

Kinetic Energy Diffusivity and Scaling Velocity Correlation Functions

Jing-Dong Bao^{1,*} and Fabio Marchesoni^{2,3,†}

¹*School of Physics and Astronomy, Beijing Normal University, Beijing 100875, China*

²*School of Physics Science and Engineering, Tongji University, Shanghai 200092, China*

³*Dipartimento di Fisica, Università di Camerino, I-62032 Camerino, Italy*

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We propose a Green-Kubo-like relation for kinetic energy diffusivity to investigate the interplay between ergodicity and anomalous diffusion. This approach introduces a fluctuation metric for the time-averaged kinetic energy, which holds for scaling velocity correlation functions. We demonstrate that as stationary diffusive systems transition into an effective ergodic phase, their kinetic energy metric converges to a universal law. This finding provides a robust framework for understanding the dynamics of such systems. Applications to protein folding and single-particle tracking illustrate the practical utility of our approach, offering a clear prescription for extracting key physical parameters, such as the friction constant and relaxation time, from finite experimental datasets. Importantly, this method remains effective even when the underlying processes exhibit weak ergodicity breaking or are bounded. Furthermore, we explore the nonergodic transition associated with the aging velocity correlation function observed in granular gases.

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The notion of diffusion and the ergodic hypothesis were key stepping stones in the development of nonequilibrium thermodynamics and continue to be the focus of active research in various fields of natural sciences. The latter provides a basis for experimenters to determine the physical behavior of a system by time averaging a small number of trajectories and regarding the result thus obtained as representative of the system's ensemble average [1–3]. Regarding the former, two decades of research on anomalous diffusion [4] led to the conclusion that there exists a great diversity of diffusive processes. Both concepts make use of the notion of the velocity correlation function (VCF), which hints at a conceptual connection between the two. In general, the asymptotic VCF of a system can be formulated as $C_v(t, t_w) \simeq C_0 t_w^{-b} \phi[(t - \epsilon t_w)/t_w^\beta]$, where C_0 is a constant, $b \in [0, 2)$, ϵ and β are 0 or 1, and ϕ denotes an arbitrary scaling function of the age time, t , and the waiting time, t_w [5–8]. Such a VCF is often invoked to characterize the slow time changes of many aging systems [9], a recurrent mechanism in nonequilibrium statistical physics. We note that the time-scaling behavior of the VCF of a Brownian particle remains a topic of significant current interest [10]. For instance, in

bulk liquids, the VCF does not exhibit an exponential decay but instead follows an algebraic decay of the form $t^{-3/2}$ in the hydrodynamic long-time limit [11]. Similarly, for a near-boundary Brownian particle confined by optical tweezers, the VCF has been reported to display a long-time tail decaying as $t^{-5/2}$ [12].

A challenging question in this context is how to determine whether a system has entered an effective ergodic regime, so that accurate steady-state measurements can be performed in a conveniently short time interval. Recently, the single-particle tracking (SPT) technique has been proposed to investigate time-averaged mean square displacements (MSDs) [13–18] under the assumption that they asymptotically coincide with the ensemble averaged MSDs in the corresponding stochastic description [19]. The diffusion constants thus extracted are consistent with the ensemble theory only under overdamped conditions or for large lag times [20]. Unfortunately, this raises the dilemma of large observation samples versus poor statistics and fails altogether in the case of bounded system [21]. Another key question is how VCFs from real experiments or numerical simulations are related with physical parameters such as diffusion and friction constants. Indeed, the standard Green-Kubo relation only applies to normal diffusion, while its generalizations [6,22] yield the fractional diffusion constant rather than the dynamical friction with physical dimension.

With this Letter, we address the aforementioned questions in terms of an ergodic criterion, earlier introduced to characterize the transition from supercooled liquids to glasses [23]. By finding a universal law below a threshold for the convergence of time-averaged observables, we

*Contact author: jdbao@bnu.edu.cn

†Contact author: fabio.marchesoni@pg.infn.it

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reexamine the relationship between diffusion and ergodicity. To demonstrate the validity of our approach, we apply our theory to three systems of experimental and theoretical relevance.

Kinetic energy Green-Kubo-like relation—To emphasize the ergodic nature of the stochastic observable $A(t) = A[x(t), v(t)]$, we use its fluctuation metric in the phase space (x, v) ,

$$\Omega_A(t) = \langle \bar{A}^2 \rangle - \langle \bar{A} \rangle^2 = \frac{1}{t^2} \int_0^t \int_0^t dt_1 dt_2 C_A(t_1, t_2), \quad (1)$$

where the finite-time average, $\bar{A} = t^{-1} \int_0^t A(t') dt'$, is itself a random observable and $C_A(t_1, t_2) = \langle A(t_1)A(t_2) \rangle - \langle A(t_1) \rangle \langle A(t_2) \rangle$. If for $t \rightarrow \infty$, $\Omega_A(t) \rightarrow 0$, its probability density, $W(\bar{A})$, with the variance of Eq. (1), converges to $\delta(\bar{A} - \langle \bar{A} \rangle)$. This means that a necessary ergodicity condition for an observable A is that its asymptotic time average tends to a deterministic value [24,25]. We first investigate the result of ergodic transition by considering $A = v^2$ in the stationary case.

Let us consider a stationary VCF with $b = 0$, $\epsilon = 1$, and $\beta = 0$, that is,

$$C_v(t, t_w) = C_0 \phi(t - t_w) + C_1 \phi(t) \phi(t_w), \quad (2)$$

where $C_0 = k_B T/m$, k_B is the Boltzmann constant, T denotes the temperature, $C_1 = \{v^2(0)\} - C_0$, and $\{\dots\}$ denotes averaging over the initial velocity, $v(0)$ [26]. Such a VCF can be obtained from the generalized Langevin equation (GLE), $m\dot{v} + m \int_0^t M_1(t-t') v(t') dt' + \partial_x U(x) = \xi(t)$, with non-Ohmic memory function $M_1(t-t') = (k_B T)^{-1} \langle \xi(t) \xi(t') \rangle$ [27–29]. In the absence of a potential, $U(x) \equiv 0$, $\phi(t)$ is the inverse Laplace transform of $[s + \hat{M}_1(s)]^{-1}$, so that the VCF can be expressed in terms of a Mittag-Leffler function [30,31], $\phi(t) = E_{2-\delta}[-(\omega_\delta t)^{2-\delta}]$, where $\omega_\delta = [\gamma_\delta \omega^{1-\delta} \sin^{-1}(\delta\pi/2)]^{1/(2-\delta)}$ and δ , with $0 < \delta < 2$, is the exponent of the power-law decaying noise spectral density. The VCF is then finite and decays at large t with power-law tail, $\sim t^{\delta-2}$ ($\delta \neq 1$). In turn, the corresponding MSD diverges asymptotically like $\langle x^2(t) \rangle \sim t^\alpha$, with diffusion exponent $\alpha = \delta$.

At large times, or when the initial velocity is prepared at thermal equilibrium, the last term of Eq. (2) vanishes, so that VCF is invariant under time translation, i.e., the process is stationary. The fluctuation metric, Eq. (1), of v^2 then reads

$$\begin{aligned} \Omega_{v^2}(t) &= \frac{4}{t^2} \int_0^t (t-z) C_v^2(z) dz \sim \frac{4}{t} \int_0^\infty C_v^2(z) dz \\ &\sim \frac{4}{t} \left(\int_0^L E_{2-\delta}^2[-(\omega_\delta z)^{2-\delta}] dz + \int_L^\infty (\omega_\delta z)^{2\delta-4} dz \right), \end{aligned} \quad (3)$$

where L is a large cutoff introduced to regularize possible singularities. However, the first integral between round parentheses is always nonzero and finite, while the second integral is so for $\delta = \alpha < \alpha_c$ with $\alpha_c = \frac{3}{2}$.

Now, the question raises naturally whether the ergodic convergent behavior reported for v^2 is universal under the condition $\alpha < \alpha_c$. For stationary processes we can answer affirmatively. Let us define the following dimensionless ratio:

$$F_{v^2}(t) = \Omega_{v^2}(0)/\Omega_{v^2}(t) \rightarrow R_{v^2} t \quad (0 < \alpha < 3/2), \quad (4)$$

where $\Omega_{v^2}(0) = 2\{v^2(0)\}^2 = 2(k_B T/m)^2$ and R_{v^2} represents a measure of the rate at which the observable v^2 explores the configuration space. As consequence, after a finite relaxation time, systems with $\alpha < \alpha_c$ eventually enter an effective ergodic regime, characterized by a universal $F_{v^2}(t)$. This remark is of help when analyzing the experimental data of systems subject to long-time transient, as is the case of many subdiffusive processes occurring in crowded and inhomogeneous media [32,33]. The square velocity metric, Ω_{v^2} , considered so far, is equivalent to the kinetic energy (KE) metric, Ω_{KE} . Indeed, since $\Omega_{KE} = (m^2/4)\Omega_{v^2}$, both exhibit the same universal law (4). For $\alpha < \alpha_c$, a Green-Kubo-like relation expresses the relaxation time of the kinetic energy in terms of the time integral of the square stationary VCF,

$$D_{KE}^{-1}(t) = 2 \left(\frac{k_B T}{m} \right)^{-2} \int_0^t \langle v(t') v(0) \rangle^2 dt'. \quad (5)$$

Note that this equation also provides an estimate of the shortest ergodic time, $t_{\text{erg}} = R_{v^2}^{-1} = D_{KE, \text{st}}^{-1}$, where $D_{KE, \text{st}} = \lim_{t \rightarrow \infty} D_{KE}(t)$ is the diffusive coefficient of the kinetic energy related with the dynamical friction γ_{dyn} [23].

In Fig. 1, we plot the asymptotic kinetic energy diffusion constant, $D_{KE, \text{st}}$, for the zero-force GLE with non-Ohmic memory friction. One notices immediately the existence of a threshold for the diffusion exponent, $\alpha_c = \frac{3}{2}$, below which ergodicity sets in and the kinetic energy diffusion constant, in units of ω_δ , increases with decreasing δ (i.e., α). Consequently, the slower the spatial diffusion, the faster the system approaches to the effective ergodic regime. Moreover, when applied to bounded systems, Eq. (5) can yield finite values for the kinetic energy diffusion constant

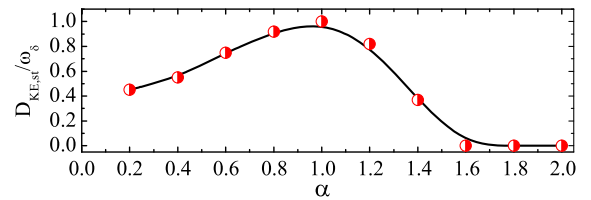


FIG. 1. Kinetic energy diffusion constant, $D_{KE, \text{st}}$, of Eq. (5) vs the diffusion exponent, α , for the GLE with non-Ohmic memory function that is reported in the text.

because of the faster convergence of VCF, whereas according to the standard Green-Kubo formula, the spatial diffusion constant would vanish.

We explain now why we chose the square velocity to define a working metric, rather than the velocity itself. To this purpose, let us consider the normal diffusion (with $\alpha = 1$), described by a stationary GLE driven by the Ornstein-Uhlenbeck noise with exponential memory function, $M_1(t) = \gamma_0 \tau_c^{-1} \exp(-t/\tau_c)$. The corresponding kinetic energy diffusion constant reads $D_{\text{KE,st}} = \gamma_0 / (1 + \gamma_0 \tau_c)$, in the white noise limit, $\tau_c \rightarrow 0$, $D_{\text{KE,st}} \rightarrow \gamma_0$, and, in the opposite case $\tau_c \rightarrow \infty$, $t_{\text{erg}} \rightarrow \infty$, the solution of GLE is $v(t) = v(0)$, thus the ergodicity is broken. However, for anomalous diffusion processes, the standard Green-Kubo formula would be less predictive because it would yield $D_x = \int_0^\infty \langle v(t)v(0) \rangle dt = 0$ for $\alpha < 1$ and $D_x \rightarrow \infty$ for $\alpha > 1$.

Protein folding—As a first application, we consider the protein folding. The evidence for the existence of pico-second time-scaling relaxation processes in protein systems has been obtained by dielectric absorption measurements at the mm-wave frequencies [34,35]. However, simple Markovian models do not accurately describe the peptide kinetics and, in particular, cannot simultaneously reproduce the folding and unfolding kinetics. Vice versa, GLE-based formulations show that memory effects in the frictional mechanisms significantly speed up peptide folding and unfolding kinetics [34,35]. One of the most direct experimental probes of protein friction is quasielastic neutron scattering [23,35]. In Ref. [35], a best fit of the experimental data sets the protein frictional damping at 0.90 ps. Here, we specialize the GLE introduced above to model the protein dynamics and make use of the kinetic energy metric to extract the relevant relaxation time. We assume that the potential energy surface explored by the protein atoms is the harmonic one and the simplest choice for the memory function $M_1(t)$ is again an exponential decaying function with time constant τ_c .

We illustrate in Fig. 2 our results for the dynamical friction constant of the system described by a GLE with the harmonic-potential $U(x) = \frac{1}{2} m \omega_0^2 x^2$ and an Ornstein-Uhlenbeck noise. In the Markovian approximation ($\tau_c \rightarrow 0$), the kinetic energy diffusivity approach of Ref. [23] predicted $\gamma_{\text{dyn}} = 1.87 \text{ ps}^{-1}$, that is, about twice the experimental value. Instead, based on the kinetic energy Green-Kubo relation of Eq. (5), we determine the noise correlation time value, $\tau_c = 0.46 \text{ ps}$, which best reproduces the experimental result, $\gamma_{\text{dyn}} = 0.90 \text{ ps}^{-1}$. Therefore, non-Markovian memory effects may well explain the slower-than-expected relaxation in protein folding [34,36].

Short-lag SPT measurement—As a second example, we address a key issue in SPT microscopy, a standard tool for probing tracer motion, particularly within living cells. Despite its utility, all experimental measurements are limited to finite observation times, regardless of the

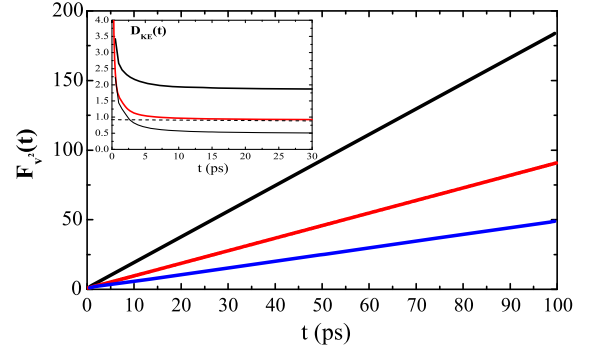


FIG. 2. $F_{v^2}(t)$ vs t with $\tau_c = 0.0, 0.46, 1.0 \text{ ps}$ from top to bottom. Inset: D_{KE} vs t is compared with the experimental data (dashed curve) [23,35]. The parameters used here are $\gamma_0 = 1.87 \text{ ps}^{-1}$ and $\omega_0 = 1.0 \text{ ps}^{-1}$.

system's lifetime. We focus here on the challenge of extracting meaningful physical parameters, such as diffusion coefficients or relaxation times, from time-averaged MSDs. This is especially critical for systems exhibiting weak ergodicity breaking or bounded dynamics, where traditional approaches often fail. The issue remains controversial, as the same phenomenon has been described by vastly different models [37–39]. Let us consider the observable $\bar{\delta}^2(\Delta, T_m) = (T_m - \Delta)^{-1} \times \int_0^{T_m - \Delta} [x(t + \Delta) - x(t)]^2 dt$, where T_m is the measurement time and Δ the lag time, which requires $\Delta \ll T_m$ [19,40]. To date, most models have revealed that the time-averaged MSD defined as $\bar{\delta}^2(\Delta, T_m)$ differs from the usual ensemble-averaged MSD, and only in a few cases, e.g., in the fractional Langevin equation representation, the two MSDs coincide asymptotically [19]. The former models imply what is termed weak ergodicity breaking, while the latter ones exhibit ergodicity in MSD sense. Nevertheless, for the ergodic system bounded by a harmonic potential, the two are not equal (see Supplemental Material [41] for details). This confirms that the information obtained from the time-averaged and the ensemble-averaged MSDs can differ.

To extract sound physical parameters for nonergodic process of this kind, in Sec. A of Supplemental Material (SM) [41] we analyze a VCF-based, time-averaged Green-Kubo-like relation, which for large- T_m reads

$$D_{\delta^2}^{-1}(\Delta, T_m) = 2 \langle [x(\Delta) - x(0)]^2 \rangle^{-2} \int_0^{T_m - \Delta} \langle [x(T_m) - x(T_m - \Delta)][x(t + \Delta) - x(t)]^2 \rangle dt. \quad (6)$$

To test the applicability of this approach, we consider the standard Brownian motion with the VCF $\langle v(t)v(0) \rangle = (k_B T / m) \exp(-\gamma_0 t)$, for which

$$D_{\delta^2}^{-1}(\Delta, T_m) = \frac{1}{\gamma_0} (1 - \exp[-2\gamma_0(T_m - \Delta)]), \quad (7)$$

certainly a correct result for large enough measurement times. In contrast, the time-averaged diffusion constant, $D_\delta = (k_B T / m \gamma_0) [1 - \exp(-\gamma_0 \Delta)]$, holds good only for large lag times [20]. The former measurement of the diffusion constant is experimentally viable, while the latter is not. Hence the convenience of replacing the nonergodic MSD metric by the ergodic kinetic-energy-like metric,

$$\overline{\delta^2} = \Delta^{-2} \overline{\delta^2}, \quad D_{\text{KE}} = \lim_{\Delta \rightarrow 0} D_{\delta^2}(\Delta, T_m). \quad (8)$$

It is thus demonstrated that the present approach, which uses short lag-time measurements, offers a statistical advantage over the original SPT method, as the latter requires large lag-time values to accurately extract the diffusion coefficient.

Aging velocity correlation function—Finally, we consider the nonstationary VCF with $b = 2 - \alpha$, $\epsilon = 1$, and $\beta = 1$, recently proposed as a paradigm for theoretical studies [6]. In the superdiffusive regime, the scaling function reads $\phi(s) = c_1 s^{-\delta_1}$ with $2 - \alpha \leq \delta_1 < 1$ and $s = (t - t_1)/t_1$ small. In the limit of large t , the fluctuation metric of v^2 becomes

$$\begin{aligned} \Omega_{v^2}(t) &= \frac{4}{t^2} \int_0^t dt_2 \int_0^{t_2} dt_1 C_v^2(t_2, t_1) \\ &\sim \frac{4}{t} \lim_{t \rightarrow \infty} \left(\int_0^L C_v^2(t, t_1) dt_1 \right. \\ &\quad \left. + c_1^2 \int_L^t t_1^{2\alpha-4+2\delta_1} t^{-2\delta_1} \left[1 + 2\delta_1 \frac{t_1}{t} \right] dt_1 \right). \end{aligned} \quad (9)$$

Since $C_v(0, 0) = \{v^2(0)\}$ is finite and $C_v(t \rightarrow \infty, t_1) \rightarrow 0$, the first single integral does not diverge, while the second one is finite for $2\alpha - 3 < 0$, i.e., $\alpha < \frac{3}{2}$. Hence, the exponent $\alpha_c = \frac{3}{2}$ appears to mark the transition onset also for this class of nonstationary processes. Although similar ideas have been proposed for various models in relation to ergodicity breaking [18,41–44], this concept remains not yet fully consolidated.

Therefore, as a third example, we discuss granular gases as a case study to explore the minimization of physical aging in terms of the kinetic-energy fluctuation metric. In low density granular systems [45], collisions are quantified by the restitution coefficient, $\kappa = |v'_{12}/v_{12}| < 1$, that is the ratio of the relative speed of two colliding particles before and after a collision. The relevant system's physical quantities, for instance the velocity relaxation time and the diffusion coefficient, are functions of time. Haff's law postulates that the temperature of free granular gas decays with time [46], so that granular particle diffusion occurs in a cooling environment. This provides the conditions for aging correlations, which we obtain from the inhomogeneous Langevin equation [47], $m\dot{v} = -mg^2(t)v + g(t)\xi(t)$, where $g(t)$ governs the friction function and $\langle \xi(t)\xi(t') \rangle = 2mk_B T \delta(t - t')$. Accordingly, one can choose $g^2(t) = \gamma_0/(1 + t/t_s)$ and $T = T_0/(1 + t/t_s)^b$, where γ_0 is the static friction constant, t_s a timescale, and T_0 a constant reference temperature [48,49].

Such a Markovian dynamics can be solved exactly. Assuming $\langle v(0)\xi(t) \rangle = 0$ and $t \geq t_w$, we obtain the VCF,

$$\begin{aligned} \{\langle v(t)v(t_w) \rangle\} &= C_0(1 + t_w/t_s)^{-b} \left(\frac{1 + t/t_s}{1 + t_w/t_s} \right)^{-\lambda} \\ &\quad + C_1[(1 + t_w/t_s)(1 + t/t_s)]^{-\lambda}, \end{aligned} \quad (10)$$

where $\lambda = \gamma_0 t_s$, $C_0 = 2k_B T_0 \lambda / [m(2\lambda - b)]$, and $C_1 = \{v^2(0)\} - C_0$. When $t \gg t_s$, the VCF of Eq. (10) exhibits the asymptotic aging behavior, $C_v(t, t_w)$, mentioned in the introduction, with $\epsilon = 0$ and $\beta = 1$. Clearly, the scaling exponent, b , is an effect of the environment cooling and, in contrast with superaging [6], here the prefactor decays with the waiting time, t_w . The corresponding MSD yields $\langle x^2(t) \rangle \sim t^\alpha$ with $\alpha = 2 - b$, while the metric of Eq. (1) for $A = v^2$ reads

$$\begin{aligned} \Omega_{v^2}(t) &= \frac{4C_0^2}{(2\lambda - 2b + 1)} \frac{1}{(t/t_s)^2} \left(\frac{1}{2 - 2b} [(1 + t/t_s)^{2-2b} - 1] - \frac{1}{1 - 2\lambda} [(1 + t/t_s)^{1-2\lambda} - 1] \right) \\ &\quad + \frac{4C_1^2}{(1 - 2\lambda)^2 (t/t_s)^2} \left(\frac{1}{2} [(1 + t/t_s)^{2-4\lambda} - 1] - [(1 + t/t_s)^{1-2\lambda} - 1] \right) \\ &\quad + \frac{8C_0 C_1}{(1 - b)(t/t_s)^2} \left(\frac{1}{2 - 2\lambda - b} [(1 + t/t_s)^{2-2\lambda-b} - 1] - \frac{1}{1 - 2\lambda} [(1 + t/t_s)^{1-2\lambda} - 1] \right). \end{aligned} \quad (11)$$

For $\alpha = 3/2$, $\Omega_{v^2}(t)$ decays inversely proportional to time because the leading asymptotic contribution comes from the first term in the rhs of Eq. (11). For $\alpha > 3/2$ ($\alpha < 3/2$), instead, $\Omega_{v^2}(t)$ decays slower (faster) than $1/t$. Thus, here, as above, $\alpha_c = 3/2$ plays the role of a critical exponent. In

the limit $t_s \rightarrow \infty$, the standard Brownian motion is recovered, i.e., $\Omega_{v^2}(t) \rightarrow 2(k_B T_0 / m)^2 \gamma_0^{-1} / t$.

Figure 3 shows the fluctuation metric of the square velocity for increasing $\alpha = 2 - b$. One expects from Eq. (11) that the decay of Ω_{v^2} grows faster with increasing

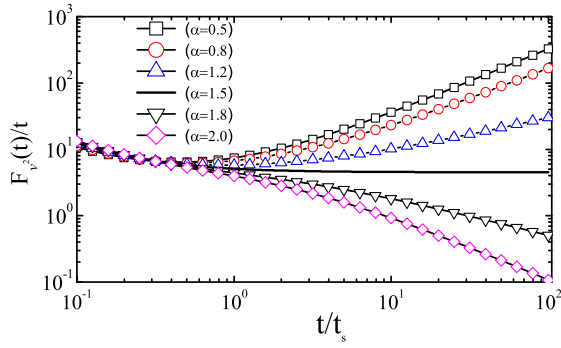


FIG. 3. Granular gas model. $F_v(t)/t$ vs t for different diffusive exponents, α , computed via Eqs. (4) and (11) for $\lambda = 5.0$.

b , because the larger b , the faster the temperature drops. A surprising finding is that in the ballistic diffusion situation with $\alpha = 2$ (or $b = 0$), the VCF of Eq. (10) vanishes but $\Omega_{v^2}(t \rightarrow \infty) \neq 0$. This is in contrast with previous results for nonergodic ballistic diffusion [26,28,50], where VCF was shown to approach a constant. In this model, when the potential is absence, the lower the temperature and larger the friction, the faster the approach to metric convergence. Also remarkable is the observation that, for $b = 1$, the MSD grows linearly in time and yet is anomalous because of the decay of the related VCF is nonexponential.

In conclusion, we established a connection between diffusion and ergodicity through scaling velocity correlation functions for both stationary and aging processes. In particular, for stationary process characterized by velocity correlation functions invariant under time translation and diffusive exponent below $\alpha_c = 3/2$, we proposed a kinetic energy Green-Kubo-like relation, which relates the dynamical friction acting on the system with the integral of the squared velocity correlation function. This result was proven in terms of the fluctuation metric of the time-averaged square velocity. The emerging nonequilibrium picture is well captured by the catchall “slow diffusion—fast ergodicity; fast aging—slow diffusion.”

We illustrated the practical advantages of the present approach with three distinct examples. First, we calculated the relaxation timescale of protein folding, in close agreement with experiments, to illustrate the importance of non-Markovian effects. Then, we had recourse to our kinetic energy metric to solve the problem of the inconsistencies between the diffusion coefficient estimated from single-particle tracking and the ensemble theory. Finally, the Langevin equation with friction and temperature varying in time according to a power law, as customary in granular matter, was proposed as a simple model to study aging velocity correlation functions at work. We are confident that our results will find application elsewhere, too.

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Data availability—No data were created or analyzed in this study.

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