

Deviations from Blob Scaling Theory for Active Brownian Filaments Confined Within Cavities

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Scaling arguments used to predict the radius of gyration of passive self-avoiding flexible polymers have been shown to hold for polymers under the influence of active fluctuations. In this Letter, we establish how the standard blob scaling theory representation of a polymer, capable of capturing the essential physics of passive polymers under a variety of settings, breaks down when dealing with active polymers under confinement. Using numerical simulations, we show how the predicted exponents associated to the forces applied by a polymer when restricted within cavities of different geometries hold only whenever the persistence length generated on the polymer by the active forces is much smaller than the size of the characteristic blob in the scaling theory.

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The way biological filaments organize within the cellular milieu is of critical importance for the proper functioning of the cell, and it is responsible for numerous processes including, among other things, cellular subdivision and cell migration. The dynamics of biopolymers such as actin filaments and microtubules is regulated by the availability, or lack thereof, of ATP or GTP, which are energy sources capable of powering their growth and locomotion. These biological filaments are beautiful examples of active systems. This is an umbrella term used to indicate all those biological [1] or synthetic systems containing components that can be driven far from equilibrium by exploiting local chemical, electrical, or thermal gradients [2]. Over the last decade, several synthetic active systems have been developed and allowed for a systematic study of their properties in the context of complex fluids. Several reviews have been dedicated to the theoretical and experimental studies of colloidal active matter [3–8], with a few reviews focusing on more specific topics such as motility-induced phase separation (MIPS) [9,10], Viscek models [11], charged particles [12], self-assembly strategies [13], and mechanisms of locomotion [14].

Because of their biological implications and their role as a minimal model where the competition between thermal, elastic, and active forces can be easily and systematically studied, active filaments have become the subject of intense scrutiny over the last few years [15–28] (see also [29] for a brief review on the subject and references therein). Specifically, two distinct variations of these systems have been put forward: one where the active forces are locally aligned to the backbone of the polymer, and another where each monomer is treated as an independent active Brownian particle and the direction of the active forces is allowed to randomly rotate within the backbone of the

polymer. The first model was proposed to describe the collective behavior and pattern formation observed in systems of actin filaments activated by molecular motors (see, for instance, [30,31]) or strings of Janus dipolar particles [32], whereas the second model has been designed to mimic the behavior of a passive filament subject to the random fluctuations of an embedding active fluid. In this Letter, we focus exclusively on the statistical properties of the second type of active polymer.

Interestingly, it has been shown [20] that the radius of gyration, R_g , of an active filament of a fixed number of monomers N has a nonmonotonic dependence on the strength of the active forces for self-avoiding chains. An initial compression of the polymer for intermediate active forces is followed by a reexpansion at large activities. Analytical results for an ideal polymer were also presented in [20], and they indicate that the effect of the activity can be simply mapped into an effective temperature. However, more recently, it was argued [22] that a more complex nonlinear dependence of the end-to-end distance of the polymer should hold even for ideal chains. Nonetheless, for a fixed active force, R_g is expected to follow the Flory scaling law exhibited by its passive counterpart when plotted as a function of N (at least for Péclet numbers that are not too large). Recent numerical simulations have indicated that the equilibrium size scaling for the radius of gyration of the polymer also holds for self-avoiding flexible chains in both 2D [20] and 3D [27]. Specifically, $R_g \sim N^\nu$, with $\nu = 3/(d + 2)$ [33]. Here, d is the dimension of the embedding space. This is a remarkable and highly nontrivial result because it suggests that the standard blob scaling theories used to understand the statistical properties of passive polymers could, in principle, be employed to also understand the behavior of polymers subject to random active fluctuations.

In this Letter, we test the limits of this assumption. Specifically, we focus on the well-established scaling laws for passive self-avoiding polymers under confinement (see [34–40] and references therein), and we discuss to what extent the results obtained for an unconstrained active filament hold when confinement is added to the picture. One of the predictions of the blob scaling theory is that the free energy cost required to confine a flexible self-avoiding polymer composed of N monomers within a cylindrical pore scales as $\Delta F \sim (R_g/R)^{1/\nu}$. When considering a confinement within a spherical cavity, the free energy scales as $\Delta F \sim (R_g/R)^{d/(d\nu-1)}$. It is well known that free energies cannot be consistently defined in active systems; yet, their derivatives, i.e., pressures and forces the polymer exerts on confining boundaries, can be easily measured. Using numerical simulations, we show how these scaling laws hold for relatively weak activities and moderate degrees of confinement, but they break down when considering active polymers under confinement in two and three dimensions; this suggests that the blob scaling picture that can be used ubiquitously for passive polymers cannot always be straightforwardly exported to active systems when constraining their fluctuations.

Our model for a flexible, active self-avoiding polymer consists of N monomers of diameter σ linearly connected with harmonic springs and subject to thermal and active forces. In this study, we consider active polymers in both two and three dimensions. Every monomer undergoes active Brownian dynamics at a constant temperature T according to the following translational and rotational equations of motion:

$$\frac{d\mathbf{r}(t)}{dt} = \frac{1}{\gamma} \mathbf{f}(\{r_{ij}\}) + v_p \hat{\mathbf{q}}(t) + \sqrt{2D} \boldsymbol{\xi}(t) \quad (1)$$

$$\frac{d\hat{\mathbf{q}}(t)}{dt} = \sqrt{2D_r} \boldsymbol{\xi}_r(t) \times \hat{\mathbf{q}}(t) \quad (2)$$

where self-propulsion is introduced via a directional propelling velocity of constant magnitude v_p and is directed along a predefined orientation unit vector $\hat{\mathbf{q}}$ centered at the origin of each monomer. The translational diffusion coefficient D is related to the temperature and the translational friction γ via the Stokes-Einstein relation $D = k_B T \gamma^{-1}$. Likewise, the rotational diffusion coefficient $D_r = k_B T \gamma_r^{-1}$, with $D_r = 3D\sigma^{-2}$. The solvent induced Gaussian white-noise terms for both the translational $\boldsymbol{\xi}$ and rotational $\boldsymbol{\xi}_r$ motions are characterized by $\langle \boldsymbol{\xi}(t) \rangle = 0$ and $\langle \xi_i(t) \xi_j(t') \rangle = \delta_{ij} \delta(t-t')$. $\mathbf{f}(\{r_{ij}\})$ indicates the excluded volume and the harmonic forces holding the polymer together. The excluded volume between the monomers is enforced via a Weeks-Chandler-Andersen (WCA) potential

$$U(r_{ij}) = 4\epsilon \left[\left(\frac{\sigma}{r_{ij}} \right)^{12} - \left(\frac{\sigma}{r_{ij}} \right)^6 + \frac{1}{4} \right].$$

Here, $\epsilon = 1k_B T$. Harmonic bonds of the form $U_b = k(|r_{i,i+1}| - \sigma)^2$ ensure chain connectivity. Here, $r_{i,i+1}$ is the distance between consecutive monomers along the chain, and $k = 400k_B T$ is set to be large enough to ensure polymer connectivity while simultaneously minimizing bond stretching that could arise from the action of the active forces.

When considering spherical, or the equivalent circular confinement in two dimensions, each monomer also experiences a force due a WCA-like potential centered around the origin and of radius λ , which is of the form

$$U(\lambda - |r_i|) = 4\epsilon \left[\left(\frac{\sigma}{\lambda - |r_i|} \right)^{12} - \left(\frac{\sigma}{\lambda - |r_i|} \right)^6 + \frac{1}{4} \right] \quad (3)$$

This potential extends up to $(\lambda - |r_i|) = 2^{1/6}\sigma$. With this setup, each monomer feels a repulsive energy from the wall of $k_B T$ when $|r_i| = \lambda - \sigma$. The effective confinement radius, R , is therefore defined as $R = (\lambda - \sigma) + \sigma/2$, where the term $\sigma/2$ is added to account for the finite size of the monomers. An identical potential is used when enforcing planar confinement, with the only difference that $|r_i|$ is replaced with $|h_i|$, which is the component of the particle position vector that is perpendicular to the confining plane. In our simulations, σ and $k_B T$ are used as the units of length and energy scales of the system, whereas $\tau = \sigma^2 D^{-1}$ is our unit of time. All simulations were typically run for at least 10^9 iterations, with a time step ranging from $\Delta t = 10^{-4}\tau$ to $\Delta t = 5 \times 10^{-5}\tau$. To quantify the strength of the active forces, it is useful to introduce the dimensionless Péclet number, defined as $Pe = v_p \sigma / D$, and the characteristic persistence length of the active path $l_p = v_p / D_r$.

We began our simulations by measuring the radius of gyration of active polymers in two dimensions for a range of numbers of monomers going from $N = 64$ to $N = 1024$, and we found a scaling exponent of $\nu = 0.74(3)$, which was consistent with the one expected by the Flory theory of $\nu_F = 3/4$ [33] for a set of Péclet numbers ranging from 10 to 80. Next, we considered the confinement of a two-dimensional ($d = 2$) active polymer and a three-dimensional ($d = 3$) active polymer between two parallel plates at a distance $2R$ from each other and within an infinitely long cylindrical channel of radius R , respectively. As discussed above, the free energy associated to the confinement of such polymers is expected to scale as $\Delta F \sim k_B T (R_g/R)^{1/\nu}$. If this scaling equation holds, then the force the polymer exerts on the constraining walls or cylinder should scale as $f_w = -\partial \Delta F / \partial R \sim R^{-\alpha}$, where α is expected to be equal to $(1/\nu + 1)$.

To test this scaling assumption, we performed a series of simulations for different degree of confinement R and a

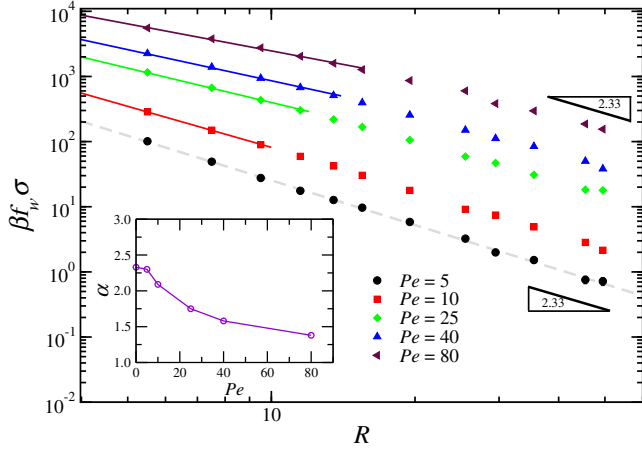


FIG. 1. Force exerted by an active polymer confined between two parallel walls at a distance $2R$ from each other for different Péclet numbers. The dashed line shows the expected dependence for the passive system $\alpha \approx 2.33$. The straight lines are fit to the data in the activity-dominated regime. The inset shows how the corresponding exponent α depends on Pe .

range of values of v_p . The results are shown in the log-log plots in Figs. 1 and 2, corresponding to polymers in two and three dimensions, respectively. Here, we plotted how the force, f_w , exerted by an active polymer depends on the degree of confinement R for different activities, Pe .

Our results show unequivocally how the predictions from the blob scaling theory fail to describe the behavior of active polymers under confinement. The results also show that a new length scale, which is irrelevant for the scaling behavior of an unbounded active polymer, emerges when confinement is imposed upon the polymer, namely, l_p . More specifically, the ratio l_p/R controls the behavior of

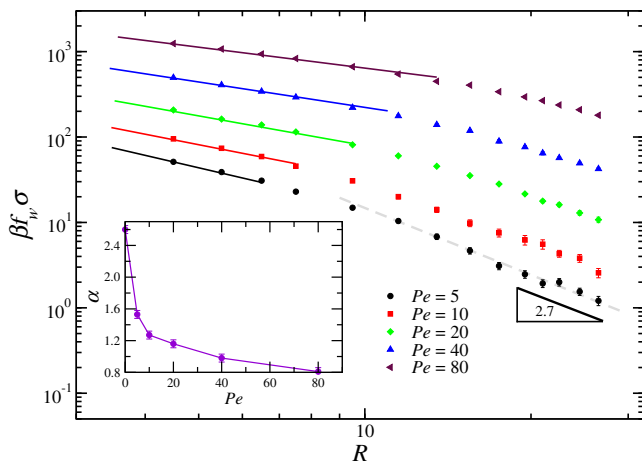


FIG. 2. Force exerted by an active polymer confined within a cylinder of radius R for different Péclet numbers. The dashed line shows the expected dependence for the passive system $\alpha \approx 2.7$. The straight lines are fit to the data in the activity-dominated regime. The inset shows how the corresponding exponent α depends on Pe .

the system. In fact, we find that the dependence of the force with the degree of confinement does not have a simple power law behavior but appears to be properly described by two distinct power laws connected by a crossover region. In the first regime, when $l_p/(2R) \ll 1$, the active polymer behaves consistently with the behavior expected from the parent passive system, albeit with a prefactor that depends on the Péclet number. In the second regime, when $l_p/(2R) > 1$, large deviations ensue. Here, the data are still properly described by a power law; however the associated exponent α becomes systematically smaller as the strength of the active forces increases. Interestingly, the decay of α with Pe is sharper for the three-dimensional polymer within the cylinder than for its two-dimensional counterpart between the two walls. This is clearly visible when comparing the insets of Figs. 1 and 2. We believe that this is because the free energy cost required to confine or compress a two-dimensional passive polymer is larger than that required to compress (to the same degree of confinement, R) a three-dimensional polymer with the same number of monomers. In fact, it can be readily shown [using, for simplicity, $\nu = 3/(d+2)$] that $\Delta F_{2d}/\Delta F_{3d} \approx (R/\sigma)^{1/3}$ for planar confinement and $\Delta F_{2d}/\Delta F_{3d} \approx N^{1/2}(N\sigma/R)^{1/4}$ (with $N\sigma > R$) for an isotropic compression, making the three-dimensional polymer effectively softer and more susceptible to the action of the active forces. Figure 3 shows typical configurations of the polymer confined between two rigid walls when $l_p/(2R) \ll 1$ (top) and for $l_p/(2R) > 1$ (bottom). In the first case, the polymer is not strongly affected by the active force and behaves effectively as a passive polymer at a higher temperature. In the second case, the polymer acquires conformations that can be best described as a mixture of collapsed configurations followed by fully stretched segments. Such highly dynamic heterogeneous conformations are clearly not amenable to be treated with the standard blob scaling representation, postulating that a linear polymer can be described as a sequence of blobs of radius R ; inside of which, the monomers are essentially unperturbed by the confinement forces and satisfy the bulk polymer statistics [38].

Analogous results are obtained when considering an active two-dimensional polymer confined within a circular cavity of radius R . According to the blob scaling theory, the

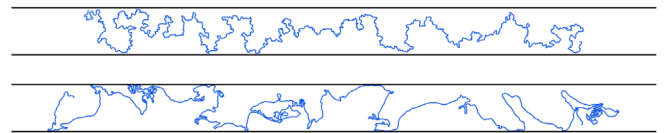


FIG. 3. Typical configurations for active polymers confined between two walls at a distance $2R$ from each other, with $R = 15.5\sigma$. The top image refers to an active polymer with $Pe = 2$, whereas the bottom image refers to the same polymer with $Pe = 80$.

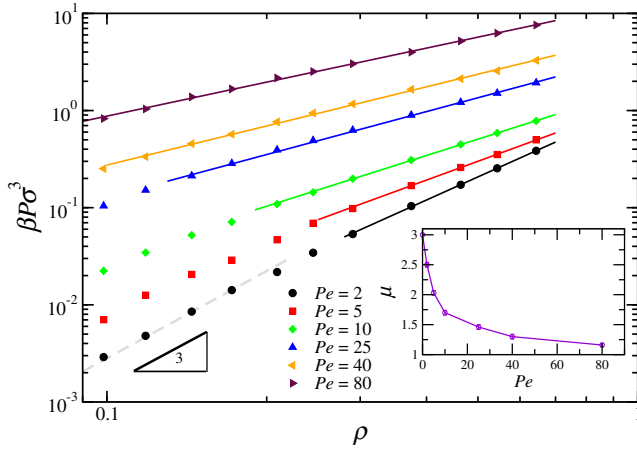


FIG. 4. Pressure exerted by an active polymer confined within a circular cavity at a number density $\rho = N/(\pi R^2)$ for different Péclet numbers. The dashed line shows the expected dependence for the passive system. Straight lines are fit to data in the activity-dominated regime. The inset shows how the corresponding exponent μ depends on Pe .

pressure exerted by a passive polymer on the boundary of the cavity is expected to scale as $P \sim \rho^\mu$, where ρ is the density of monomers inside the cavity and the exponent for the passive system is expected to be $\mu = d\nu/(d\nu - 1)$. In this case, the size of the blob is equal to the correlation length $\xi \sim [R^2/(\sigma^2 N)]^{\mu/d}$. We therefore expect, consistent with our results shown in Fig. 4, deviations from the standard scaling theory for $(l_p/\xi) > 1$. Notice that, because ξ is much smaller than R for even moderate degrees of confinement, already, when $Pe = 25$, most of the data points (obtained with a polymer of $N = 1024$ monomers) are within the activity-dominated regime; and no crossover from an effective passive behavior is even detectable. In conclusion, we studied under what conditions the blob scaling theory is applicable to active polymers under confinement. Our results suggest a breakdown of the scaling predictions as soon as l_p becomes larger than the size of the smallest unperturbed blob in the polymer representation. In this regime, the forces or pressures applied on the confining geometries have a power law behavior that becomes dependent on the Péclet number. The associated scaling exponents systematically decrease upon increasing Pe . Given the conformations of the active polymer inside the cylinder (see Fig. 3), it is tempting to map it into a passive semiflexible polymer with an effective persistence length of $P \sim l_p$. In the strong confinement regime, the free energy cost to force such a polymer within a cylindrical pore of radius R scales as $\Delta F \sim k_B T N \sigma / \lambda$, where $\lambda \sim P^{1/3} R^{2/3}$ is the average distance between successive deflection points of the chain in the pore [41]. This would lead to an average force applied to the cylinder that scales as $f_w \sim R^{-5/3}$. This exponent is inconsistent with our results. In fact, we observe a significantly weaker

dependence of f_w on R for large Pe . Although we have not driven our system to extreme activities, it is well known that, in this limit, the force exerted by a single active particle on the surface of a container of lateral dimension R scales as $f_w = \gamma v_p \exp[-R/(2l_p)]$ [42,43] so that f_w is only weakly dependent on R when $(R/l_p) \rightarrow 0$. This is because the time spent by a particle on the surface applying a force becomes much larger than that required to cross the dimension of the confining cavity in the large v_p limit, and it is not implausible to also expect a similar behavior for our system. Indeed, in this limit, we expect that the major contribution to the forces on the boundaries should be due to the active forces rather than to entropic confinement, thus making the radius of the cavity less and less relevant.

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