Large Fluctuations for Spatial Diffusion of Cold Atoms

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We use a new approach to study the large fluctuations of a heavy-tailed system, where the standard large-deviations principle does not apply. Large-deviations theory deals with tails of probability distributions and the rare events of random processes, for example, spreading packets of particles. Mathematically, it concerns the exponential falloff of the density of thin-tailed systems. Here we investigate the spatial density $P_t(x)$ of laser-cooled atoms, where at intermediate length scales the shape is fat tailed. We focus on the rare events beyond this range, which dominate important statistical properties of the system. Through a novel friction mechanism induced by the laser fields, the density is explored with the recently proposed non-normalized infinite-covariant density approach. The small and large fluctuations give rise to a bifractal nature of the spreading packet. We derive general relations which extend our theory to a class of systems with multifractal moments.

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In many diffusion processes, the concentration of particles starting at the origin spreads out like a Gaussian, which is fully characterized by the mean-squared displacement. This results from the widely applicable Gaussian central limit theorem (CLT) [1]. Many other physical systems are described by Lévy's CLT (see, e.g., [2]). The latter deals with the sum of independent identically distributed random variables whose own distribution is heavy tailed; as a result it yields a density so broad that its variance diverges [3]. In this Letter we study the spatial expansion of an atomic cloud undergoing Sisyphus laser cooling [4,5]. Here the diffusion switches between three statistical phases, depending on the relative strength of the noise versus the damping [6–8]. In the Gaussian phase, which corresponds to deep optical lattices (see below), small fluctuations mean that the central shape of the particle packet obeys the standard CLT. The Lévy phase is dominated by rare large fluctuations and the particle packet, not too far from the origin, is described by the Lévy distribution [7,9]. In the heating phase, at shallow lattices, fluctuations overcome the damping. Our focus is on the two CLT phases.

In an experimental situation, e.g., [9], diverging moments such as the infinite dispersion predicted by the Lévy CLT are unphysical because at finite times no particles traveling at finite velocities can be found infinitely far from their origin. The finiteness of all the moments requires that the fat tail of the distribution be cut off beyond some point. A full characterization of the system demands that this far asymptotic regime be captured correctly, beyond the intermediate asymptotic power law of the Lévy density. This requires a new theory for the large fluctuations of the system. Similarly, the standard CLT may correctly predict the variance of the system, as in the Gaussian phase of the Sisyphus process, but completely fail in the description of the rare events which determine higher moments.

In simple coin-tossing random walks (e.g., [10]) and other processes (e.g., [11–13]) when the falloff of the probability of the observable of interest is exponential, rare fluctuations are often studied with large-deviations theory [14]. Mathematically, this requires that the cumulant-generating function be smooth. However, Lévy processes [2,15,16] and other fat-tailed systems, where the decay rate is a power law and cumulants may diverge, do not meet this requirement [14]. We show, with the example of the physically tunable system of cold atoms, that the recently introduced infinite-covariant density (ICD) approach [17,18], based on the moment- (as opposed to the cumulant-) generating function, constitutes a superior option for correctly handling heavy-tailed distributions. We discuss the applicability of this approach and its results to a large class of systems via universal relations.

Model.—In the semiclassical approximation, the trajectory of a Sisyphus-cooled atom, starting at the origin $x(0) = 0$, with $v(0) = 0$, is determined by the Langevin equations [6] [see the Supplemental Material (SM) for a more in-depth review [19]],

$$\dot{v}(t) = \mathcal{F}(v) + \sqrt{2D}\Gamma(t), \qquad \dot{x}(t) = v(t), \qquad (1)$$

where $\mathcal{F}(v) = -v/(1+v^2)$ is the deterministic cooling force, in dimensionless units [17] (physical units in SM [19]). Asymptotically, $\mathcal{F}(v) \sim -v$ when $v \ll 1$ and $\mathcal{F}(v) \sim -1/v$ when $v \gg 1$. $\Gamma(t)$ is a Gaussian white noise with zero mean and $\langle \Gamma(t)\Gamma(t')\rangle = \delta(t-t')$. $D = cE_R/U_0$, where U_0 is the depth of the optical lattice, E_R is the recoil energy, and $c \approx 20$ is a constant whose precise value is specific to the type of atoms used in the experiment [5,6]. U_0, and hence D, may be tuned in the lab, and are the control parameters of the system. Several anomalous statistical predictions of this model, Eq. (1), both in and out of equilibrium, have been confirmed in experiments [9,20,21].

We wish to study the large fluctuations of the probability density function (PDF) of the particles' positions at time t, $P_t(x)$. Its Fourier transform, $\int_{-\infty}^{\infty} \exp(ikx) P_t(x) dx$, from $x \to k$, is the moment-generating function [3]

$$\hat{P}_t(k) = 1 + \sum_{m=1}^{\infty} \frac{(ik)^{2m}}{(2m)!} \langle x^{2m}(t) \rangle. \quad (2)$$

The strategy we will employ is to derive the moments of the process, $\langle x^{2m}(t) \rangle$, for $m = 1, 2, \ldots$ (odd moments are zero by symmetry), perform the summation in Eq. (2) and invert this function to obtain the density in x space. Naively, we would expect a normalized density to emerge, but this, as we will show, is not the case.

Scaling arguments for a non-normalizable state.—An initial insight into $P_t(x)$ is gained as follows: Let $W_t(x, v)$ be the phase-space distribution of the particle packet, at time t. Because for $v \gg 1$ the friction vanishes as per Eq. (1), in this case we expect a scaling $v \propto t^{1/2}$. By integration over time, this implies $x \propto t^{3/2}$. Based on these scaling arguments we may write $W_t(x, v) \sim t^{-\xi} f(x/t^{3/2}, v/t^{1/2})$. To determine the exponent ξ we may first use a simple argument (later we derive this rigorously): Note that when $D < 1$ the marginal velocity equilibrium density is [17,20,22–24]

$$\lim_{t\to\infty} \mathbf{P}_t(v) \to \mathbf{P}_{\text{eq}}(v) \sim |v|^{-1/D}, \quad (\text{when } v \gg 1). \quad (3)$$

The heating phase, where $D > 1$, is left out of the context of this Letter because an equilibrium state does not exist there. By definition, this velocity density is related to the phase-space distribution via

$$\mathbf{P}_{\text{eq}}(v) = \lim_{t\to\infty} t^{-\xi+3/2} \int_{-\infty}^{\infty} f\left(\frac{x}{t^{3/2}}, \frac{v}{t^{1/2}}\right) d\left(\frac{x}{t^{3/2}}\right). \quad (4)$$

Hence, from Eqs. (3) and (4) we find $\xi = 3/2 + 1/(2D)$.

Using this result, integration of the scaling solution over $d(v/t^{1/2})$ yields $P_t(x) \sim \mathcal{I}(z)/t^{1+1/(2D)}$, where $z = x/t^{3/2}$. This suggests, and indeed our rigorous theory shows, that there exists a limit such that

$$\mathcal{I}(z) = \lim_{t\to\infty} t^{\beta} P_t(x), \quad (5)$$

where $\beta = 1 + 1/(2D)$. This limit is interesting because if we integrate Eq. (5) over $dz = d(x/t^{3/2})$, we get from the normalization of $P_t(x)$ that the integral $\int_{-\infty}^{\infty} \mathcal{I}(z) dz \to \infty$. Therefore, $\mathcal{I}(z)$ is not a normalized density, but rather a scaling solution that captures the nonuniform convergence of the particle packet. Given $D < 1$, the process is also described by the Gaussian or Lévy CLT; however, as mentioned earlier, this does not describe the rare fluctuations. In this sense, the non-normalized state $\mathcal{I}(z)$, being a limiting solution, is complementary to the CLT. The scaling limit, Eq. (5), can also be argued for from the Kramers equation for $W_t(x, v)$, see the SM [19]. Figure 1 presents simulation data

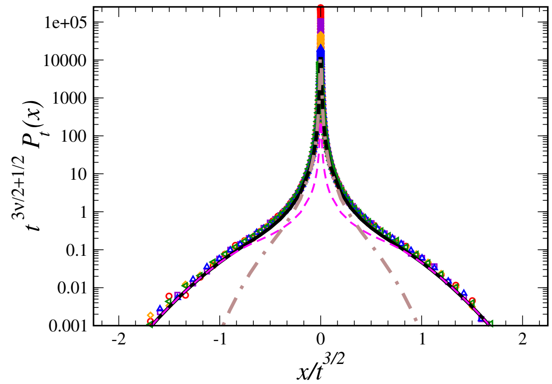

FIG. 1. Convergence of the particles position density in Sisyphus cooling, with $D = 0.4$ ($\nu = 7/6$), to the form of the ICD, Eq. (12). Langevin simulation results [25] are shown for $t = 1000$ (green left-pointing triangles), $t = 1778$ (blue up-pointing triangles), $t = 3162$ (orange diamonds), $t = 5623$ (purple squares), and $t = 10000$ (red circles). The scaling limit function $\mathcal{I}(z)$, based on the areal PDFs of the Bessel excursion and the meander, is presented by the solid black line. Asymptotic theory for $(x/t^{3/2}) \ll 1$ (dot-dashed brown line) and $(x/t^{3/2}) \gg 1$ (magenta dashed line) correspond to Eqs. (10) and (11), respectively. Notice how $\mathcal{I}(z)$ diverges as $x/t^{3/2} \to 0$ and that it is not integrable at this pole.

from the cold-atom system with $D = 0.4$ [25], which converges nicely with increasing time to the theory.

Excursions to untangle Langevin dynamics.—To derive our main results we use a connection between the properties of constrained stochastic paths and Langevin dynamics, established in [6,26]. Let the times $t_1, t_2, \ldots, t_n$ denote the zero crossings of the stochastic process $v(t)$, Eq. (1). The time intervals between the crossing events, $\tau_1 = t_1 - 0, \ldots, \tau_n = t_n - t_{n-1}$, are independent identically distributed random variables, due to the Markovian Langevin process under investigation. The total measurement time is $t = \sum_{i=1}^{n} \tau_i + \tau^*$, where τ^* is the duration of the last interval, in which the velocity does not return to zero. The displacement accumulated during each step is $\chi_i = \int_{t_{i-1}}^{t_{i-1}+\tau_i} v(t) dt$ [for the last step, $\chi^* = \int_{t-\tau^*}^{t} v(t) dt$], and the final random position of the particle at time t is $x(t) = \sum_{i=1}^{n} \chi_i + \chi^*$. In this construction, the velocity path in all but the last interval starts and ends at zero, and is either strictly positive or negative in between; hence, the τ_is are determined by the first-passage time (to the velocity origin) distribution, $g(\tau)$, where $g(\tau) \approx g^* \tau^{-3/2-1/(2D)}$ for large τ [17,26]. The slowly decaying power-law tail of this function means that the last step might be longer than all its predecessors combined, and it cannot be neglected. This is a consequence of the weak friction at large velocities, which allows for very long flights without velocity zero crossings.

Each segment of the path $v(t)$, between zero crossings, is approximated by a Bessel excursion in velocity space [26,27] (see Fig. 2). An excursion in the time interval $[0, \tau_i]$ is a stochastic trajectory constrained to begin close to

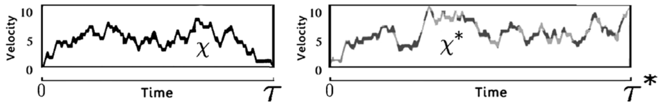

FIG. 2. Left: A Bessel excursion in velocity space follows $\dot{v} = -1/v + \sqrt{2D}\Gamma(t)$, with the path constrained to start at $v(0) = \epsilon$ and end at $v(\tau) = 0$, and remain strictly positive in the time interval $(0, \tau)$. The random area under the path is χ. Right: A velocity Bessel meander, with duration τ^* and area χ^*, starts at $v(0) = \epsilon$ and remains positive, while the final value $v(\tau^*)$ is random.

the velocity origin at $v(0) = \epsilon \to 0$, end at $v(\tau_i) = 0$, and never reach zero between $(0, \tau_i)$ (see, e.g., [28–32]). The area under the ith excursion is $\chi_i \propto \tau_i^{3/2}$, which is naturally correlated to its duration, because longer duration means larger displacement. The last segment, where the velocity path is not conditioned at its final point, is called a velocity Bessel meander [26,33] (Fig. 2).

The term Bessel derives from the fact that for $v \gg 1$, Eq. (1) is related to the Bessel process, describing the radial component of Brownian motion in arbitrary dimensions [7,34–36]. Because we investigate the limit $t \gg 1$, where excursions and meanders are long, we use $F(v) = -1/v$ to calculate the distribution of χ. Our case is equivalent to the Bessel process in an effective fractional negative dimension $d = 1-2/D$ (see the SM [19]). The statistics of χ and the zero-crossing times, τ, determine the random position of the particle, $x(t)$. Because the τs are independent and identically distributed, the zero crossings form a renewal process [26,37], which allows us to analyze the problem analytically based on a generalization of the Montroll-Weiss equation [37,38]. Note that in a previous work [7], our original presentation of this equation incorrectly ignored meanders, which were introduced in our later publication [26].

In the SM [19], we find the following asymptotic expression for the $2m$th moment of the particles' positions, valid for $m \geq 1$ at long times, in the range $1/5 < D < 1$ (the range $D < 1/5$ is addressed below, and details on the prefactor $g^*/\langle\tau\rangle$ are provided in the SM):

$$\langle x^{2m}(t)\rangle \sim \frac{g^*}{\langle\tau\rangle} t^{3m-3\nu/2+1}\left(\frac{\langle\chi^{2m}\rangle_E}{|(3m-3\nu/2)(3m-3\nu/2+1)|} + \frac{2\langle\chi^{2m}\rangle_M}{3\nu|(3m-3\nu/2+1)|}\right), \tag{6}$$

where

$$\nu = \frac{1}{3D} + \frac{1}{3}, \qquad (2/3 < \nu < 2). \tag{7}$$

We denote by $\langle\chi^{2m}\rangle_E = \int_{-\infty}^{\infty}\chi^{2m}B_E(\chi)d\chi$ the $2m$th moment of the areal distribution of the Bessel excursion, $B_E(\chi)$, in the time interval [0, 1]. We denote by $B_M(\chi)$ and $\langle\chi^{2m}\rangle_M$ the distribution and moment, respectively, of the meander in the same time interval. Explicit expressions for $B_E(\chi)$ and $B_M(\chi)$ are provided in the SM (which contains also Ref. [39]) [19]. Note, importantly, that Eq. (6) does not apply for $m = 0$.

Non-normalizable limit function for the PDF.—Using the long-time asymptotic moments provided in Eq. (6) in the moment-generating function, Eq. (2), yields an approximation for $\hat{P}_t(k)$, which we denote as $\hat{P}_t^A(k)$, valid at long times,

$$\hat{P}_t^A(k) = 1 + t^{-3\nu/2+1}\sum_{m=1}^{\infty}\frac{(-1)^m g^*(kt^{3/2})^{2m}}{\langle\tau\rangle(2m)!} \times\left[\int_{-\infty}^{\infty}\chi^{2m}B_E(\chi)d\chi\left(\frac{1}{3m-3\nu/2} - \frac{1}{3m-3\nu/2+1}\right) + \int_{-\infty}^{\infty}\chi^{2m}B_M(\chi)d\chi\frac{2}{3\nu(3m-3\nu/2+1)}\right]. \tag{8}$$

Rearranging, and using the Taylor expansion $\cos(\omega^{3/2}y) = \sum_{n=0}^{\infty}(-1)^n(\omega^{3/2}y)^{2n}/(2n)!$ for the summation, we obtain

$$\hat{P}_t^A(k) = 1 + \frac{g^* t^{-3\nu/2+1}}{\langle\tau\rangle} \times\int_{-\infty}^{\infty}d\chi\int_0^1 d\omega[\cos(\omega^{3/2}k\chi t^{3/2}) - 1] \times\left[\frac{B_E(\chi)}{\omega^{3\nu/2-1}} + \frac{2B_M(\chi) - 3\nu B_E(\chi)}{3\nu\omega^{3\nu/2}}\right]. \tag{9}$$

Immediately below, taking the inverse-Fourier transform from $k \to x$, we drop the term proportional to $\delta(x)$ because this analysis applies only at large x. Calculating the integral over ω, we now obtain the limit function $\mathcal{I}(z)$ in Eq. (5) explicitly. When $z = x/t^{3/2} \ll 1$,

$$\mathcal{I}(z) \approx \frac{g^*}{3\langle\tau\rangle}\langle|\chi|^\nu\rangle_E|z|^{-\nu-1}, \tag{10}$$

where $\langle|\chi|^\nu\rangle_E$ is the νth absolute moment of the excursion [27]. This equation means that $\mathcal{I}(z)$ is nonintegrable around the origin. For $z \gg 1$,

$$\mathcal{I}(z) \approx \frac{4g^*}{9\nu\langle\tau\rangle}|z|^{-\nu-1/3}\int_z^{\infty}|\chi|^{\nu-2/3}B_M(\chi)d\chi. \tag{11}$$

Using the properties of $B_M(\chi)$, in the SM we show that the very far tail is Gaussian, $\mathcal{I}(z) \propto \exp[-3z^2/(4D)]$, at $z \to \infty$ (in our previous work [7], this Gaussian decay was observed from numerics) [19].

The function $\mathcal{I}(z)$ is the ICD of the spatial diffusion of the cold atoms. For every z we find

$$\mathcal{I}(z) = \frac{2g^*}{3\langle\tau\rangle}\frac{1}{|z|^{\nu+1}}\left[\int_{|z|}^{\infty}B_E(\chi)|\chi|^\nu d\chi + |z|^{2/3} \times\int_{|z|}^{\infty}\left(\frac{2}{3\nu}B_M(\chi) - B_E(\chi)\right)|\chi|^{\nu-2/3}d\chi\right]. \tag{12}$$

The term "infinite" means that it is non-normalizable, despite being a limit function of the (obviously normalized)

PDF. "Covariant" means that it is a function of the scaled variable $x/t^{3/2}$. We obtained this non-normalizable solution from the standard moment-generating function, Eq. (2), because we summed over the long-time asymptotic approximations of the moments, rather than their exact finite time values. Equation (12) constitutes the long-time asymptotics of the tail of the PDF, and in that sense it describes the rare fluctuations of the system. Its asymptotic limits, Eqs. (10) and (11) (also plotted in Fig. 1), are controlled exclusively by the excursions for $z \ll 1$ and the meander for $z \gg 1$; thus the far tail is described by a path that did not switch its velocity direction for a duration of the order of measurement time. This is clearly a rare event.

The ICD, $\mathcal{I}(z)$, Eq. (12), gives the long-time limit of all the absolute integer and fractional moments $\langle |x|^q \rangle$ of order $q > \nu$. This also includes the second moment, generally used in experiments as a characterization of the diffusion process. Looking back at Eq. (6), the mean-squared displacement, which is sensitive to the large fluctuations and the tails of the PDF, is obtained via $\langle x^2(t) \rangle = t^{4-3\nu/2} \langle z^2 \rangle_{\mathcal{I}}$, where $\langle z^2 \rangle_{\mathcal{I}} = \int_{-\infty}^{\infty} z^2 \mathcal{I}(z) dz$. For every $q \in \mathbb{R}$, if $\mathcal{I}(z)$ [Eq. (12)] is integrable with respect to $|z|^q$, then the ICD determines $\langle |x|^q(t) \rangle$ (i.e., the qth absolute moment [40]) via $\langle |x|^q(t) \rangle = t^{3q/2-3\nu/2+1} \langle |z|^q \rangle_{\mathcal{I}}$, where $\langle |z|^q \rangle_{\mathcal{I}} = \int_{-\infty}^{\infty} |z|^q \mathcal{I}(z) dz$.

When $q \le \nu$, $\mathcal{I}(z)$ is nonintegrable with respect to the observable $|z|^q$ and small absolute moments that are less sensitive to large fluctuations are given by the Lévy distribution, $\mathcal{L}_\nu(\cdot)$, as found in [7]. For this CLT result the effect of the meander is irrelevant; further details are provided in the SM [19]. This second long-time limit function has the scaling shape $(K_\nu t)^{-1/\nu} \mathcal{L}_\nu [x/(K_\nu t)^{1/\nu}]$, where K_ν is a constant (see the SM for more information [19]) [7,26]. For all the absolute moments we find the biscaling behavior

$$\langle |x|^q(t) \rangle \propto \begin{cases} t^{q/\nu} & q < \nu \\ t^{3q/2-3\nu/2+1} & q > \nu \end{cases}. \tag{13}$$

Such multifractality is known as strong anomalous diffusion [41]. It represents the multiscaling nature of the underlying PDF. Note that as $q \to \nu$ from above, the coefficient of $\langle |x|^q \rangle$, given by the analytic continuation of Eq. (6), diverges. The same happens when evaluating the moments using the Lévy scaling function, approaching ν from below.

The derivation of $\mathcal{I}(z)$, Eq. (12), was performed in the range of D where the variance is provided by the ICD. However, the scaling arguments at the beginning of this Letter suggest that such a function should be found whenever the power-law equilibrium state in velocity space, Eq. (3), exists —namely, for all $0 < D < 1$. In the range $0 < D < 1/5$ one will find that $\langle x^2 \rangle \propto t$ and the central part of the spreading packet is Gaussian. But even in this Gaussian regime, standard large-deviations theory does not apply; instead, the ICD given by Eqs. (5) and (12) ensures the finiteness of large moments, beyond the mean-squared displacement.

Generality of the infinite-covariant density approach.— ICDs may be naturally related to multifractality (physical examples provided below). In particular we now derive a general relation between exponents describing the bifractal moments, the central part of the packet (with bulk fluctuations described by the Lévy CLT), and the exponents describing the ICD. When absolute moments of order $q > q_c$, where $q_c > 0$ defines some critical moment, scale faster in time than smaller ones, a scaling function $\mathcal{I}(\tilde{z} = x/t^\alpha)$ may describe the large fluctuations at long times via $\langle |x|^q(t) \rangle \to t^{q\alpha-\beta+\alpha} \int_{-\infty}^{\infty} |\tilde{z}|^q \mathcal{I}(\tilde{z}) d\tilde{z}$ $(\beta > \alpha > 0)$. These absolute moments and exponents should be obtained specifically for any given model or system, for example, using experimental data. In this case, one will find that $\mathcal{I}(\tilde{z}) = \lim_{t\to\infty} t^\beta P_t(x)$, where $P_t(x)$ is the normalized PDF. This limit function is, hence, a non-normalizable ICD [since obviously $\langle x^0(t) \rangle = 1$, then $\int_{-\infty}^{\infty} \mathcal{I}(\tilde{z}) d\tilde{z} \to \infty$]. If around the origin the PDF is represented by the Lévy distribution [42] $t^{-1/\nu} \mathcal{L}_\nu(x/t^{1/\nu})$ (for arbitrary $0 < \nu < 2$, not necessarily related to D), one will find (by "stitching" this limit function and the ICD at a central region of x, as in [18], see the SM [19]) the following relation between the scaling exponents: $\alpha - \beta + \alpha\nu = 1$. In [43], for example, the authors study a nonlinearly coupled continuous-time random walk with $(\alpha, \beta) = (\boldsymbol{\alpha}, \boldsymbol{\alpha} + \boldsymbol{\beta} - 1)$, which according to our analysis yields $\nu = \boldsymbol{\beta}/\boldsymbol{\alpha}$ ($\boldsymbol{\alpha}, \boldsymbol{\beta}$, in bold, refer to the parameters in this reference). Our prediction is consistent with their results. A more general relation links the exponents α, β of the ICD and the central power-law where $x \sim t^{1/\nu}$ to the critical moment of the biscaling, q_c, as follows: $\alpha - \beta + q_c \alpha = q_c/\nu$. This is consistent, e.g., with the exponents found for transport on two-dimensional Lévy quasicrystals, studied in [44]. The agreement with [43,44] suggests an ICD in these systems as well.

The moment-generating function is a natural tool for deriving the ICD for many processes, e.g., random walks with multiplicative noise [45]. While nonanalytical behavior of the moments raises a red flag for standard large-deviations theory, it argues for the use of the ICD approach as the appropriate theory for the large fluctuations. Finding this function is crucial for characterizing the rare events. The limit law given by the ICD in Eq. (5) for the rescaled PDF replaces the large-deviations principle, according to which the falloff of the tails in thin-tailed systems is controlled by the rate function $Q(x/t)$, where $Q(x/t) = \lim_{t\to\infty} \ln [P_t(x)]/t$.

Discussion.—CLTs play an important role in statistical physics, but of no less importance may be the proper characterization of the deviations from them. The ICD was previously found, e.g., for Lévy walks [18,46]. Because dual scaling of the moments and fat-tailed distributions are very common, we speculate that ICDs will describe a large class of systems, e.g., Lévy glasses [47], fluctuating surfaces [48], motion of tracer particles in the cell [49]. and diffusion on lipid bilayers [50]. To identify the ICDs in these diverse systems requires further work. The Sisyphus-cooled atoms system is special because it allows us, by tuning the intensity of the lasers, to find regimes where large fluctuations in the tails are non-negligible. Our ICD solves the serious problem of the diverging variance expected by the Lévy distribution in this system; however, the latter insures the normalizability of the PDF. A full description of the system requires both functions.

Generally, the ICD may depend on the protocol of the preparation of the system. Its shape may change if, prior to measurement, the particles are relaxed by interacting with the lasers in a spatial trap for some time t^*, where $t^* \gg t$. Our results apply in the opposite limit. The dependence on t^* leads to transport that depends on the preparation time. Dechant and Lutz found theoretically not biscaling, but triscaling of the moments in this case [8]. Finally, we point out that the function $f(z, \tilde{v})$ (where $\tilde{v} = v/t^{1/2}$) in Eq. (4) is itself an ICD, as it is clearly not normalizable. Elucidating its properties is an important future goal.

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