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Universal fluctuations in subdiffusive transport

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Abstract – Subdiffusive transport in tilted washboard potentials is investigated within the fractional Fokker-Planck equation approach by making reference to the associated continuous time random walk (CTRW) framework. The scaled subvelocity is shown to obey a universal law. The latter is defined by the index of subdiffusion α and the mean subvelocity only. Interestingly this law depends neither on the size of the system or measurement time, nor on the bias strength or on the specific form of the washboard potential. These scaled, universal subvelocity fluctuations emerge due to the weak ergodicity breaking and are vanishing in the limit of normal diffusion. The results of the analytical reasoning are corroborated by Monte Carlo simulations of the underlying CTRW.

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Introduction. – A process of directed motion, for example, the motion of a Brownian particle under influence of a constant force, can be characterized by its mean velocity v. The mean velocity is measured using a ruler and a stopwatch in one of two different setups: One can either measure the distance l covered within a fixed time interval T and define the mean velocity as $v = l/T$ or, like it is done in track-and-field competitions, fix the distance L and measure the time interval t necessary to cover it, yielding $v = L/t$. Thus, one can distinguish between the fixed time (FT) velocities and the time-of-flight (TOF) velocities. Both these definitions are known from elementary physics course and these *do not* imply any special averaging procedure. However, when the instantaneous velocity exists (*e.g.*, for normal Brownian motion with inertia) the FT mean velocity can be considered as the time-average of the instantaneous velocities over the time span T. The TOF velocity obeys a somewhat different statistical nature as it implies the averaging over the random time span. For our Brownian particle moving under the influence of the constant force F both setups yield values of (mean) velocity which in the limit of $T \to \infty$ for the FT measurement or $L \to \infty$ for the TOF measurements reach the *same* sharp value v. In the "normal" situation the typical time t necessary to overcome the distance L grows linearly with L in the TOF setup, and the relative fluctuations of the corresponding times in individual measurements decay. On the other hand, the typical distance l covered during the time T grows on the average linearly with T in the FT setup, while the relative fluctuations in the distances decay as well. This implies that monitoring a *single particle* over a long path or during a long time yields a sharp value for its velocity, without any necessity to additionally average the results over an ensemble of repeated measurements. The property of a physical observable to reach a sharp value in a large system or, likewise, for a long observation time is typically referred to as "self-averaging" property.

For the case of biased anomalous subdiffusion the situation drastically differs. In what follows the subdiffusive motion $x(t)$ is modeled by a continuous time random walk (CTRW) with the waiting time probability density (WTD) on sites assuming (for $t \gg \tau$) a power law; *i.e.*,

$$\psi(t) \sim c(t/\tau)^{-1-\alpha} \quad (1)$$

with a diverging mean waiting time, *i.e.*, with $0 < \alpha < 1$. In (1), τ is a characteristic time scale and the prefactor $c = \alpha[\tau\Gamma(1-\alpha)]^{-1}$ is introduced for the sake of simplicity of further calculations. For example, charge transport processes in disordered, amorphous media can be subdiffusive due to a trap-like transport mechanism with a trapping time distribution [1–6] similar to (1). This constitutes an approximation which can be justified for samples of macroscopic, but finite size L [3]. The considered CTRW model can also be deduced from a Markovian diffusion model with quenched disorder [7]. The corresponding averaged current $J(t) \propto \mathrm{d}\langle \delta x(t)\rangle/\mathrm{d}t$, $\delta x(t) = x(t) - x(0)$, is a transient which decays to zero upon increasing time [2,3]. This decay of the current is due to the fact that with a subdiffusive CTRW the typical displacement under a constant force F grows as $\delta x \propto t^\alpha$. In the case of the measured decaying photocurrent occurring in thin amorphous films [3] one can, however, define an anomalous current as $J_\alpha = \int_0^t J(t')\mathrm{d}t'/t^\alpha = Q(t)/t^\alpha$, where $Q(t)$ is the transferred charge. This quantity is quasi-stationary (at shorter times) and then subsequently fades anyway when the particles reach the boundaries of the system.

The measured current in a macroscopic sample presents a multi-particle, ensemble-averaged quantity. Concentrating on the motion of a single particle, we can introduce the subvelocity v_α as the characteristics of its motion, namely as $v_\alpha = \Gamma(1+\alpha)l/T^\alpha$ in the FT setup, or as $v_\alpha = \Gamma(1+\alpha)L/t^\alpha$ for the TOF setup. As it will be elucidated below, the corresponding subvelocities are no longer self-averaging quantities; *i.e.*, these do not tend to sharp values for $T \to \infty$ or $L \to \infty$. Moreover, we demonstrate that in the limit of a large size L or a large span T the distributions of v_α measured in both setups are identical and of nonvanishing width. This means that the subvelocity dynamics exhibits *universal fluctuations*. Nevertheless, one can define an *ensemble*-averaged mean subvelocity $\overline{v}_\alpha = \Gamma(1+\alpha)\langle \delta x(t)\rangle/t^\alpha$ [8,9] which presents a quasi-stationary quantity for a sufficiently large L (neglecting finite size effects, *i.e.*, $L \to \infty$ when assuming limit $t \to \infty$). We further demonstrate that this mean subvelocity is the only quantity (apart from α) which fully determines the distribution of subvelocities in the individual runs.

The absence of a finite mean trapping time leads to the weak ergodicity breaking [10–12] in the relevant transport processes. This feature is at the root for the absence of self-averaging. In particular, the mean subvelocity of individual particles (before ensemble averaging) is a random quantity; this is so because the time and the ensemble averages are not equivalent. This random character is also preserved for the measurement of the diffusion coefficient of individual particles [13,14], where fluctuations of the corresponding mobility have also been discussed. It should be noted, however, that the moving time averaging procedures discussed in those latter references are different from the ones implied by the FT or TOF setups introduced here.

Using numerical simulations we show that the same conclusions hold as well in more complex situations, as, for example, for a CTRW in a tilted washbard potential, *i.e.*, the situation discussed in refs. [8,9]. In the continuous space limit, it is described by the fractional Fokker-Planck equation (FFPE) [4,8,9]

$$\frac{\partial^\alpha P(x,t)}{\partial t^\alpha} = \kappa_\alpha \frac{\partial}{\partial x}\left[e^{-\beta U(x)}\frac{\partial}{\partial x}e^{\beta U(x)}P(x,t)\right], \qquad (2)$$

which we formulate here in the form with the fractional Caputo derivative $\partial^\alpha P(x,t)/\partial t^\alpha = (1/\Gamma(1-\alpha))\int_0^t \mathrm{d}t'[t-t']^{-\alpha}\partial P(x,t')/\partial t'$ [5,8]. This equation is equivalent to the original FFPE of ref. [4] with the Riemann-Liouville fractional derivative acting on the right-hand side. In eq. (2), $U(x) = V(x) - Fx$, where $V(x+\lambda) = V(x)$ denotes a periodic potential with period λ, and $F > 0$ is the biasing force; $\beta = 1/(k_BT)$ is the inverse temperature, and κ_α is the subdiffusion coefficient. A self-averaging in time does not occur [9] and mean subvelocity remains a random variable even in the strict limit $T \to \infty$. As we shall show, the same is valid in the TOF setup: only upon an additional ensemble averaging does the averaged value of subvelocity coincide with the one given by solving analytically the fractional Fokker-Planck equation [8,9]. Again, the distribution of subvelocities in individual runs is governed only by this ensemble-averaged mean subvelocity value and by the index α of the waiting-time distribution.

It is surprising from a physics point of view that this universality class does not depend on imposing a periodic static potential $V(x)$ in addition to an applied constant force F (what seems feasible experimentally, *e.g.*, for a situation with charged particles). It also does not depend on the environmental temperature provided that α is temperature independent. This universality feature is similar in nature with an established universal scaling relation [8,9] between anomalous current and biased diffusion, originally suggested for free (*i.e.*, in the absence of a periodic potential) biased subdiffusive CTRW transport [1,2]. This result is derived below by use of an heuristic argumentation; *i.e.*, by use of a reduction to a coarse-grained CTRW. On the level of ensemble-averaged quantities, we therefore obtain a universal law for the relative fluctuations of (sub-)velocity, or fluctuations of anomalous current, which can be tested experimentally.

Theory for the biased CTRW. – We start out from a CTRW on a one-dimensional lattice with period length a and the WTD in (1). The walk is biased and solely nearest neighbors jumps (this assumption is not restrictive and can be relaxed) occur with force-dependent splitting probabilities q^+ (toward the right) and q^- (toward the left), with $q^+ + q^- = 1$. After n steps, the mean displacement is $l_n = \langle x\rangle = na(q^+ - q^-)$. From now on, we measure distance l in the units of $a^*(F) = a(q^+ - q^-)$. Time will be measured in units of τ and the subvelocity v_α in units of $v_0(F) = \Gamma(1+\alpha)a^*/\tau^\alpha$.

In the *fixed time* setup, we fix the final time T and ask for the probability $p(n,T)$ to make n steps. The answer is well known (see p. 248 in ref. [2]): in the Laplace domain, it reads

$$\hat{p}(n,u) = \frac{1-\hat{\psi}(u)}{u}\left[\hat{\psi}(u)\right]^n. \qquad (3)$$

For $u \to 0$ (*i.e.*, for $T \to \infty$), the leading term expansion of the Laplace transform of WTD in eq. (1) is $\hat{\psi}(u) \sim 1 - u^\alpha$.

This leads to

$$\hat{p}(n,u) \simeq u^{\alpha-1}\exp\left[n\ln(1-u^{\alpha})\right] \simeq u^{\alpha-1}\exp(-nu^{\alpha}) \quad (4)$$

in the limit of large $n\to\infty$. The expression $\exp(-nu^{\alpha})$ is related to the Laplace transform of the one-sided Lévy stable law $\mathcal{L}_{\alpha}(t)$ of index α, being $\tilde{\mathcal{L}}(u)=\exp(-u^{\alpha})$; *i.e.*, in the original time domain it corresponds to $n^{-1/\alpha}\mathcal{L}_{\alpha}\left(n^{-1/\alpha}t\right)$. Considering n as a continuous parameter (distance being $l=n$ in units of a^*) and noting that eq. (4) equals the Laplace transform of

$$-\int_0^T \frac{\mathrm{d}}{\mathrm{d}l}\frac{1}{l^{1/\alpha}}\mathcal{L}_{\alpha}\left(\frac{t}{l^{1/\alpha}}\right)\mathrm{d}t,$$

we obtain, upon applying a change of variable of integration from t to $\xi=t/l^{1/\alpha}$,

$$p(l,T)\simeq -\frac{\mathrm{d}}{\mathrm{d}l}\int_0^{T/l^{1/\alpha}}\mathcal{L}_{\alpha}(\xi)\mathrm{d}\xi = -\frac{\mathrm{d}}{\mathrm{d}l}C_{\alpha}\left(\frac{T}{l^{1/\alpha}}\right).$$

Here, $C_{\alpha}(x)$ is the cumulative distribution function of the one-sided Lévy stable law; *i.e.*,

$$p(l,T)\simeq \frac{1}{\alpha}\frac{T}{l^{1+1/\alpha}}\mathcal{L}_{\alpha}\left(\frac{T}{l^{1/\alpha}}\right).$$

This finding presents in essence a well-known result for the asymptotic behavior of the number of steps in CTRW at fixed time. Thus, we can extract the distribution for the FT-subvelocity $v_{\alpha}=\Gamma(1+\alpha)l/T^{\alpha}$ via a change of the random variable from l to v_{α}, yielding in terms of the scaled subvelocity $\zeta_{\alpha}=v_{\alpha}/v_0(F)$, for all $F>0$,

$$p(\zeta_{\alpha})=\frac{\Gamma(1+\alpha)^{1/\alpha}}{\alpha\zeta_{\alpha}^{1+1/\alpha}}\mathcal{L}_{\alpha}\left[\left(\frac{\Gamma(1+\alpha)}{\zeta_{\alpha}}\right)^{1/\alpha}\right]. \quad (5)$$

This universal form of the subvelocity distribution presents a first major result of our study. In this context it is pertinent to mention that yet another random quantity, namely the time-averaged mobility of a single subdiffusive particle obeys the very same law [14]. The subvelocity here, however, has a quite different physical meaning, despite the fact that the randomness of both quantities, *i.e.*, the time-averaged mobility and the subvelocity, constitutes a manifestation of weak ergodicity breaking. Notably, the time-averaged mobility vanishes in the limit $T\to\infty$ [14]; in contrast, the subvelocity remains finite.

Let us demonstrate that this very same result is recovered also within the *time-of-flight* setup. That is, we are looking for the asymptotic distribution of times to make a large number of n steps. The corresponding distance will assume a sharply peaked distribution around its mean value which can be identified with the sample size L. The random time t necessary to traverse the system of length L in the TOF setup is essentially the time necessary to make n steps. The overall time to make $n\gg 1$ steps tends in distribution to a one-sided Lévy law $n^{-1/\alpha}\mathcal{L}_{\alpha}\left(n^{-1/\alpha}t\right)$. To see this it is sufficient to notice that the Laplace transform of the probability to find this time is given by $\hat{p}(u)=[\hat{\psi}(u)]^n\simeq(1-u^{\alpha})^n\simeq\exp[n\ln(1-u^{\alpha})]\simeq\exp(-nu^{\alpha})$. The distribution of v_{α} is then obtained by the same change of variable as invoked above to arrive again at the identical result in (5). Here, the only difference being that t (instead of T) is now a random variable and $l=L$ is fixed.

The averaged value of the scaled subvelocity with probability density (5) is one, $\overline{\zeta}_{\alpha}=1$, *i.e.*, the subvelocity in (5) is scaled in fact through its averaged value $\overline{v}_{\alpha}(F)=v_0(F)$. All the higher moments are obtained using the change of variable $y=[\Gamma(1+\alpha)/v_{\alpha}]^{1/\alpha}$ and the relation

$$\int_0^{\infty} y^{\eta}\mathcal{L}_{\alpha}(y)\mathrm{d}y=\frac{\Gamma(1-\eta/\alpha)}{\Gamma(1-\eta)} \quad (6)$$

being valid for any $\eta\in(-\infty,\alpha)$ (see ref. [15]). With $\eta=-2\alpha$, one obtains the second moment as

$$\overline{\zeta_{\alpha}^2}=\frac{2\Gamma^2(1+\alpha)}{\Gamma(1+2\alpha)}. \quad (7)$$

The relative fluctuation of subvelocity $\delta v_{\alpha}=\sqrt{\overline{v_{\alpha}^2}-(\overline{v}_{\alpha})^2}/\overline{v}_{\alpha}$ equals the universal scaling relation between the averaged subdiffusion current and the biased diffusion of refs. [1,2,8,9], *i.e.*,

$$\begin{aligned}\delta v_{\alpha} &= \frac{\sqrt{\overline{\delta x^2(t)}}}{\overline{\delta x(t)}}=\sqrt{\frac{2\Gamma^2(1+\alpha)}{\Gamma(1+2\alpha)}-1}\\ &= \lim_{t\to\infty}\frac{\sqrt{\langle\delta x^2(t)\rangle}}{\langle\delta x(t)\rangle}.\end{aligned} \quad (8)$$

This result is by no means trivial: this is so because $\overline{(\cdots)}$ denotes the average over the stationary subvelocity density $p(v_{\alpha})=p(\zeta_{\alpha}=v_{\alpha}/\overline{v}_{\alpha})/\overline{v}_{\alpha}$, while $\langle\cdots\rangle$ is the average over the time-dependent population probabilities $p_i(t)$ of the lattice sites. This constitutes our second main result: It shows that weak ergodicity breaking is at the root of this remarkable scaling relation (8). Put differently, weak ergodicity breaking is responsible for the startling change of the law of subdiffusion from $\langle\delta x^2(t)\rangle\propto t^{\alpha}$ —when $F=0$— to $\langle\delta x^2(t)\rangle\propto t^{2\alpha}$ —when $F\neq 0$; *i.e.*, subdiffusion turns over into superdiffusion for $0.5<\alpha<1$. We remark in this context that the scaling relation (8) between the current and the biased diffusion cannot be employed to deduce the main result in (5).

In particular, for $\alpha=0.5$, eq. (5) simplifies to one-sided Gaussian form (cf., fig. 1),

$$p(\zeta_{1/2})=\frac{2}{\pi}\exp\left(-\frac{1}{\pi}\zeta_{1/2}^2\right) \quad (9)$$

and $\delta v_{1/2}=\sqrt{\pi/2-1}$. For other values of α, a handy approximation to the subvelocity distribution can be obtained using the corresponding small x asymptotic behavior of the Levy-stable distribution [15,16]. It reads

$$p(\zeta_{\alpha})\simeq A(\alpha)(\alpha\zeta_{\alpha})^{\frac{\alpha-1/2}{1-\alpha}}\exp\left[-B(\alpha)\zeta_{\alpha}^{1/(1-\alpha)}\right], \quad (10)$$

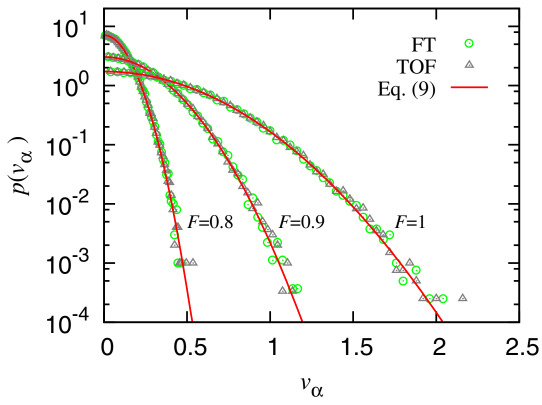

Fig. 1: (Color online) Numerical subvelocity distribution $p(v_\alpha)$ for $\alpha = 0.5$ in both FT and TOF setups for a periodic potential $V(x) = V_0\cos(2\pi x/\lambda)$ and differing bias forces F. v_α is scaled in units of $v_0^*(F) = \overline{v}_\alpha(F)/v_{cr}$, where $\overline{v}_\alpha$ is given by (11) and $v_{cr} = F_{cr}\kappa_\alpha/(k_BT)$. The solid lines depict the theoretical result (9): $p(v_\alpha) = p(\zeta_\alpha = v_\alpha/v_0^*)/v_0^*$.

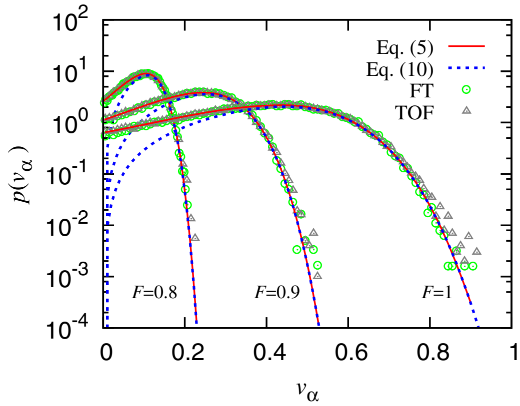

Fig. 2: (Color online) Numerical subvelocity distribution $p(v_\alpha)$ *vs.* the analytical approximation in eq. (10) and the exact result in eq. (5) for $\alpha = 0.8$. The same scaling applies as in fig. 1.

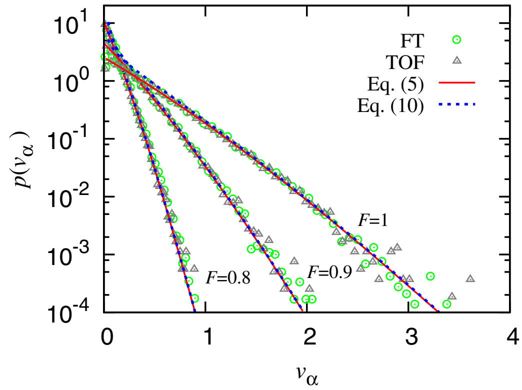

Fig. 3: (Color online) Same as in fig. 1 for $\alpha = 0.2$.

where $A(\alpha) = [\sqrt{2\pi(1-\alpha)}\Gamma(1+\alpha)]^{-1}$ and $B(\alpha) = (1-\alpha)\alpha^{\alpha/(1-\alpha)}\Gamma(1+\alpha)^{-1/(1-\alpha)}$. For $\alpha = 0.5$ this approximation reproduces the exact result in (9). For $0.5 < \alpha < 1$, it correctly predicts that the distribution function is non-monotonic, possessing a maximum (see fig. 2) which becomes sharp for $\alpha \to 1$. In this limit, the relative fluctuation vanishes, $\delta v_\alpha \to 0$, and the velocity distribution tends to the delta function centered at $\overline{v}_\alpha$. The correct value $p(0)$, however, always remains finite for $\alpha < 1$, implying that there are always particles which become immobilized. For $\alpha \leqslant 0.5$, $p(v_\alpha)$ decays monotonically. Moreover, for small $\alpha \to 0$, the distribution becomes nearly exponential, consistent with $\delta v_\alpha \to 1$ in this limit (see fig. 3).

All our analytical findings are confirmed by the numerical simulations of the underlying CTRW in a biased cosine potential $U(x) = V(x) - Fx$ with $V(x) = V_0\cos(2\pi x/\lambda)$, using the Mittag-Leffler distribution $\psi(\tau)$ and the numerical algorithm detailed in [9]. In doing so, we use here a different method to extract the numerically exact generator for the Mittag-Leffler distribution by using the relation due to Kozubowski [17] (see also ref. [18]). It is surprising that all these results hold *universally*, *i.e.*, they are independent of the details of periodic potential and the temperature. This fact is numerically confirmed with figs. 1–3 for a washboard potential (details are given below). To elucidate our main finding we next make use of a reasoning put forward originally in ref. [8].

Theory for washboard potentials. – We dilate the lattice by introducing many more points with separation $\Delta x \to 0$. The residence time distributions on every point are chosen to be Mittag-Leffler distributions [5] with different time scaling parameters $\tau_i = 1/\nu_i$. This distribution belongs to the same class in (1). Each point i is characterized also by the right and the left nearest-neighbor jump probabilities $q_i^{\pm} = g_i^{\pm}/(g_i^+ + g_i^-)$, and by the fractional forward and backward rates, $g_i^{\pm} = q_i^{\pm}\nu_i^{\alpha}$, respectively. These quantities follow from the potential $U(x)$ as $g_i^{\pm} = (\kappa_\alpha/\Delta x^2)\exp[-\beta(U_{i\pm 1/2} - U_i)]$ so that the Boltzmann relation $g_{i-1}^+/g_i^- = \exp[\beta(U_{i-1} - U_i)]$ is fulfilled. Here, $U_i \equiv U(i\Delta x)$, $U_{i\pm1/2} \equiv U(i\Delta x \pm \Delta x/2)$, and $\nu_i = (g_i^+ + g_i^-)^{1/\alpha}$. The generalized master equation for such a CTRW is [5,8]

$$\frac{\partial^\alpha P_i(t)}{\partial t^\alpha} = g_{i-1}^+ P_{i-1}(t) + g_{i+1}^- P_{i+1}(t) - (g_i^+ + g_i^-)P_i(t).$$

In the spatial continuous limit $\Delta x \to 0$, it yields the FFPE (2). In this way, we simulate the stochastic dynamics associated with (2) on a sufficiently dense grid with step Δx, using the Monte Carlo algorithm from [9].

We next consider a periodic potential with period λ subjected to a finite bias force F. One can course-grain the corresponding limiting CTRW and map it onto a new biased CTRW with the lattice period λ, *i.e.*, we average over spatial period λ. The precise form of the coarse-grained WTD is not known. However, it belongs to the same class as (1); only the time parameter τ is correspondingly changed together with the coarse-grained splitting probabilities $q^{\pm}$. We note that such course-graining of Markovian, normal diffusion in washboard potentials yields a *non-Markovian* CTRW that gives rise to such profound effects as giant acceleration of diffusion [19, 20]. In clear contrast, our original CTRW constitutes a non-Markovian, weakly non-ergodic stochastic process, possessing infinite memory. Coarse-graining it further does not change the universality class because no correlations between the residence times in non-overlapping spatial domains occur.

For arbitrary periodic tilted potentials, the result for the ensemble-averaged subvelocity was obtained in refs. [8,9]. It reads

$$\overline{v}_\alpha(F) = \frac{\kappa_\alpha \lambda\ [1-\exp(-\beta F\lambda)]}{\int_0^\lambda \mathrm{d}x \int_x^{x+\lambda} \mathrm{d}y \exp(-\beta\,[U(x)-U(y)])}\,. \qquad (11)$$

In all our numerical simulations we used the archetype cosine potential $V(x)=V_0\cos(2\pi x/\lambda)$. The grid contains 200 points within each spatial period. A scaled temperature of $k_BT=0.1\,V_0$ is used throughout and the force F is scaled in units of the critical force F_{cr}, where with $F>F_{cr}$ the potential $U(x)$ becomes monotonic without barriers in between. The number of particles is $N=10^5$. The different lines for fixed α and different bias values F are due to the different values of the scaling parameter $\overline{v}_\alpha(F)$, calculated in accordance with (11). In accordance with our theory, all the related lines perfectly coincide (not shown) after re-scaling $v_\alpha \to \zeta_\alpha = v_\alpha/\overline{v}_\alpha(F)$, $p\to p\cdot\overline{v}_\alpha(F)$, for all $F>0$. The numerical results thus corroborate well with theory.

In conclusion, a prominent finding of this work is that the feature of weak ergodicity breaking is responsible for the universal scaling relation (8) between the anomalous current and the subdiffusion occurring in arbitrary tilted periodic potentials. This intriguing result follows from the universal law for the theoretically deduced subvelocity distribution in (5), this being the main result of this work.

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