Anomalous Dynamical Scaling Determines Universal Critical Singularities

Attilio L. Stella
Department of Physics and Astronomy, University of Padova, Via Marzolo 8, I-35131 Padova, Italy and INFN, Sezione di Padova, Via Marzolo 8, I-35131 Padova, Italy

Aleksei Chechkin
Institute of Physics and Astronomy, University of Potsdam, D-14476 Potsdam-Golm, Germany, Faculty of Pure and Applied Mathematics, Hugo Steinhaus Center, University of Science and Technology, Wyspianskiego 27, 50-370 Wrocław, Poland, and Akhiezer Institute for Theoretical Physics National Science Center "Kharkov Institute of Physics and Technology," 61108, Kharkiv, Ukraine

Gianluca Teza*
Department of Physics of Complex Systems, Weizmann Institute of Science, Rehovot 7610001, Israel

(Received 6 October 2022; accepted 19 April 2023; published 18 May 2023)

Anomalous diffusion phenomena occur on length scales spanning from intracellular to astrophysical ranges. A specific form of decay at a large argument of the probability density function of rescaled displacement (scaling function) is derived and shown to imply universal singularities in the normalized cumulant generator. Exact calculations for continuous time random walks provide paradigmatic examples connected with singularities of second order phase transitions. In the biased case scaling is restricted to displacements in the drift direction and singularities have no equilibrium analogue.

DOI: 10.1103/PhysRevLett.130.207104

Scaling laws are at the basis of our understanding of equilibrium systems at criticality and of the singularities associated with second order phase transitions [1–3]. A key role in this context is played by the scaling of probability density functions (PDFs) of observables [4], like the spatial span of a self repelling polymer at varying backbone lengths [5–7] or the magnetization of a finite Ising model for different system sizes [8,9].

PDFs with an analogous type of scaling, but with time t replacing the chain length or system size, are also often met outside of equilibrium. A paradigmatic example is that of anomalous spatial diffusion, where the mean squared displacement grows as $\langle x^2\rangle \sim t^{2\nu}$ with $\nu \neq 1/2$ ($\nu = 1/2$ provides Brownian diffusion [10]). At long times, the associated PDF asymptotically satisfies

$$p(x,t) \sim t^{-\nu} f(x/t^{\nu}) \tag{1}$$

where f is a non-Gaussian scaling function [11,12]. The importance of this characterization follows from the ubiquity of anomalous diffusion in nature, which can be observed in a variety of experiments carried out on different scales ranging from astrophysical to intracellular ones [10,11,13–26]. The scaling function f is expected to decay exponentially fast at large values with a power of the argument linked to ν by a relation first established by Fisher for polymers in equilibrium [6], and supported by probabilistic arguments and numerical model calculations [11,12,27], simulations [28], and renormalization group results [29].

Besides the polymer case, stretched exponential decays of scaling functions have been conjectured or numerically estimated also for equilibrium criticality, especially for the PDF of the magnetization of finite Ising systems at the Curie temperature and in zero magnetic field [8,9,30–32]. On the basis of these decays, analogies between magnetic critical phenomena and anomalous diffusion were already stressed in early work [11]. In the magnetic Ising case, the power quantifying the stretching is expected to be directly connected to the Kadanoff exponent determining the magnetic field singularity of the free energy density [2,33]. In the attempt to deepen its connection with singularities and other universal aspects of equilibrium criticality, this type of stretched exponential decay, but also modulated by a power law factor, was conjectured in Refs. [34,35]. However, such conjecture could never be proven or fully confirmed numerically [36–40].

Progress in the understanding of nonequilibrium dynamics largely relies on parallels one can draw with equilibrium [41–43]. Thus, it is fundamental to investigate the possible connections established by the scaling function decays encountered in anomalous diffusion with the singularities observed in equilibrium systems at criticality. A characterization in this perspective of scaling and its consequences for anomalous diffusion is also of interest for general nonequilibrium theory.

In this Letter, we show that the scaling property of a diffusing system implies a specific form of decay of the scaling function, which determines power-law singularities in the scaled cumulant generating function of displacement. Remarkably, we show that such singularities originate from the same form of decay of the scaling function once postulated for the magnetization of Ising systems, including the modulating power law factor [34,35]. The singularities propagate to the large deviation functions [41,42] and cause divergences of a dynamical response function analogous to a magnetic susceptibility in equilibrium. The Fisher relation is shown to follow from the property of extensivity in time of the generator of cumulants. The universality of this relation for equilibrium and nonequilibrium problems emerges, in our approach, from its clear connection to large deviation properties. We also address the case of biased diffusion, obtaining singularities corresponding to scaling forms that have no analogue in equilibrium systems. All this is verified by exact calculations for continuous time random walks (CTRWs) and related fractional drift-diffusion equations as generic models for anomalous diffusion [10,13,14]. In this way we establish bridges among three pillars of statistical mechanics: scaling, anomalous diffusion, and large deviation theory.

We start by considering the general case of a particle diffusing on a one-dimensional landscape, assuming that the scaling hypothesis of Eq. (1) holds for some exponent $0 < \nu < 1$ [44]. The generating function is expressed in terms of a Laplace transform as $G(\lambda, t) = \int_{-\infty}^{+\infty} dx e^{\lambda x} p(x, t)$. For long enough times, on the basis of Eq. (1), it takes the form

$$G(\lambda, t) \sim \int_{-\infty}^{+\infty} dz e^{\lambda t^\nu z} f(z) \tag{2}$$

where we performed the variable change $z = x/t^\nu$. The cumulants are generated by differentiation with respect to λ at $\lambda = 0$ of the cumulant generator $\log G$, which here we assume to be linearly extensive in time. Hence, the scaled cumulant generating function (SCGF) can be expressed through the limit

$$\varepsilon(\lambda) = \lim_{t\to\infty} \frac{1}{t} \log G(\lambda, t). \tag{3}$$

It is already apparent how the existence of this finite limit cannot exclude the possibility of a singularity of $\varepsilon(\lambda)$ at $\lambda = 0$. Indeed from Eq. (1) it follows that, for a non-Gaussian scaling function, the nth order cumulant grows as $t^{n\nu}$, implying a divergence to infinity for the cumulant scaled by t for $n > 1/\nu$ when $t \to \infty$. Consistently, this can cause a divergence of the nth derivative of the SCGF as soon as $n > 1/\nu$.

In the case of free diffusion the scaling function $f(z)$ is symmetric like that of the magnetization of an Ising system at criticality. To the contrary, for biased diffusion we could expect an asymmetry [46] or even a restriction of the domain in which Eq. (1) holds. For both free and biased diffusion, the dominant behavior of the integral in Eq. (2) for large λt^ν with $\lambda > 0$ is determined by the decay of the scaling function $f(z)$ for $z \to +\infty$ (throughout this Letter when considering a bias we assume it to be in the positive direction). Indeed, with f differentiable and monotonically decreasing sufficiently fast to zero as $z \to +\infty$, the integrand in Eq. (2) reaches a maximum at some $\bar{z}$ which increases towards $+\infty$ as time grows. Applying Laplace's method [47] the leading contribution to G will come from the integrand in Eq. (2) computed at the value $\bar{z}$ satisfying $f'(\bar{z})/f(\bar{z}) = -\lambda t^\nu$, which maximizes the argument of the exponential in terms of which one can write the integrand in Eq. (2). Assuming that $\bar{z}$ for long enough times grows as $(\lambda t^\nu)^{1/\delta}$ for some $\delta > 0$, we get that the differential equation $f'(\bar{z})/f(\bar{z}) \sim -\bar{z}^\delta$ asymptotically holds. It admits the solution

$$f(\bar{z}) \sim \bar{z}^\psi e^{-c\bar{z}^{\delta+1}} \tag{4}$$

for some positive constant c and any exponent ψ [48]. The factor $\bar{z}^\psi$ is introduced to allow the possibility of cancellation of a term $\propto \log(\lambda t^\nu)$ in $\log G$, as shown below, and implies a correction $\propto \bar{z}^{-1}$ to the differential equation.

The exponent δ not only enters the tails of the scaling function, but also determines the asymptotic dominant term in ε. Indeed, substituting $\bar{z}$ in Eq. (2) and using Laplace's method we get

$$\log G(\lambda, t) \sim \lambda t^\nu \bar{z} - c\bar{z}^{\delta+1} + \frac{2\psi + 1 - \delta}{2} \log \bar{z} + \text{const} \tag{5}$$

up to a correction $\propto \bar{z}^{-1-\delta}$ [49]. Recalling that $\bar{z}^\delta \sim \lambda t^\nu$, we find that the first two terms are proportional to the same powers of t and of λ, the third term is a logarithmic correction [disappearing only for $\psi = (\delta - 1)/2$] and the fourth is a time independent constant [49]. The term $\propto \log(\bar{z})$ is the only one which actually allows us to split the λ and t dependencies into the sum of two separate terms. Therefore its presence would introduce a logarithmic singular dependence on λ in the whole t-independent part of $\log G$, implying a divergence for $\lambda = 0$. For such reason, this dependence should be dropped by the above choice of ψ.

A contribution, extensive in t for $\log G$, can result from the first two terms. Since the exponent of t depends on both ν and δ, Eq. (3) provides $\delta = \nu/(1 - \nu)$, known as the Fisher relation [6]. We also find that Eq. (5) predicts a singular dependence for $\varepsilon(\lambda) \sim \lambda^{1/\nu}$ at $\lambda = 0^+$. Therefore, the larger is δ, i.e., the faster the decay of f, the larger also ν and thus the stronger the singularity in λ. The fact that it is determined by the asymptotic rate of decay of f confers a universal character to the singularity: different f's can have the same law of decay and thus cause the same singularity.

Below we demonstrate the existence of a critical singularity in the SCGF of the CTRW in both free (subdiffusive) and biased (subdiffusive and superdiffusive) regimes [49–52]. Experimental evidence of systems described by this model is found in many different contexts, such as charge-carrier transport in amorphous semiconductors [53], dynamical chaos [54], transport in a groundwater aquifer [55,56], cell biology [21,57–60], and finance [61–63], to name a few. In this model, a particle jumps on a one-dimensional lattice with spacing L with a rate r (l) to the right (left) nearest neighboring site. The waiting times τ in between the jumps occur according to a certain PDF $\omega(\tau)$. Anomalous diffusion occurs when this PDF decays to zero as a power law [$\omega(\tau) \sim \tau^{-1-\alpha}$ with $0 < \alpha < 1$] for $\tau \to \infty$, so that its first moment is infinite. We underline that the freedom in the choice of α, together with the universal character of the singularities, testifies the applicability of our approach to a wide class of nonequilibrium scenarios.

The probability $P_i(t)$ to observe the particle on the ith site at a certain time t evolves according to the generalized master equation

$$\partial_t^\alpha P_i(t) = rP_{i-1}(t) + lP_{i+1}(t) - (r+l)P_i(t) \tag{6}$$

where ∂_t^α is the α-order Caputo fractional derivative (implying that the unit of r and l is $[\text{time}]^{-\alpha}$) which has the integral representation [49,64]

$$\partial_t^\alpha P_i(t) = \frac{1}{\Gamma(1-\alpha)} \int_0^t \frac{\partial_\tau P_i(\tau)}{(t-\tau)^\alpha} d\tau \tag{7}$$

where $\Gamma(\cdots)$ is the complete Gamma function. With initial condition $P_i(0) = \delta_{i,0}$, the generating function $G(\lambda, t) = \Sigma_i e^{\lambda L i} P_i(t)$ satisfies

$$[\partial_t^\alpha - \varepsilon_B(\lambda)]G(\lambda, t) = 0 \tag{8}$$

where $\varepsilon_B(\lambda) = r(e^{L\lambda} - 1) + l(e^{-L\lambda} - 1)$ is the SCGF of Brownian ($\alpha = 1$) diffusion [65,66]. The solution can be found, passing to Laplace space [49], to be

$$G(\lambda, t) = E_\alpha(\varepsilon_B(\lambda)t^\alpha) \tag{9}$$

where E_α is the one-parameter Mittag-Leffler function [67]. The asymptotics of this function are proportional to $e^{\varepsilon_B(\lambda)^{1/\alpha}t}$ and $-1/\varepsilon_B(\lambda)t^\alpha$ for positive and negative arguments, respectively.

Let us first address the unbiased case, with $r = l$. We find that $\varepsilon_B(\lambda) = (4r)\sinh^2(L\lambda/2) \geq 0$. Therefore, taking the long time limit provides us with the following SCGF:

$$\varepsilon(\lambda) = (4r)^{1/\alpha} \sinh^{2/\alpha}(L\lambda/2) \sim \lambda^{2/\alpha} + O(\lambda^{2+2/\alpha}). \tag{10}$$

Thus we get a leading singularity $\sim\lambda^{2/\alpha}$ in the scaled cumulant generating function at $\lambda = 0$. This singularity is qualitatively of the same type encountered for the free energy density of Ising systems at criticality, with λ here playing the role of magnetic field there [30]. The first derivative of $\varepsilon(\lambda)$ divergent at $\lambda = 0$ is that of even order n with n just exceeding $2/\alpha$. This derivative assumes the meaning of a diverging dynamical response function related to counting and analogous to the magnetic susceptibility of an Ising model at criticality. The divergence $\sim|\lambda|^{(2/\alpha)-n}$ of this derivative heralds the fact that for large t the nth cumulant of the total displacement grows as $t^{n\alpha/2}$.

The correctness of our general argument connecting the singularity of the SCGF to a specific form of asymptotic decay of the scaling function can be exactly verified in this example. Indeed, in the case $r = l$ the continuum limit of Eq. (6) yields the fractional diffusion equation [14,49] regulating the PDF $P_i(t)/L \to p(x, t)$ of observing the particle at position $iL \to x$ at time t

$$\partial_t^\alpha p(x, t) = D\partial_x^2 p(x, t), \tag{11}$$

where $rL^2 \to D$ is a diffusion constant, which to the purpose of our further discussion can be assumed to be equal to 1. The solution to this equation satisfies exactly the scaling form of Eq. (1) for all x and t with $\nu = \alpha/2$. The scaling function can be expressed via the Fox H function [68], M-Wright function [69], or the one-sided Lévy stable density [70]. Choosing the M-Wright function, we can express the generating function as in Eq. (2) with $M_{\alpha/2}(z)$ replacing $f(z)$. The tails of this scaling function are given by [69,71]

$$M_\nu(z) \sim |z|^{\frac{\nu-1/2}{1-\nu}} e^{-\frac{1-\nu}{\nu}|\nu z|^{1/(1-\nu)}} \tag{12}$$

and have precisely the general form argued in Eq. (4). The exponent δ is found to take the value $\nu/(1-\nu)$, consistent with the Fisher relation and implying that $\log G$ grows linearly with time [from Eq. (5)]. The multiplicative power factor with $\psi = (\nu - 1/2)/(1-\nu)$, which translates in $\psi = (\delta - 1)/2$, allows us to drop the possible logarithmic dependence on $\bar{z}$ for $\log G(\lambda, t)$, and the leading singularity is $\varepsilon(\lambda) \sim \lambda^{2/\alpha}$, confirming the result in Eq. (10).

Let us now address the biased CTRW case, given by Eq. (6) when $r \neq l$. In such a scenario, the factor $\varepsilon_B(\lambda)$ determining the lattice generating function $G(\lambda, t)$ has an additional zero $\lambda_0 = L^{-1}\log l/r < 0$ for $r > l$, as shown by the yellow curve in Fig. 1(a). This implies for $G(\lambda, t)$ a power-law asymptotic behavior $\propto -1/\varepsilon_B(\lambda)t^\alpha$ in the infinite time limit for $\lambda_0 < \lambda < 0$, while an exponential dependence $\propto e^{\varepsilon_B(\lambda)^{1/\alpha}t}$ holds elsewhere. This causes the associated SCGF to be identically zero in that interval, giving us

$$\varepsilon(\lambda) = \begin{cases} \varepsilon_B(\lambda)^{1/\alpha} & \lambda \leq \lambda_0 \text{ and } \lambda \geq 0 \\ 0 & \lambda_0 < \lambda < 0 \end{cases} \tag{13}$$

which shows a power law singularity $\sim\lambda^{1/\alpha}$ for $\lambda \to 0^+$ [see Fig. 1(a)]. An additional singularity $\sim(\lambda_0 - \lambda)^{1/\alpha}$ appears for $\lambda \to \lambda_0^-$. The simultaneous presence of these singularities is consistent with the fact that the SCGF satisfies the Gallavotti Cohen identity, $\varepsilon(\lambda) = \varepsilon(-\lambda + \lambda_0)$, making the function symmetric with respect to the $\lambda_0/2$ axis and heralding validity of the fluctuation theorem. Both these

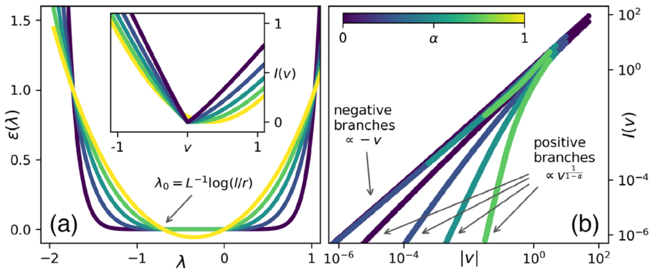

FIG. 1. (a) SCGF for a biased ($r = 2/3$) subdiffusive random walk [Eq. (13)] for different values of α: $\varepsilon(\lambda)$ identically zero for $\lambda_0 \leq \lambda \leq 0$. The nonsingular case of Brownian diffusion (ε_B) is also reported for reference (yellow). (b) The branches of the rate functions exhibit a dependence $\sim v^{1/(1-\alpha)}$ for positive values, while $\sim -v$ for negative values, which ensures validity of the fluctuation theorem.

singularities are totally asymmetric, since $\varepsilon(\lambda)$ is identically zero for $\lambda_0 < \lambda < 0$. This asymmetry and the simultaneous existence of two singularities have no counterpart in equilibrium systems at criticality. Nevertheless, the singularities can still be justified along the lines proposed here, but in terms of a scaling which now holds only in the positive x domain.

The continuum limit of Eq. (6) for $r > l$ leads to the fractional drift diffusion equation [49]

$$\partial_t^\alpha p(x,t) = [-K\partial_x + D\partial_x^2]p(x,t), \qquad (14)$$

where $(r-l)L \to K$ defines the drift constant and $(r+l)L^2/2 \to D$ the diffusion constant, implying that Eq. (11) is immediately recovered in the case $r = l$. As in the free case, these limit prescriptions are justified by their consistency with the scaling of the solution of the resulting continuum equation. Local detailed balance [66,72,73] allows us to link the coefficients to the rates of the CTRW [49,74]. Since also in this case the actual values of the constants do not affect our results, for simplicity we set $K = 1$ and $D = 1$ below. An asymptotic ($t \to \infty$) solution of Eq. (14) for the positive branch ($x > 0$) is found to be $t^{-\alpha}M_\alpha(x/t^\alpha)$ [71,74–76], with the M-Wright function playing again the role of scaling function. Indirect evidence of such scaling comes also from renormalization group calculations [77].

The singularity implied by Eq. (13) at $\lambda = 0^+$ can again be directly obtained by our asymptotic analysis, since the behavior of $M_\nu(z)$ given in Eq. (12) at large positive z holds in the whole $0 < \nu < 1$ interval and is of the form proposed in Eq. (4), now with $z = x/t^\alpha$. Thus, our derivation shows that also in this biased case the scaling function at large positive z is consistent with $\varepsilon(\lambda) \sim \lambda^{1/\alpha}$ for $\lambda \to 0^+$ and with the Fisher relation, for both subdiffusion ($\nu = \alpha < 1/2$) and superdiffusion ($\nu = \alpha > 1/2$). We notice that for $1 < 1/\alpha < 2$ the second-order derivative of $\varepsilon(\lambda)$ diverges, meaning that the scaled variance of the displacement diverges for long times as in the case of the total magnetization in Ising criticality [2].

A remarkable feature of the biased case is that the solution of Eq. (14) is not satisfying scaling in the $t \to \infty$ limit for $x < 0$. Indeed, for $x < 0$ one finds $p(x,t) \sim t^{-\alpha}e^x$ [49], which also shows that the total probability of negative displacements tends to annihilate in the limit. However, the global behavior of $\varepsilon(\lambda)$ can be exactly connected with the solution $p(x,t)$ of Eq. (14) on the whole x axis by switching to Laplace transform in time for both p and G [49] and exploiting results in [74]. The biased case provides a remarkable example of the consequences for critical singularities of a probability distribution exhibiting one-sided scaling, outlining a scenario with no counterpart in equilibrium.

Large deviation theory was recently employed to estimate the propagator of CTRWs with exponential and gamma waiting time distribution [78] and its asymptotics in the fat-tailed case [79]. Integrating our results with the framework of large deviation theory allows us to characterize direct implications on fluctuations [41]. Since $\varepsilon(\lambda)$ is convex and differentiable, for the PDF of the "velocity" variable $v = x/t$ a rate function $I(v)$, satisfying for $t \to \infty$

$$p(x/t = v, t) \sim e^{-tI(v)}, \qquad (15)$$

can be obtained by application of the Gärtner-Ellis theorem [80,81] as the Legendre-Fenchel transform of ε [82]:

$$I(v) = \sup_{\lambda \in \mathbb{R}}[v\lambda - \varepsilon(\lambda)]. \qquad (16)$$

This rate function is equal to zero at $v = 0$, consistently with the fact that there is no conventional current in biased anomalous diffusion. Simple calculations [49] also show that the branches of ε at $\lambda \sim 0^+$ and $\lambda \sim \lambda_0^-$ determine, respectively, $I(v) \sim v^{1/(1-\alpha)}$ around $v = 0^+$ and $I(v) \sim -v$ around $v = 0^-$. So, the two singularities of ε merge into a single singular point at $v = 0$, where $I(v)$ is clearly asymmetric [83]. Analogously, we find that also in the unbiased case the rate function is singular for $v = 0$. The fact that this singularity is determined by the asymptotics of the scaling function leads us to expect an analogous behavior for the rate function of the magnetization in Ising criticality, which was the subject of most recent investigations [88].

The Gallavotti-Cohen symmetry of the SCGF in Eq. (13) allows us also to show that $I(v) - I(-v) = \lambda_0 v$ for any v [49], confirming validity of the fluctuation theorem [65,66,89] for biased anomalous diffusion, consistently with earlier results in Ref. [74]. Remarkably, the separate analysis of the branches of the rate function clearly shows that only the negative branch—the one contrary to the bias—is providing a linear contribution in v, which ultimately ensures validity of the fluctuation theorem.

The examples discussed above are of anomalous diffusion satisfying the Fisher relation $\delta = \nu/(1-\nu)$, even in the biased case. This relation, which we could clearly link to the requirement of standard extensivity in time for the cumulant generator, is expected to be satisfied by a large class of problems in which the diffusion step does not depend on position in space [12]. This class includes diffusion on fractals [29] and on percolation clusters [28]. Still, if one gives up the requirement of standard extensivity for $\log G$ our approach applies also to problems belonging to the Richardson class [12,90,91]—related to processes with position-dependent step sizes—by enforcing the characterizing relation $\delta = (1-\nu)/\nu$. For this class our Eq. (5) foresees a nonstandard extensivity in time, i.e., $\log G \propto t^{\nu/(1-\nu)}$, and a singularity $\propto \lambda^{1/(1-\nu)}$ for the consistently defined ε. We could verify [92] these properties by exact calculations for a process of anomalous diffusion in an inhomogeneous medium introduced in [93]. Thus, our approach enables us to characterize anomalous diffusion belonging to both Fisher and Richardson classes. Other directions we point out are related to systems dealing with diffusion on fractals [94], subdiffusion with static disorder [95], correlated CTRWs [96], and superstatistical subdiffusion in viscoelastic environments [97]. It would be of great interest to use our methods to explore the existence of singularities in the generating functions of such systems. A remarkable feature of our analysis is also the possibility of its application to other counting observables. An example is that of the total entropy produced by the biased process at a given time [89]. Using methods from [65,66] one can realize that, while the zero "velocity" in such a model implies a zero average rate of entropy production, critical singularities determine a divergence of the scaled variance for $t \to \infty$. Thus, anomalously large fluctuations take place for this quantity like for the total magnetization of an Ising model at criticality [2]. We plan to investigate the directions outlined above in a more extended work.

In summary we demonstrated the existence of power law critical singularities for the SCGFs of paradigmatic models of anomalous diffusion. Exact results fully support the link we predict between these singularities and the decay of the non-Gaussian scaling function of the displacement at asymptotic absolute values of its argument. The Fisher relation linking this decay to the diffusion exponent is shown to follow from the extensivity in time of the cumulant generator, while a peculiar power law prefactor excludes corrections $\propto \log(t)$ for this function. For biased diffusion two singularities with extreme asymmetry simultaneously result from a PDF with one-sided scaling, determining peculiar singular behavior in the rate function, but not preventing validity of the fluctuation theorem. We have also shown that the singularity generation mechanism valid for anomalous diffusion is based on exactly the same form of scaling function decay once postulated, but never proved, for Ising systems in equilibrium [34,35]. This suggests the possibility of a general probabilistic explanation of this form in anomalous scaling.

G. T. is supported by the Center for Statistical Mechanics at the Weizmann Institute of Science, the Grant No. 662962 of the Simons foundation, the grants HALT and Hydrotronics of the EU Horizon 2020 program and the NSF-BSF Grant No. 2020765. A. C. acknowledges support of the Polish National Agency for Academic Exchange (NAWA). We thank David Mukamel, Oren Raz, and Gregory Falkovich for useful discussions.

*gianluca.teza@weizmann.ac.il

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