitted just because it does not change the thermodynamical state of the system. Our conclusion may seem paradoxical if we recall that the Gaussian diffusion law, which periodically flows from the pens of theoreticians in various physical contexts, occupies a central place in the idealized world of mathematical physics. But the paradox is resolved in a very simple way: the Gaussian statistics always appeared as a consequence of the explicit or implicit use of hypotheses (or establishing of postulates) about 'independences' of random events or quantities. We have managed without such hypotheses and thus showed their fallacy in application to Brownian motion.

We did not bind ourselves to them in the past either, and therefore arrived at the same paradoxical conclusion, in the framework of both a phenomenological statistical analysis of diffusion and transport processes [1–4, 10] and an analysis based on the full hierarchy of Bogoliubov–Born–Green– Kirkwood–Yvon (BBGKY) equations [7, 10, 16, 18, 19, 23], as well as on the basis of exact 'generalized fluctuation– dissipation relations' (FDRs) or 'dynamical virial relations' [12, 17, 18, 22], and other methods [10, 11, 15], including those for quantum systems [13, 15, 21].

In Section 4, we again touch on the 'paradox of independence', but before that, just below, we consider an example of a physically correct alternative to the Gaussian law.

3.4 Thermodynamics of Brownian motion and statistics of large deviations

From the left-hand side expression in (14), it is clear that the boundedness of the force $\overline{\mathbf{F}}$ in general implies the relation

$$\left| \overline{\mathbf{F}}(t, \mathbf{R}) \right| \leqslant \overline{F}_{\max}(t) \sim \frac{T}{\sqrt{2Dt}},$$
(16)

whose right-hand side can easily be surmised on dimensional grounds. Of course, the symbol ~ here hides some dimensionless coefficient that reflects the specificity of the concrete system and details of the construction of its function $-\ln \Psi(z)$. A comparison of (16) and (15) shows that in the region of 'tails' of the diffusion law, at $z \gtrsim 1$, the Gaussian law requires that the real force value be increased by at least $\sim \sqrt{z}$ times, thus entirely incorrectly describing (greatly underestimating) the probabilities of large displacements of a BP at $z \ge 1$.

In reality, according to (16), the function $-\ln \Psi(z)$ grows no faster than $\propto \sqrt{z}$, that is, $-\ln \Psi(z)/\sqrt{z} < \infty$, and therefore the decrease in $W(t, \mathbf{R})$ at large $|\mathbf{R}| \to \infty$ is always subexponential (anyway, not faster than a simple exponential, to say nothing about the 'Gaussian').

The difference between reality and the 'Gaussian ideal' becomes aggravated when it is not only the force itself that is bounded, but also the characteristic energy (work) conjugate to this force:

$$A(z) \equiv \frac{|\mathbf{R}||\overline{\mathbf{F}}|}{2} \leqslant A_{\max} = A(\infty) \sim T \tag{17}$$

(with the same remark about \sim). This is a natural expectation because, as a result of any walk (any path **R**), the medium takes no more energy from the **BP** than the **BP** had been able to take from the medium before.

The boundedness of A(z) implies the boundedness of the force; moreover, this implies that under an increase in $|\mathbf{R}|$, the force passes through a maximum and then decreases to zero, approximately as $|\overline{\mathbf{F}}| \approx 2A_{\text{max}}/|\mathbf{R}| \propto T/|\mathbf{R}|$. This asymptotic

behavior is again prompted by the dimensionality of quantities we put at the disposal of statistical thermodynamics.

As a result, according to equality (14), the tails of the diffusion law and hence the probabilities of large deviations $(z \ge 1)$ from typical behavior $(z \sim 1)$ decrease as $|\mathbf{R}|$ grows even much more slowly than in a mere sub-exponential fashion generally dictated by inequality (16); namely, they now decay in a power-law fashion:

$$\Psi(z) \propto z^{-A_{\max}/T}, \quad z \to \infty.$$

This can be seen after scalar multiplication of (14) by **R**, then solving the obtained differential equation, which yields

$$\Psi(z) = \Psi(0) \exp\left(-\int_0^z \frac{A(z)}{Tz} dz\right),$$

and finally applying inequality (17).

We emphasize, however, that the boundedness of the force, (16), also logically implies the vanishing of the force at infinity (excluding the extreme case where $F_{\text{max}}(t) = |\mathbf{F}(t, \infty)|$), such that the medium's 'spring' resists small 'stretching' only and always loses elasticity at large stretching.

An appropriate example of the diffusion law satisfying (17), that is, having power-law tails, is

$$\Psi(z) = \frac{(3/2+\eta)!}{(2\pi\eta)^{3/2}\eta!} \left(1 + \frac{z}{2\eta}\right)^{-5/2-\eta},$$
(18)

with a free parameter $\eta > 0$ [the factorial x! is a standard 'synonym' of the gamma function $\Gamma(x+1)$]. Obviously, $A_{\text{max}} = (5/2 + \eta) T$. The condition $\eta > 0$ is necessary for the finiteness of the mean diffusivity in (4).

Such a distribution, with $\eta = 1$, was first obtained in [16] from a consideration of Brownian motion ('self-diffusion' [7]) of a probed, or marked, atom of a gas. A similar distribution was found for molecular Brownian motion in a liquid [17, 18]), although, strictly speaking, this is an approximation of formally more exact but more complicated expressions, which, in particular, take the violation of the scale invariance into account.

Formula (18) turned out to be a reasonable approximation also for a BP whose mass M differs from the mass m of medium (gas) atoms. Here, various mathematical approaches [19, 22, 23] to the BBGKY equations lead to an identical estimate of the parameter η as a function of the mass ratio: $\eta = M/m$.

Hence, the investigation of the complete (infinite-dimensional) Liouville equation qualitatively justifies the results of our semi-heuristic analysis of the initial terms of this equation.

We may further lower formal rigor and try to interpret mathematical connections between statistics of Brownian motion and its microscopic mechanism in 'layman's terms'. For instance, let Π be internal pressure of the medium (gas) and the quantity $A_{\text{max}} = A(\infty)$ be identified with $3T/2 + \Pi\Omega$, where Ω is the gas volume displaced by a far walking BP, and $\Pi\Omega$ is the related displacement work. In essence, Ω represents the deficiency of the BP collisions with gas atoms, facilitating its nontypically far excursion. Because the position of the center of mass of the system stays fixed, an effective decrease in the gas mass near the BP, $m\Omega n$, with n being the mean concentration of gas atoms, just compensates the local mass excess M + m accompanying the current BP-atom collision. We hence have $\Omega = (M/m+1)/n$ and, assuming that $\Pi/n = T$ for a not too dense gas, $A_{\text{max}} = (5/2 + M/m) T$.

This reasoning, of course, by itself is unsafe, but can be supported by exact results on pair and many-particle nonequilibrium statistical correlations [12, 17, 18, 22], in particular, a theorem stating that the short-range character of a one-time spatial BP–atom pair correlation (finiteness of the 'correlation volume' Ω) implies a long-range ('long-lived') behavior of many-time self-correlations in the BP motion and, hence, invalidity of the Gaussian diffusion law (reciprocally, the validity of the latter requires nonlocality of the BP–gas correlations in space) [17, 18].

3.5 Uncertainty and flicker fluctuations of the diffusion rate Decomposing non-Gaussian diffusion law (18) with respect to Gaussian ones in accordance with (8), we obtain the probability distribution of the random quantity $\zeta = \Delta/D$ in the form

$$U(\zeta, 0) = \frac{1}{\eta! \zeta} \left(\frac{\eta}{\zeta}\right)^{\eta+1} \exp\left(-\frac{\eta}{\zeta}\right).$$
(19)

According to the FDR [1, 4, 12], this distribution is transferred to the BP mobility (at least the 'low-field' one) and therefore can be observed in measurements of diversity of the 'time-of-flight' (time of drift) values for BPs under the influence of an external force (for example, electrons or holes injected in semiconductors) [20].

Effects of non-Gaussian statistics were also observed directly in equilibrium, by measuring 'fourth cumulants' (irreducible fourth-order correlations) of electric current or voltage noise [1]. Here, low-frequency fluctuations of power spectral density of thermal electric noise (at frequencies $f \sim 1/t$) were under investigation, which, in essence, is the uncertainty and fluctuations of the charge transfer rate and the rates (coefficients) of diffusion of charge carriers.

In our example (18), (19), assuming that $\eta > 1$, it is not difficult to calculate

$$\frac{\langle (\mathbf{R}^2)^2 \rangle}{\langle \mathbf{R}^2 \rangle^2} - 3 = 3\left(\frac{\langle \Delta^2 \rangle}{\langle \Delta \rangle^2} - 1\right) = \frac{3}{\eta - 1}$$

with $\langle \Delta \rangle = D$ and $\langle \mathbf{R}^2 \rangle = 6Dt$. This formula shows not only a degree of uncertainty of the diffusion rate but also defects in the approximation of pure scale invariance: a divergence of the variance of the diffusion rate for $\eta \leq 1$ and the complete independence of the variance from the duration of observations. The last circumstance means that the rate fluctuations are similar to quasistatic ones, with the spectrum (spectral power density) $S_D(f) \propto D^2 \delta(f)$ concentrated at the zero frequency. This is the usual result of the simplest (although nontrivial) approach to 1/f noise from microscopic theory [13–15].

Undoubtedly, in a more precise theory beyond ideal scale invariance [16, 19], the tails of the diffusion law are cut off at least at $\mathbf{R}^2 \gtrsim V_0^2 t^2$ [$\Delta \gtrsim D/\xi \sim V_0^2 t$ in (8)], such that the variance of Δ , as well as all higher-order statistical moments of **R** and Δ , are definitely finite and hardly exceed values corresponding to a free BP flight,

$$\langle (\mathbf{R}^2)^k \rangle \lesssim (2k+1)!! (V_0^2 t^2)^k, \quad \langle \Delta^k \rangle \lesssim (2k+1)!! \left(\frac{V_0^2 t}{2}\right)^k.$$

Simultaneously, the delta-function $\delta(f)$ does somehow 'spread', keeping the dimensionality and singularity at zero

and taking the form $\sim 1/f$, where \sim includes a function of $\ln(\tau f)$.

The first of these corrections can be easily described by replacing $U(\zeta, 0)$, for instance, in (19), with the approximate expression $U(\zeta, \xi) \approx U(\zeta, 0) \Xi(\zeta\xi)$, in which $\Xi(0) = 1$ and $\Xi(\cdot)$ sufficiently rapidly tends to zero at infinity. Then, instead of (18), we obtain

$$\Psi(z,\xi) \approx \Psi(z) \Theta(z\xi)$$

where the scale-invariant factor $\Psi(z)$ is the same as before, for instance, in (18), and also $\Theta(0) = 1$ and $\Theta(\cdot)$ rapidly tends to zero at infinity, thus cutting off the tail of $\Psi(z)$. As a result, the quadratic cumulant of Δ becomes finite even at $\eta \leq 1$. At once, it acquires time dependence, such that the fourth cumulant of the BP displacement increases with time, $\propto t^{3-\eta}$ if $\eta < 1$, and $\propto t^2 \ln(t/\tau)$ at $\eta = 1$. Correspondingly, the quasistatic spectrum $\propto \delta(f)$ transforms into the flicker spectrum

$$S_D(f) \sim \frac{D^2}{\pi f} \left(\frac{1}{\tau f}\right)^{1-\eta},$$
 (20)

at $\tau f \ll 1$, and, in particular, into the 1/f spectrum at $\eta = 1$.

However, at $\eta > 1$, such a correction is insufficient for a full 'spreading' of the frequency delta function — which means that the scale invariance violation has now a more complex or different character. We can obtain a notion of how else it may look, for example, if we consider [1–4, 10, 34] a diffusion law that has infinite divisibility and stability properties in the sense of probability theory [32] but only asymptotically as $\xi = \tau/t \rightarrow 0$, because no real transport process can be physically divided into infinitely small independent fragments. The corresponding kernel in expansion (8) is simplistically describable by the formula

$$U(\zeta,\xi) \approx \frac{\alpha(\xi) \exp_{+} \left[-\left(\zeta - \zeta_{0}(\xi)\right)/c \right]}{\left[\zeta - \zeta_{0}(\xi) + \alpha(\xi)\right]^{2}},$$
(21)

where

$$\begin{aligned} &\alpha(\xi) = \frac{1}{\ln(1/\xi)} = \frac{1}{\ln(t/\tau)} \,, \\ &\exp_{+}(x) = \begin{cases} \exp(x) \,, & x > 0 \,, \\ 0 \,, & x < 0 \,, \end{cases} \end{aligned}$$

the function $\zeta_0(\xi)$ is determined by condition (4), that is, $\int \zeta U(\zeta, \xi) d\zeta = 1$, and $c = r_0^2/D\tau_0$, with r_0 and τ_0 being the minimal space scale and the time scale down to which the 'infinite divisibility' of a random walk is physically meaningful [for simplicity, it is presumed in (21) that the constant *c* is not too small, $c \ge \alpha(\xi)$]. Obviously, as $\xi \to 0$, expression (21) becomes $U(\zeta, 0) = \delta(\zeta - 1)$. Thus, the scale-invariant 'seed' of such a diffusion law is purely Gaussian, which motivates us to name it quasi-Gaussian [10]. In [34], it was considered in detail along with its generalizations and a comparison with experimental data [20].

For tails of the quasi-Gaussian law at $z \ge 1$, Eqns (8) and (21) yield

$$\frac{\Psi(z,\xi)}{\Psi(0,\xi)} \sim \alpha(\xi) \; \frac{2c\sqrt{\pi}}{z^{3/2}} \exp\left(-\sqrt{\frac{2z}{c}}\right),$$

and hence the tails satisfy boundedness requirement (16), although they lie on the boundary of the set of diffusion laws permitted by inequality (16). As regards the spectrum of flicker fluctuations of the diffusion rate (or, generally, the transport process rate), we find

$$S_D(f) \approx \frac{D^2 c}{f} \left(\ln \frac{1}{\tau f} \right)^{\gamma},$$
 (22)

where $\gamma = -2$.

Spectrum (22) reflects the difference between the most probable value and the (ensemble) average of the transport rate, $\zeta_0(\xi)$ and 1 in relative units in (21); the difference decreases with the observation time, albeit logarithmically slowly: $1 - \zeta_0(\xi) \approx \alpha(\xi) \ln (c/\alpha(\xi))$. Kernel (19) is similar in form to (21) (a more or less sharp 'wall' on the left and comparatively gentle slope on the right), but the analogous difference in (19) is fixed. Imparting a time dependence to it may be one more, parallel, scenario of the spectrum spreading of $\delta(f)$ under improved analytic approximations of solutions of the BBGKY equations. From our standpoint, this is a relevant question in statistical mechanics.

However, even the already available approximations of microscopic theory are able to produce realistic quantitative estimates of the 1/f noise amplitude. Although the estimates obtained in [7] and in [16, 20] in different approximations [(22) with $\gamma = 1$ and (20) with $\eta = 1$ or (22) with $\gamma = 0$] differ from one another by the factor $\ln \left[\frac{1}{\tau f} \right]$, they are both in reasonable agreement with experimental data on liquids and gases [1, 20], taking their diversity into account. On the other hand, the quasi-Gaussian random walk scheme rather well predicts or explains the magnitudes of electric 1/f noise in various systems [1–3, 10, 20]. Because we here deal with the transfer of charge instead of mass (in view of the smallness of the mass of usual charge carriers), while the interaction of walking charges with the medium is long-range, it is not surprising that the transport statistics are non-Gaussian in an essentially other manner than in the case of molecular Brownian motion. Analysis of the relation of these statistics to a quantum many-particle Liouville equation or equivalent 'quantum BBGKY hierarchy', for example, for standard electron-phonon Hamiltonians, also seems to be a relevant issue.

At the current stage of development of statistical mechanics, it is useful to state that unprejudiced treatment of this science inevitably discovers flicker fluctuations of rates of transport processes, even the rate of the random walk of a particle interacting with an ideal gas [17, 18, 22, 23] and, moreover, even in the formal Boltzmann–Grad limit (the infinitesimal gas parameter) [35].

This fact excellently highlights the inconsistency of attempts to reduce 1/f noise and related long-lived statistical correlations and dependences to some very long memory or relaxation times, and thus the inconsistency of the opinion nourishing such attempts by presuming any statistical correlations between random phenomena to be evidence of literal or at least indirect physical connections between them.

In Section 4, we show with the help of elementary logic that in reality, just the physical disconnectedness of interparticle collisions leads to the uncertainty and 1/f noise of the relative frequency of collisions and rate of wandering of each of the particles in many-particle systems. Thus, from a new standpoint, we justify both the general logic of the introduction and elementary mathematical treatment in Sections 2 and 3 of molecular random walk.

4. Myths and reality of molecular random walks

4.1 Gaussian probability law

and two meanings of the independence of random events

We first recall why the Gaussian law appeared and continues to appear in various theoretical models. One reason is that it naturally comes from the assumption of the statistical independence of BP displacements (random walk increments) at nonoverlapping time intervals. But the main reason is that physicists have become accustomed to identifying the statistical independence of random events in the sense of probability theory with their independence in the worldly sense of their lack of influence on one another.

Both these circumstances have more than a 300-year history. The history of the Gaussian law began from the celebrated 'law of large numbers' by J Bernoulli [36], who investigated the statistics of sequences of observations on the vicissitudes of life or, for instance, playing dice, under the assumption that unpredictable outcomes of successive 'random trials' are mutually independent or, to be more precise, that their probabilities are independent, that is, the joint probability of several random events decomposes (factores) into the product of their individual probabilities.

In exactly the same way, statistical independence is introduced in modern probability theory [31]. But in the probability theory, it is nothing but a formal mathematical definition and therefore, as Kolmogorov warned in [31], deducing this property of probabilities from the independence of physical phenomena as such is possible as a hypothesis only, which should be verified in experiment.

In other words, any evidence of the independence of physical random events in each of their concrete realizations (e.g., in the sense of the absence of cause-and-effect connections between them) by itself is not sufficient for declaring the statistical independence of these events in a set (statistical ensemble) of realizations.

Logically inverting this thesis, we conclude that if statistical experiments reveal a statistical dependence in an ensemble of realizations of random events, this does not necessarily mean that some real interaction between the events exists. That is precisely the case where one encounters 1/f noise.

Hence, identifying the two senses of 'independence' is nothing but a fallacy, which, unfortunately, traditionally governs physicists' relation to randomness, even despite its careful disclosure — in the context of fundamental statistical mechanics — made by Krylov more than 60 years ago [37].

4.2 Collisions, chaos, and noise in a system of hard balls

Mathematicians know Krylov as one of the founders of the modern theory of dynamical chaos. According to this theory [38, 39], notably, the motion of $N \ge 3$ elastic hard balls in a box or disks on a torus obeying deterministic laws of mechanics is indistinguishable from a random process [39, 40]. For us, it is important that statistical characteristics of this process are crucially sensitive to the ratio of the duration of observing it and the total number of balls participating in it.

More precisely, we consider the role of the parameter $t/\tau N$, where τ is the mean time separation of collisions of a

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given ball with others and therefore the relaxation time of the ball velocity due to collisions with other balls [16, 18]. For

$$\frac{t}{\tau} \gg N$$

clearly, the number $\propto t/\tau$ of quantities describing the trajectory of any of the balls is much greater than the number $\propto N$ of quantities establishing the initial state of the whole system, and hence each particular ball trajectory contains the same exhaustive information on the system. Moreover, this information is already contained in any small part of a particular trajectory with duration $\sim N\tau \ll t$.

Due to this circumstance, fluctuations in the numbers of collisions of a given ball, from one time subinterval with a duration $\sim N\tau$ to the next such subinterval, behave like statistically independent random quantities, or white noise (which is quite understandable: the presence of some relation or correlation between them would reveal some special features, not yet realized, of the system initial state, which would contradict the condition $t \ge N\tau$). Accordingly, the relative frequency of a particular ball collisions averaged over the whole observation time is almost nonrandom, i.e., is the same for all balls and all initial conditions (at a fixed total energy of the system, of course), while the statistics of fluctuations in the number of collisions (of a given ball) at intervals $t \ge N\tau$ obey the law of large numbers, i.e., are asymptotically Gaussian.

4.3 Paradox of independence

Such a picture of the chaos of collisions — as with usual noise — is quite attractive for physicists. But we should not forget that it required the condition $t/\tau \ge N$ establishing a rigid (deterministic) physical (cause-and-effect) inter-dependence between collisions, which is nonlocal in time and space (nonvanishing as $t/\tau \rightarrow \infty$ and pertaining to all the balls). Just this dependence, very paradoxically, provides the statistical independence of events (collisions) that are distant from one another in time or in space.

We see that maintaining the ideal disorder with which statistical independence is usually associated requires vigilant underlying control of it and, in this sense, a global order.

At this point, it involuntarily comes to mind how the Dodo in Lewis Carroll's *Alice's Adventures in Wonderland* (as translated into Russian by B Zakhoder) agitated other characters to 'fit into strict disorder', or 'stand up strictly anyhow'. In addition, analogies to the mysterious 'quantum nonlocality' and 'entangled quantum states' suggest themselves.

4.4 Uncertainty and 1/f noise of the relative frequency of collisions and rate of diffusion

However, in real world, it is not easy to measure the temporal disorder of random events so strictly as to subordinate it to the law of large numbers. This is not easy for the simple reason that real many-particle systems are characterized by the opposite ratio of the duration of observations achievable in experiments and the number of particles in the system:

 $\frac{t}{\tau} \ll N \,.$

Therefore, the appeal to arbitrary large averaging times, so much beloved in mathematical physics, has no factual grounds [40].

The above inequality is satisfied even for very small volumes of fluids isolated from the rest of the world [18]. This inequality is all the more true in view of the impossibility of complete isolation and hence the necessity of adding to N the number of particles (and generally degrees of freedom) of all huge surroundings of the system. And this inequality definitely covers objects of the Gibbs statistical mechanics, in which the number of particles N is not bounded. Exactly this was under Krylov's critical analysis [37].

Now, the number $\propto t/\tau$ of quantities sufficient for a description of the observed trajectory of one particle (ball) or another all the time stays small compared with the number $\propto N$ of independent causes (parameters of the system initial state) influencing the trajectory.

But averaging over a relatively few number of consequences of a larger number of causes cannot produce a certain result, because the result remains dependent on many unknown free parameters and does not represent all possible variants of the course of events, and all the more cannot represent them in some definite proportion. Therefore, time averaging over time intervals that are available for observing the life of a particle in any particular experiment (at each part or realization of a phase trajectory of the system) inevitably gives an unpredictable new value of the relative frequency of particle collisions and, moreover, a new distribution (histogram) of collisions (or more complex events) with respect to their internal characteristics. In other words, an experimenter encounters 1/f noise (see the Introduction).

It hence follows that instead of thinking up hypotheses on relative frequencies or 'probabilities' and the 'independence' of events constituting random walks, it would be better to imitate Newton [8] and devote ourselves to investigating equations of statistical mechanics.

4.5 Independence game and problems of statistical mechanics

To investigate the equations of statistical mechanics rather than producing hypotheses was Krylov's appeal in his book [37] when explaining the falseness of widespread prejudices "as if a probability law exists regardless of theoretical scheme and full experiment" and "as if 'obviously independent' phenomena should have independent probability distributions."[†]

The full experiment here means a concrete realization of a phase trajectory of a system considered as a single whole — as an image of a practical experiment-without its artificial division into 'independent' time fragments (in Sections 2 and 3, we considered exactly a full experiment). At $N \gg t/\tau$, the time-smoothed relative frequency of a given sort of random event (collisions of a given particle with others) varies from one experiment to another, and therefore we cannot introduce a definite a priori 'probability' for separate events. This means that they are all mutually statistically dependent because, figuratively speaking, all are equally responsible for the resulting (and each time new) rate of their appearance (a posteriori probability). This is so although all the events are physically independent, because at $N \gg t/\tau$ they arise from interactions with different groups of a total set of N particles. Correspondingly, Bernoulli's law of large numbers breaks down, because it is based on the postulate of statistical independence.

† Author's translation of the Russian original.

Here, we clearly see another side of the 'paradox of independence': genuine full-fledged chaos involves infinitely long statistical dependences and correlations.

It is also clear why molecular Brownian motion, being associated with such full-fledged chaos, does not want to fit the Procrustean bed of Gaussian statistics and, all the more, Boltzmann's kinetics.

5. Conclusion

Unfortunately, the above-mentioned popular careless ideas of independences and probabilities of random phenomena "are so much habitual that even a person who agreed with our argumentation then usually automatically returns to them as soon as he faces a new question. The origin of the stability of these ideas lies in the fact that they are based on a common intuitive notion about statistical laws, and therefore they would be permissible and advisable if the issue was learning phenomena of empirical reality. However, such ideas turn out to be quite unsatisfactory as a benchmark for substantiation of probability laws when the question is the connections between statistical laws and the principles of micro-mechanics" [37].

Fortunately, we currently have an understanding of the errors of replacing micro-mechanics by speculative probabilistic constructions, even if these are beautiful in and of themselves. Besides, as we noted above, there is already an experience of systematic investigation of equations of statistical mechanics in application to transport processes. It clearly shows that the mechanics of systems of very many interacting particles, or degrees of freedom, in no way prescribes interactions to keep the certainty of the rates of changing the system micro-states (transition probabilities), even when molecular chaos takes the form of macroscopic order.

The point is that any realization of an 'elementary' act of interactions is in fact a product of the full (initial) micro-state of the system, and therefore the number of causes of visible randomness always greatly exceeds the number of its manifestations under time averaging over factually achievable durations of experiments. As a consequence, any particular experiment presents to the researcher its own unique assortment of relative frequencies ('probabilities') of random events composing a process under observation. That is 1/f noise.

Hence, being surprised by 1/f noise is no more grounded than being surprised by noise in general. Nature needs 1/f noise as a manifestation in any particular 'irreversible' process of all inexhaustible resources of nature's randomness, and on the whole the uniqueness of the observed realization of our Universe's evolution at all of its time scales. A purely stochastic world, without 1/f noise, where anything can be easily time-averaged, would be too boring (and even, possibly, would suppress free will [41]).

Unfortunately, as we have seen above, 1/f noise has 'bad' statistics absolutely alien to the law of large numbers and resembling those that sometimes force their observers (see, e.g., [42]) to suspect the action of mysterious 'cosmophysical factors'. This circumstance strongly complicates theoretical tasks.

Fortunately, although influence from the cosmos is never undoubtedly ruled out, a source of randomness quite sufficient for 1/f noise creation is already hidden, as we noted above, in such a simple system as a molecular Brownian particle interacting with an ideal gas. As we have demonstrated, such a source definitely exists in any medium that allows Brownian motion. Hence, we have every prospect of success in building and experimentally verifying the theory of 1/f noise (and accompanying statistical anomalies) starting from quite usual Hamiltonians.

We hope that this article will induce some interested reader to work in this intriguing area of statistical physics.

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