Dynamics of a Tagged Monomer: Effects of Elastic Pinning and Harmonic Absorption

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We study the dynamics of a tagged monomer of a Rouse polymer for different initial configurations. In the case of free evolution, the monomer displays subdiffusive behavior with strong memory of the initial state. In the presence of either elastic pinning or harmonic absorption, we show that the steady state is independent of the initial condition that, however, strongly affects the transient regime, resulting in nonmonotonic behavior and power-law relaxation with varying exponents.

It is known that the dynamics of a mesoscopic particle embedded in a viscous fluid is Markovian and well described by the Brownian motion. The particle mean-squared displacement (MSD) grows diffusively in time as $2Dt$, where D is the diffusion coefficient. However, in a crowded environment of interacting particles, the single particle may display anomalous diffusion. Let us consider a long Rouse polymer composed of L monomers connected to their nearest neighbors by harmonic springs of constants Γ and immersed in a good solvent. Its global dynamics is Markovian, and the center of mass diffuses with MSD behaving as $2(D/L)t$. However, the dynamics of a single tagged monomer is non-Markovian, with the MSD subdiffusing as $\sqrt{2/(\pi\Gamma)}Db_0\sqrt{t}$ for times $t \ll L^2/\Gamma$ [1]. Here, b_0 encodes the memory of the polymer configuration at $t = 0$. In particular, if the polymer at $t = 0$ is in equilibrium with the solvent, the dynamics of the tagged monomer is well described [2–4] by a fractional Brownian motion (FBM), which generalizes the Brownian motion to the case of nonindependent Gaussian increments [5,6]. On the other hand, if the polymer at $t = 0$ is out of equilibrium, the dynamics displays *aging*, in that the increments are not only correlated (as in FBM) but also drawn from a Gaussian distribution with a time-dependent variance. These non-Markovian processes are relevant for many biological phenomena, such as the unzipping of DNA [7], translocation of polymers through nanopores [8–11], subdiffusion of macromolecules inside cells [12–15], and single-file diffusion [16].

In the above applications, often the tagged particle is subject to either pinning by an elastic spring or absorption. The first case, e.g., corresponds to employing optical tweezers to confine specific molecules in order to contrast their dynamical behavior inside the crowded environment of a cell with that outside [17]. The second situation arises when a reactant attached to a single monomer encounters an external reactive site fixed in space [18,19]. Moreover, in the problems of polymer translocation and DNA unzipping, the time to translocate or unzip corresponds to the absorbing time of a one-dimensional subdiffusive Gaussian process inside a finite interval with absorbing boundaries.

In general, these problems are investigated numerically either by molecular dynamics simulations or by simulation of the underlying Gaussian process [20,21]. Recently, it has been shown that subdiffusive Gaussian dynamics can be studied by the fractional Langevin equation [16,22,23]. This approach has been fruitfully used in the presence of elastic pinning [24–26] but cannot easily incorporate absorption.

In this Letter, we propose a general analytical framework to compute relevant quantities such as the MSD and the absorbing time distribution of the tagged monomer, for the case of elastic pinning and harmonic absorption. These problems are relevant for practical applications: the pinning by optical tweezers is indeed elastic, whereas harmonic absorption mimics well a finite interval with absorbing boundaries. Our approach naturally incorporates the initial condition of the system. In the following, we specifically consider a one-dimensional Rouse chain and mention higher dimensions in the conclusions. Our main results, summarized in Table I, show that whereas the steady state is independent of the initial condition, the transient behavior exhibits very strong memory effects: (i) If a quench in temperature is performed at $t = 0$, the MSD displays a bump in time and converges to the steady-state value as a power law. This behavior, predicted for both pinning and absorption, could be observed in experiments. (ii) For harmonic absorption, the absorption time distribution decays exponentially with a characteristic time
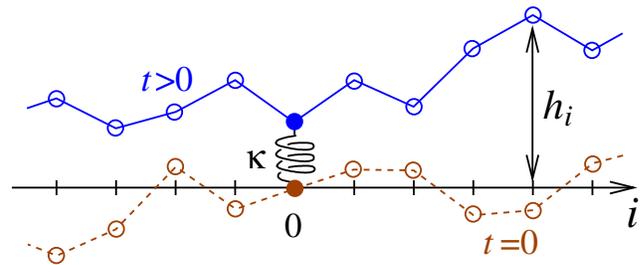

FIG. 1 (color online).   Schematic of an EW interface pinned by a harmonic spring acting on the tagged monomer at $i = 0$. The initial configuration h^0 (dashed line) has $h_0^0 = 0$.

that is independent of the initial condition. Hence, we expect the translocation or the unzipping time to have a distribution with exponential tails, independent of the initial condition of the system.

The Rouse chain is equivalent to the one-dimensional discrete Edwards-Wilkinson (EW) interface shown in Fig. 1 [27,28]. Here, $h_i(t)$ is the displacement of the ith monomer at time t with respect to the origin. The elastic energy of the system is $E_{el} = (\Gamma/2)\sum_i (h_{i+1} - h_i)^2$, where Γ is set to unity below. Additionally, the monomers are subjected to friction (set to unity) in an overdamped regime. The dynamics of the interface is described by a set of L coupled Langevin equations

$$\frac{\partial h_i(t)}{\partial t} = -\frac{\partial E_{el}}{\partial h_i} + \eta_i(t) = \sum_j \Delta_{ij} h_j(t) + \eta_i(t), \quad (1)$$

where Δ denotes the discrete Laplacian matrix, $\{\eta_i(t)\}$ are independent Gaussian white noises $\langle \eta_i(t) \rangle = 0$, $\langle \eta_i(t)\eta_j(t') \rangle = 2T\delta_{i,j}\delta(t - t')$, with the temperature T set to unity below, and $\langle \cdots \rangle$ denotes thermal averaging.

Elastic pinning.—We consider the situation where the "tagged" monomer at $i = 0$ is pinned around the origin by an additional elastic force (Fig. 1). This is described by adding the term $\kappa h_0^2/2$ to the energy. In this case, the Langevin equations are similar to Eq. (1) with Δ_{ij} substituted by $-\Lambda_{ij} = \Delta_{ij} - \kappa\delta_{i,j}\delta_{i,0}$ and can be solved (cf. the Supplemental Material [29]). In order to adopt a unified formalism to deal with both pinning and absorption, we follow a Fokker-Planck approach. Let $\mathcal{W}_t[h|h^0]$ be the probability density to observe the interface in the configuration h at time t, given that the configuration at time $t = 0$ was h^0, where h (respectively, h^0) denotes the vector $\{h_i\}$ (respectively, $\{h_i^0\}$). It obeys the Fokker-Planck equation (FPE)

$$\frac{\partial \mathcal{W}_t[h|h^0]}{\partial t} = \left[\sum_i \frac{\partial^2}{\partial h_i^2} + \sum_{i,j} \frac{\partial}{\partial h_i}\Lambda_{ij}h_j\right]\mathcal{W}_t[h|h^0], \quad (2)$$

which is an L-dimensional generalization of the FPE for the one-dimensional Ornstein-Uhlenbeck process [30,31]. Equation (2) can be solved through a mapping onto the imaginary time Schrödinger equation for L coupled quantum harmonic oscillators (see the Supplemental Material [29])

$$\mathcal{W}_t[h|h^0] = \sqrt{\det\left(\frac{\Lambda}{2\pi(1 - e^{-2\Lambda t})}\right)}$$
$$\times \exp\left[-\frac{1}{2}(h - e^{-\Lambda t}h^0)^T \frac{\Lambda}{1 - e^{-2\Lambda t}}(h - e^{-\Lambda t}h^0)\right], \quad (3)$$

where the superscript T denotes transpose operation. Note that replacing the matrix Λ in Eq. (3) by the spring constant λ, we recover the well-known Ornstein-Uhlenbeck result for the dynamics of a particle submitted to a harmonic force.

Since Eq. (3) has a Gaussian form, all statistical information about the dynamics of the tagged monomer are encoded in the first two moments of $\mathcal{W}_t[h|h^0]$, which are conveniently obtained by introducing the local field $b = \{b_i\}$ acting on individual monomers. We consider the generating function

$$\mathcal{G}_t[b] = \int \prod_i dh_i e^{\sum_i b_i h_i} \mathcal{W}_t[h|h^0]. \quad (4)$$

Using Eq. (3) in Eq. (4), changing variables $h \to h - e^{-\Lambda t}h^0$, and doing the Gaussian integration, we get

$$\mathcal{G}_t[b] = \exp\left[\frac{1}{2}b^T \frac{1 - e^{-2\Lambda t}}{\Lambda}b + b^T e^{-\Lambda t}h_0\right]. \quad (5)$$

Note that $\mathcal{G}_t[0] = 1$ represents the normalization of $\mathcal{W}_t[h|h^0]$. The connected correlation functions are obtained by differentiation of $\mathcal{F}_t[b] = \ln\mathcal{G}_t[b]$. In particular, using $\langle h_i(t) \rangle = \partial \mathcal{F}_t[b]/\partial b_i|_{b=0}$ and $\langle h_i(t)h_j(t) \rangle_c = \partial^2 \mathcal{F}_t[b]/\partial b_i \partial b_j|_{b=0}$, we get

$$\langle h_i(t) \rangle = (e^{-\Lambda t}h^0)_i, \quad \langle h_i(t)h_j(t) \rangle_c = \left(\frac{1 - e^{-2\Lambda t}}{\Lambda}\right)_{ij}. \quad (6)$$

TABLE I. Summary of our results for the MSD and the survival probability: single particle versus tagged monomer of an infinite Rouse chain. We prepare the chain in equilibrium at temperature T_0, and the overbars denote the average over the ensemble of initial configurations. At time $t = 0$, the system is quenched to temperature $T = 1$ and let to evolve following three protocols, namely, (i) free evolution, (ii) elastic pinning acting on the tagged monomer, and (iii) harmonic absorption acting on the tagged monomer. The friction constant Γ and D are both set to unity.

	Free evolution	Elastic pinning, long time $t \to \infty$ behavior	Harmonic absorption, long time $t \to \infty$ behavior
Single particle	$\langle h_0^2(t) \rangle = 2t$	$\langle h_0^2(t) \rangle = \frac{1}{\kappa}(1 - e^{-2\kappa t})$	$\langle h_0^2(t) \rangle = 2\mu^{-1/2}\tanh(t/\sqrt{\mu})$
			$S(t) \sim \exp(-2\mu^{1/2}t)$
	Brownian process		
Tagged monomer	$\overline{\langle h_0^2(t) \rangle} = \sqrt{\frac{2}{\pi}}b_0\sqrt{t}$	$\overline{\langle h_0^2(t) \rangle} = \frac{1}{\kappa} + \frac{c_0}{\kappa^2\sqrt{t}} + \cdots$	$\overline{\langle h_0^2(t) \rangle}^{abs} = a_0\mu^{-1/3} + O(1/t)$
$T_0 \neq 1$	$b_0 = 1 + T_0(\sqrt{2} - 1)$	$c_0 = \sqrt{2/\pi}(T_0 - 1)$	$S(t) \sim \exp(-a_0\mu^{2/3}t)$
	Aging process		
Tagged monomer	$\overline{\langle h_0^2(t) \rangle} = \frac{2}{\sqrt{\pi}}\sqrt{t}$	$\overline{\langle h_0^2(t) \rangle} = \frac{1}{\kappa} + \frac{c_1}{\kappa^3 t} + \cdots$	$\overline{\langle h_0^2(t) \rangle}^{abs} = a_0\mu^{-1/2} + O(1/t)$
$T_0 = 1$	FBM process	$c_1 \approx 0.0711$	$S(t) \sim \exp(-a_0\mu^{2/3}t)$

At long times, we expect from the equipartition theorem that $\langle h_0^2(t \to \infty) \rangle = 1/\kappa$, independent of the number of monomers in the polymer. In the case of a single particle, the steady-state value is reached exponentially fast in time (Table I). For a long polymer, the analysis of Eq. (6) shows that the steady-state value is reached with a power-law decay where the exponent depends on the initial configuration. In particular, we study an initial configuration h^0 randomly sampled from the ensemble of configurations equilibrated at temperature T_0 and conditioned on $h_0^0 = 0$. At equilibrium, the displacements h_i^0's are Gaussian distributed as $p_{eq}(h^0) = \exp[-\frac{1}{2}(h^0)^T \sigma^{-1} h^0]/\sqrt{\det(2\pi\sigma)}$, where $\sigma_{ij} = \overline{h_i^0 h_j^0}$ is the covariance matrix, with the overbar denoting averaging with respect to $p_{eq}(h^0)$. In the limit $L \to \infty$, the equilibrated EW interface corresponds to two Brownian trajectories starting at 0 with diffusion constant equal to $T_0/2$. The covariance then reads $\sigma_{ij} = T_0 \theta_H(ij) \min(|i|, |j|)$, where $\theta_H(x)$ is the Heaviside function. On the other hand, for a finite interface with periodic boundary conditions, we have $\sigma_{ij} = T_0[\min(i, j) - ij/L]$, where $i, j \in \{0, \ldots, L-1\}$. The computation of $\langle h_0^2(t) \rangle$ for long times can be performed analytically in the limit $L \to \infty$. The details are given in the Supplemental Material [29]. We get

$$\overline{\langle h_0^2(t) \rangle} \simeq \frac{1}{\kappa}\left[1 + \frac{T_0 - 1}{\kappa}\sqrt{\frac{2}{\pi t}} - \frac{T_0 c_1}{\kappa^2 t} + \cdots\right], \quad (7)$$

where $c_1 = 0.0711 \ldots$. We thus see that the MSD tends to the steady-state value $1/\kappa$ as $1/\sqrt{t}$ if T_0 is different from unity. For $T_0 = 1$, which corresponds to the temperature of the noise for $t > 0$, the relaxation to steady state is as $1/t$. Moreover, for $T_0 > 1$, the MSD has a nonmonotonous behavior in time with a bump. This behavior may be understood as the effect of the large initial *spatial* fluctuations of the polymer for $T_0 > 1$ that propagate towards the tagged monomer and increase its *temporal* fluctuations in the transient regime. Note that the calculation in Refs. [24–26] applies to polymers equilibrated with the solvent, whereas here we study the effects of different initial conditions.

Harmonic absorption.—The FPE is

$$\frac{\partial \mathcal{W}_t[h|h^0]}{\partial t} = \left[\sum_i \frac{\partial^2}{\partial h_i^2} - \sum_{i,j}\left(\frac{\partial}{\partial h_i}\Delta_{ij}h_j + h_i A_{ij}h_j\right)\right]\mathcal{W}_t[h|h^0],$$

$$\quad (8)$$

where the positive definite matrix A describing absorption is $A_{ij} = \mu\delta_{i,j}\delta_{i,0}$, with $\mu > 0$ being the absorption rate. Since the absorption probability increases quadratically with distance, the FPE (8) can be solved using the mapping to a system of coupled quantum harmonic oscillators (see details in the Supplemental Material [29]). We obtain

$$\mathcal{G}_t[b] = \mathcal{G}_t[0]\exp[b^T\Omega_t^{-1}b + b^T\Omega_t^{-1}Y_t h^0], \quad (9)$$

$$\mathcal{G}_t[0] = \sqrt{\det(e^{-t\Delta}Y_t\Omega_t^{-1})}\exp\left[-\frac{1}{2}(h^0)^T Q_t h^0\right], \quad (10)$$

where we have introduced the four symmetric matrices

$$K = \sqrt{\Delta^2 + 4A}, \qquad \Omega_t = K\coth(Kt) - \Delta,$$

$$Y_t = K/\sinh(Kt), \qquad Q_t = (\Omega_t + 2\Delta - Y_t\Omega_t^{-1}Y_t)/2.$$

In the presence of absorption, $\mathcal{W}_t[h|h^0]$ is not normalized to unity, and $\mathcal{G}_t[0]$ is the survival probability $S(t)$, namely, the probability that an initial configuration h^0 has not been absorbed up to time t [32,33]. Note that the survival probability is the cumulative of the absorbing time distribution. In the long time limit, we have $\Omega_t \approx K - \Delta$ and $Y_t \sim \exp(-Kt)$ so that the survival probability asymptotically decays as $S(t) \sim \sqrt{\det(e^{-(K+\Delta)t})}$. Using $\det[\exp(A)] = \exp(\text{Tr}[A])$, we get

$$S(t) \underset{t\to\infty}{\sim} \exp[-t\text{Tr}\{K + \Delta\}/2]. \quad (11)$$

Note that the decay rate is independent of h^0.

Alternatively, one can obtain an exact expression for $S(t)$ in terms of the tagged monomer MSD, as follows. Using $S(t) = \int \prod_i dh_i \mathcal{W}_t[h|h^0]$ and the FPE (8), we obtain the evolution equation $\partial_t S(t) = -\mu\langle h_0^2(t)\rangle S(t)$, where $\langle \cdots \rangle$ in the presence of absorption involves averaging over surviving realizations only; see Eq. (14) below. With the use of the initial condition $S(0) = 1$, the solution is

$$S(t) = \exp\left(-\mu\int_0^t d\tau\langle h_0^2(\tau)\rangle\right). \quad (12)$$

As before, the mean displacement and the connected correlation function are obtained by differentiating the generating function $\mathcal{F}_t[b] = \ln\mathcal{G}_t[b]$; one finds

$$\langle h_i(t)\rangle = (\Omega_t^{-1}Y_t h^0)_i, \quad \langle h_i(t)h_j(t)\rangle_c = 2(\Omega_t^{-1})_{ij}. \quad (13)$$

The correlation function $\langle h_i(t)h_j(t)\rangle_c$ is independent of the initial condition h^0 and has a finite value in the long time limit, whereas $\langle h_i(t)\rangle$ vanishes in that limit. In particular, the MSD in the long time limit reaches a steady-state value: $\langle h_0^2(t \to \infty)\rangle = [2/(K - \Delta)]_{00}$.

A dimensional analysis in the limit of a long polymer $L \to \infty$ allows one to deduce that $\langle h_0^2(t \to \infty)\rangle = a_0\mu^{-1/3}$, where a_0 is a dimensionless constant of order unity. Noting that in the absence of absorption the tagged monomer subdiffuses as $\langle h_0^2(t)\rangle \sim \sqrt{t}$, we see from the absorbing term in the FPE (8) that absorption is effective over times such that $\mu t^{3/2} \sim O(1)$. Thus, we have $\langle h_0^2(t)\rangle \sim \sqrt{t}F(\mu t^{3/2})$, where the scaling function $F(x)$ is a constant as $x \to 0$. Since $\langle h_0^2(t \to \infty)\rangle$ approaches a constant, it follows that $F(x \to \infty) \sim x^{-1/3}$, giving $\langle h_0^2(t \to \infty)\rangle = a_0\mu^{-1/3}$. Equation (12) gives $S(t) \sim \exp[-a_0\mu^{2/3}t]$ in the long time limit, independently of h^0; see Table I.

We now discuss the full time evolution of $\langle h_0^2(t)\rangle$ for a given initial configuration h^0. The MSD is

$$\langle h_0^2(t)\rangle = \frac{\int \prod_i dh_i h_0^2 \mathcal{W}_t[h|h^0]}{\int \prod_i dh_i \mathcal{W}_t[h|h^0]}. \quad (14)$$

In order to evaluate the MSD involving an average over an ensemble of initial configurations, we should weigh the contribution (14) with $p_{\mathrm{eq}}(h^0)S(t)/\overline{S(t)}$, where $S(t)/\overline{S(t)}$ is the probability that the configurations starting from h^0 at time $t = 0$ belong to the ensemble of surviving configurations at time t. Denoting the average MSD as $\overline{\langle h_0^2(t)\rangle}^{\mathrm{abs}}$, we compute it from the generating function

$$\ln(\overline{\mathcal{G}_t[b]}) = \ln\overline{S(t)} + \frac{1}{2}b^{\mathrm{T}}C_t b, \quad (15)$$

$$\overline{S(t)} = \sqrt{\frac{\det(e^{-t\Delta}Y_t\Omega_t^{-1})}{\det(1 + \sigma Q_t)}}, \quad (16)$$

$$C_t = 2\Omega_t^{-1} + \Omega_t^{-1}Y_t(1 + \sigma Q_t)^{-1}\sigma Y_t\Omega_t^{-1}, \quad (17)$$

with 1 the identity matrix. In particular, we obtain

$$\overline{\langle h_0^2(t)\rangle}^{\mathrm{abs}} = \partial^2 \ln(\overline{\mathcal{G}_t[b]})/\partial b_0^2|_{b=0} = (C_t)_{00}. \quad (18)$$

We compute numerically Eqs. (16) and (18) for different initial temperatures T_0. The results are shown in Figs. 2 and 3. As expected by our scaling arguments, both the decay rate of $S(t)$ and the steady-state value of the MSD are independent of the initial configuration. For the MSD, the approach to the steady-state value $a_0\mu^{-1/3}$ is always as $1/t$ (inset of Fig. 3), i.e., faster than the behavior $1/\sqrt{t}$ obtained for the case of pinning. For initially flat interface (i.e., $T_0 = 0$), we see from Fig. 3 that Eq. (18) behaves monotonically in time. Whereas for elastic pinning, a bump appears only above $T_0 = 1$, with absorption a bump is observed already for $T_0 = 1$ and further enhanced for larger T_0 values (Fig. 3). It would be interesting to understand why the approach to steady state differs in the

FIG. 3 (color online). MSD in the presence of harmonic absorption; Eq. (18). The MSD converges to a constant that is independent of T_0. Inset : Plot of $\delta h^2 = |\langle h_0^2(t)\rangle^{\mathrm{abs}} - \langle h_0^2(\infty)\rangle|$ shows the $\sim 1/t$ approach to the steady state.

two cases. Our numerical results are supported by direct Monte Carlo simulations of the interface dynamics and by a careful finite-size analysis presented in the Supplemental Material [29].

Conclusion.—In this Letter, we analyzed tagged monomer dynamics under the action of elastic pinning or harmonic absorption. Our solution stems from the crucial observation that in the presence of harmonic interactions, the stochastic evolution of the tagged monomer remains Gaussian. Some of our results, e.g., the presence of a unique steady state or the bump in MSD corresponding to a temperature quench, can be intuitively understood. Others such as the exponential decay of the survival probability or the power-law transient behaviors in the presence of absorption were observed in numerical simulations [8] but were not analytically known before. Finally, some of our results like the change of power law for $T_0 \neq 1$ (pinning case) or the bump observed when $T_0 = 1$ (harmonic absorption) were unexpected.

In this work, we focused on the case of one-dimensional polymers. However, it is straightforward to generalize our analysis to either a Rouse chain in d dimensions [34] or a d-dimensional EW interface by using the corresponding Laplacian matrix in place of Δ. Moreover, hydrodynamic effects for the chain or long-range elastic interactions for the interface can also be included by replacing Δ with the corresponding fractional Laplacian $-(-\Delta)^{z/2}$ [2]; in this case, the MSD of the tagged particle subdiffuses as $t^{(z-1)/z}$ with $z > 1$ for the chain and as $t^{(z-d)/z}$ with $z > d$ for the interface [35]. It would be interesting to study the effect of the pinning and absorption in the case of nonlinear models such as self-avoiding polymers and Kardar-Parisi-Zhang interfaces [36]. Another open issue is to go beyond the harmonic approximation and study absorption in the presence of localized targets.
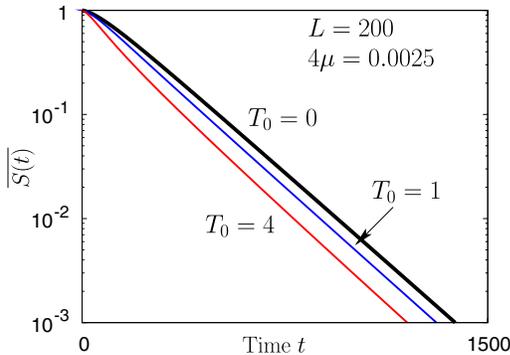
FIG. 2 (color online). Survival probability for different initial temperatures T_0. We observe at long times an exponential decay $\overline{S(t)} \sim \exp[-a_0\mu^{2/3}t]$ independent of the initial condition.
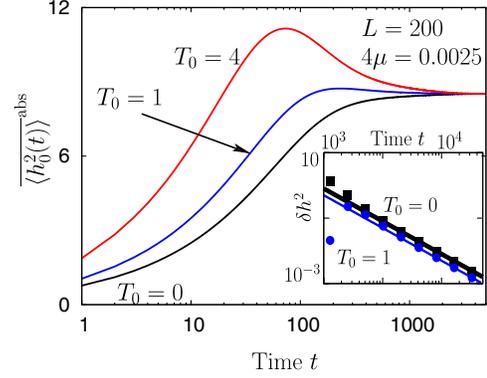
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