Polymer stretching in the inertial range of turbulence

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We study the deformation of flexible polymers whose contour length lies in the inertial range of a homogeneous and isotropic turbulent flow. By using the elastic dumbbell model and a stochastic velocity field with nonsmooth spatial correlations, we obtain the probability density function of the extension as a function of the Weissenberg number and of the scaling exponent of the velocity structure functions. In a spatially rough flow, as in the inertial range of turbulence, the statistics of polymer stretching differs from that observed in laminar flows or in smooth chaotic flows. In particular, the probability distribution of polymer extensions decays as a stretched exponential, and the most probable extension grows as a power law of the Weissenberg number. Furthermore, the ability of the flow to stretch polymers weakens as the flow becomes rougher in space.

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I. INTRODUCTION

One of the main phenomena that characterize the deformation of flexible polymers in a fluid flow is the coil-stretch transition, which consists in an abrupt increase in polymer extension as the intensity of the velocity gradient exceeds a critical value [1-3]. The coil-stretch transition was predicted by de Gennes [4] in the case of a planar extensional flow. The relevant dimensionless parameter for the transition is the Weissenberg number Wi = $\gamma \tau$, where γ is the amplitude of the velocity gradient and τ is the polymer relaxation time. In an extensional flow, a polymer stays in the equilibrium coiled configuration as long as Wi < 1/2 and unravels almost completely as Wi exceeds 1/2. Correspondingly, the probability density function (PDF) of polymer extensions consists of a narrow peak that, as Wi exceeds 1/2, rapidly moves from extensions close to the equilibrium size of the polymer, R_0 , to extensions close to its contour length L. In his study of the coil-stretch transition, de Gennes [4] used the elastic dumbbell model, which describes a flexible polymer as two inertialess beads connected by an elastic spring and subjected to thermal fluctuations [5]. In the frame of reference of the center of mass, the configuration of a dumbbell is specified by the separation vector between the beads, \mathbf{R} , which satisfies the stochastic ordinary differential equation [5]

$$\dot{\mathbf{R}} = \mathbf{R} \cdot \nabla \mathbf{u} - \frac{f(R)}{2\tau} \mathbf{R} + \sqrt{\frac{R_0^2}{\tau}} \, \boldsymbol{\eta}(t), \tag{1}$$

where u is the incompressible velocity field, R = |R|, $\eta(t)$ is vectorial white noise, f(R) = 1 for a Hookean dumbbell and $f(R) = 1/(1 - R^2/L^2)$ for a finitely extensible nonlinear elastic (FENE) dumbbell. Perkins *et al.* [6] have observed the coil-stretch transition in a planar extensional flow by examining the deformation of fluorescently labeled DNA macromolecules.

Lumley [7,8] and subsequently Balkovsky *et al.* [9] have employed the elastic dumbbell model to show that the coilstretch transition also occurs in chaotic or random flows. This phenomenon has been confirmed experimentally by Steinberg and coworkers [3,10,11] via the direct observation of single-polymer dynamics in chaotic flows generated by elastic instabilities. It has also been studied in numerical

simulations of the dumbbell model for randomly fluctuating flows [12,13] and turbulent flows [14–17]. The criterion for the transition is that the Weissenberg number $Wi = \lambda \tau$ exceeds 1/2, where λ is the Lyapunov exponent of the flow [9,18]. However, in chaotic flows the statistics of polymer stretching has different properties compared to the laminar case. Indeed, for intermediate extensions $(R_0 \ll R \ll L)$, the PDF of R behaves as a power law of the form $R^{-1-\alpha}$. Thus, for a comparable intensity of the velocity gradient, the distribution of the extensions is much broader than in a laminar flow; coiled and stretched polymers coexist in the flow, and the value of Wi determines which configuration is predominant. The exponent α is a decreasing function of Wi; it is positive for Wi < 1/2, equals zero for Wi = 1/2, and is negative for greater values of Wi [9]. Hence, in the Hookean dumbbell model $(L = \infty)$, the probability distribution of the extension ceases to be normalizable for Wi = 1/2, and this behavior is interpreted as the sign of the coil-stretch transition in chaotic flows [9]. In the more realistic FENE model, the emergence of the stretched state manifests itself through the fact that the slope of the PDF of intermediate extensions changes from negative to positive, and consequently the maximum of the PDF of R abruptly moves from close to R_0 to close to L [19,20].

The phenomenology described above holds when the correlation time of u is short compared to λ^{-1} . In the opposite regime, the dynamics of the coil-stretch transition is different. The PDF of the extension is bimodal, and the appearance of the stretched state occurs through the drop of the peak at R_0 and the simultaneous rise of a second peak at L as Wi increases [21]. At large values of Wi, the PDF of intermediate extensions behaves as R^{-1} [21].

All the aforementioned studies of the dumbbell model assume that u is spatially smooth in the neighborhood of the polymer and that ∇u is uniform over the size of the molecule. For sufficiently large Reynolds numbers, the polymer contour length can exceed the viscous scale and polymers can stretch into the inertial range of turbulence, where the flow is spatially rough [22]. The Reynolds number required for polymers to experience inertial-range fluctuations is lower for long biological macromolecules, such as those used in the study of the coil-stretch transition in laminar velocity fields [23]; long fibers can even approach the integral

scale of a turbulent flow [24]. Wiese [25] has studied polymer deformation in a random field with nonsmooth spatial correlations by using renormalization-group techniques. Davoudi and Schumacher [26] have examined the dynamics of dumbbells that can stretch into the inertial range in a numerical simulation of polymer dynamics in a turbulent flow. Here we study this problem analytically by using a stochastic model of inertial-range turbulence. We obtain the PDF of polymer extension as a function of the scaling exponent of the velocity structure functions and show how, in a spatially rough flow, the phenomenology of polymer stretching differs from that observed before in laminar or in smooth chaotic flows.

II. SHORT-CORRELATED RANDOM FLOW

A fully analytical study of the coil-stretch transition in random flows can be performed in the limit in which the correlation time of u is short. This has been achieved in the case of a smooth random flow by using the Batchelor regime of the Kazantsev-Kraichnan model [27,28] to mimic small-scale velocity fluctuations (see Ref. [29] for a review on the application of this model to turbulent transport). The velocity field is Gaussian, statistically homogeneous and stationary, and has zero mean and correlation:

$$\langle u_i(\mathbf{x} + \mathbf{r}, t)u_i(\mathbf{x}, t')\rangle = D_{ii}(\mathbf{r})\delta(t - t'), \tag{2}$$

where the incompressibility of u requires $\sum_i \partial D_{ij}/\partial r_i = 0$. The velocity gradient thus plays the role of a multiplicative noise in Eq. (1). Note that, in virtue of the incompressibility of u, whether Eq. (1) is interpreted in the Itô or in the Stratonovich sense is immaterial as far as the statistics of R is concerned [29]. The PDF of the separation vector, P(R), indeed satisfies the Fokker-Planck equation [30]:

$$\partial_t P = \left[d_{ij}(\mathbf{R}) + \frac{R_0^2}{2\tau} \delta_{ij} \right] \partial_{R_i} \partial_{R_j} P - \frac{1}{2\tau} \partial_{R_i} [f(R)R_i P], \quad (3)$$

where $d_{ij}(\mathbf{r}) = D_{ij}(\mathbf{0}) - D_{ij}(\mathbf{r})$ are the structure functions of the velocity field. If \mathbf{u} is smooth and statistically isotropic and invariant under reflections, $d_{ij}(\mathbf{r})$ takes the form [29]

$$d_{ij}(\mathbf{r}) = D_1 r^2 [(d+1)\delta_{ij} - 2\hat{r}_i \hat{r}_j], \tag{4}$$

where $D_1 > 0$ determines the amplitude of the fluctuations of the velocity and d is the dimension of the flow. In this case, the stationary PDF of R only depends on R and can be sought as the long-time solution of the following Fokker-Planck equation in one variable:

$$\partial_T P = -\partial_R [C_1(R)P] + \partial_R^2 [C_2(R)P], \tag{5}$$

where $T = t/2\tau$, $C_1(R) = 2(d+1)$ Wi $R/d - f(R)R + (d-1)R_0^2/R$, and $C_2(R) = 2$ Wi $R^2/d + R_0^2$ [13,19,20,31] (for further details on the notation, see Ref. [32]). Here Wi = $\lambda \tau$, where $\lambda = D_1 d(d-1)$ is the Lyapunov exponent of the flow [33,34].

As R must be positive, the solution of Eq. (5) must satisfy a reflecting boundary condition at R = 0. Thus, in the stationary state the probability current must vanish everywhere, and the stationary solution of Eq. (5) is (e.g., Ref. [35])

$$P_{\rm st}(R) \propto \exp\left[\int^R d\zeta \ C_1(\zeta)/C_2(\zeta)\right]/C_2(R).$$
 (6)

In particular, for the FENE model, the explicit expression of the exponent α that characterizes the power-law behavior of $P_{\text{st}}(R)$ for intermediate extensions is [20]:

$$\alpha = (R_0^2/L^2 + 2Wi/d)^{-1} - d.$$
 (7)

For realistic values of R_0 and L, the ratio R_0^2/L^2 is much smaller than 1; hence $\alpha \approx -d(1-1/2\text{Wi})$ and approaches -d as Wi increases.

III. STRETCHING OF POLYMERS IN A SPATIALLY ROUGH FLOW

In a turbulent flow, the assumption that ∇u is uniform over the size of the polymer is appropriate provided the contour length of the polymer is much smaller than the Kolmogorov dissipation scale ℓ_K (e.g., Ref. [36]). The velocity field can then be Taylor expanded and, to a first approximation, the stretching effect of the flow is entirely given by the velocity gradient [see Eq. (1)]. However, long polymers with $R_0 < \ell_K$ but $L > \ell_K$ may extend beyond the Kolmogorov scale into the inertial range, where u is nonsmooth [36]. Davoudi and Schumacher [26] have considered an elastic dumbbell with maximum length greater than ℓ_K in a turbulent shear flow. The numerical simulations show that the PDF of R reaches a stationary form even for Wi greater than the critical value and for a Hookean force. Indeed, whereas in a smooth flow fluid particles separate exponentially, when the velocity is only Hölder continuous the mean-square separation grows asymptotically in time as t^{β} with $\beta > 1$ [29]. As a consequence, for extensions $R \gg \ell_K$ the stretching effect of the flow is weaker than the elastic relaxation even for a linear elastic force [26]. These findings are systematized below within the Kazantsev-Kraichnan model.

If R can take values greater than ℓ_K , the full difference between the velocities of the polymer ends must be retained in the dumbbell equation [26,37,38]:

$$\dot{\mathbf{R}} = \mathbf{u}(\mathbf{x}_2, t) - \mathbf{u}(\mathbf{x}_1, t) - \frac{f(R)}{2\tau} \mathbf{R} + \sqrt{\frac{R_0^2}{\tau}} \, \eta(t),$$
 (8)

where x_1 and x_2 are the positions of the two beads that compose the dumbbell (the separation vector is defined as $\mathbf{R} = x_2 - x_1$). For $r \gg \ell_K$, the tensor $d_{ij}(\mathbf{r})$ can be written in such a way as to reproduce the structure functions of a non-smooth velocity field [29]:

$$d_{ij}(\mathbf{r}) = a(\xi)D_1 \ell_K^{2-\xi} r^{\xi} [(d-1+\xi)\delta_{ij} - \xi \hat{r}_i \hat{r}_j], \quad (9)$$

where $a(\xi)$ is a positive constant and $\xi/2$ can be interpreted as the Hölder exponent of the velocity field and varies between 0 (white noise) and 1 (smooth flow). The value of ξ corresponding to the inertial-range scaling of a turbulent flow is $\xi = 4/3$ [39]. In this flow, the separation between two fluid particles grows in time according to a generalized Richardson law: $\langle r^{\mu} \rangle \sim t^{\mu/(2-\xi)}$ [29]. The impact of this property of the flow on the statistics of polymer stretching can be examined by substituting $d_{ij}(\mathbf{R})$ from Eq. (9) into Eq. (3) and by setting $f(\mathbf{R}) = 1$ and $R_0 = 0$. For $\ell_K \ll R \ll L$, the drift and

diffusion coefficients in Eq. (5) thus take the form

$$C_1(\rho) = \frac{2(d-1+\xi)a(\xi)\text{Wi}}{d}\rho^{\xi-1} - \rho,$$
 (10)

$$C_2(\rho) = \frac{2a(\xi)\text{Wi}}{d}\rho^{\xi},\tag{11}$$

with $\rho = R/\ell_K$, and from Eq. (6) the zero-current solution of Eq. (5) is

$$P_{\rm st}(\rho) \propto \rho^{d-1} \exp\left[-\frac{d\rho^{2-\xi}}{2a(\xi)\text{Wi}(2-\xi)}\right],$$

$$1 \ll \rho \ll L/\ell_K. \tag{12}$$

Note that $P_{\rm st}(\rho)$ has the same form as the instantaneous PDF of the distance between two fluid particles that separate in the inertial range of a turbulent flow [39]. Contrary to the case of a smooth flow, $P_{\rm st}(\rho)$ is normalizable for all Wi even in the limit $L \to \infty$. Thus, according to the criterion introduced by Balkovsky et al. [9], there is not a coil-stretch transition if the maximum length of polymers lies within the inertial range of a turbulent flow. Nevertheless, as Wi increases, polymers can stretch significantly according to the following phenomenology. For Wi $\lesssim d/2(d-1)$, $P_{st}(R)$ has a maximum near to R_0 , it decays as a power law $R^{-1-\alpha}$ with $\alpha > 0$ for $R_0 \ll R \ll \ell_K$, and decays as a stretched exponential for $R \gg \ell_K$. For Wi $\gtrsim d/2(d-1)$, the PDF of R increases as a power law for $R_0 \ll R \ll \ell_K$ and behaves as in Eq. (12) for $\ell_K \ll R \ll L$. For all Wi, $P_{\rm st}(R)$ falls fast to zero near to L. Hence, the peak of $P_{\rm st}(R)$ is at $R_{\star} = \ell_K [2(d-1)a(\xi){\rm Wi}/d]^{1/(2-\xi)}$ for the Hookean model and near to the smaller of R_{\star} and L for the FENE model.

The full PDF of R can be calculated by choosing $d_{ij}(\mathbf{r})$ in such a way as to interpolate between the smooth form given in Eq. (4) and the inertial-range behavior discussed in this section. The structure functions of a three-dimensional (d = 3), statistically homogeneous, isotropic, and parity invariant velocity field can be written as [40,41]

$$d_{ij}(\mathbf{r}) = d_{NN}(r)\delta_{ij} + [d_{LL}(r) - d_{NN}(r)]\hat{r}_i\hat{r}_j, \tag{13}$$

where $d_{LL}(r)$ and $d_{NN}(r)$ are the longitudinal and transverse structure functions, respectively, and can be expressed in terms

of the scalar spectrum E(k) as follows [41]:

$$d_{LL}(r) = 4 \int_0^\infty \left[\frac{1}{3} + \frac{\cos(kr)}{k^2 r^2} - \frac{\sin(kr)}{k^3 r^3} \right] E(k) dk, \quad (14)$$

$$d_{NN}(r) = d_{LL}(r) + \frac{r}{2}d'_{LL}(r). \tag{15}$$

The equation for $d_{NN}(r)$ follows from the incompressibility of u. In order to obtain structure functions that interpolate between the smooth behavior for $r \ll \ell_K$ and the Hölder behavior with exponent $\xi/2$ for $r \gg \ell_K$, we choose [29]

$$E(k) = ce^{-\ell_K^2 k^2} k^{-1-\xi}, \qquad 0 < \xi < 2,$$
 (16)

where c is a positive constant and has the dimensionality of length^{$2-\xi$} time⁻¹. The above expression assumes that the integral scale of the flow is set to infinity or, in other words, that L is much smaller than this scale. Equation (14) then yields (see formulas 3.952.7, 3.952.8, 9.212.1, in Ref. [42])

$$d_{LL}(r) = c\ell_K^{\xi} \Gamma\left(-\frac{\xi}{2} - 1\right) \left\{ \frac{2\ell_K^2}{r^2} \left[{}_1F_1\left(-\frac{\xi}{2} - 1; \frac{1}{2}; -\frac{r^2}{4\ell_K^2}\right) - {}_1F_1\left(-\frac{\xi}{2} - 1; \frac{3}{2}; -\frac{r^2}{4\ell_K^2}\right) \right] - \frac{\xi + 2}{3} \right\},$$
(17)

where Γ is the Euler gamma function and ${}_1F_1$ is the confluent hypergeometric function of the first kind. The asymptotic expressions of $d_{LL}(r)$ can be obtained by using formulae 13.1.2 and 13.1.5 in Ref. [43]:

$$d_{LL}(r) \sim 2D_1 r^2 \qquad (r \ll \ell_K), \tag{18}$$

and

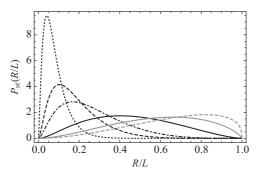
$$d_{LL}(r) \sim 2D_1 a(\xi) \ell_K^{2-\xi} r^{\xi} \qquad (r \gg \ell_K), \tag{19}$$

with

$$D_1 = \frac{c\xi \ell_K^{\xi - 2}}{60} |\Gamma(-\xi/2)|,\tag{20}$$

and

$$a(\xi) = 15 \frac{2^{1-\xi} \sqrt{\pi}(\xi+4)}{\xi(\xi+2)(\xi+5)\Gamma(\frac{\xi+3}{2})}.$$
 (21)



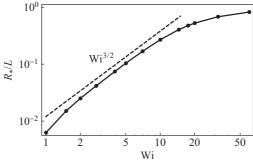


FIG. 1. Left: Stationary PDF of R/L for $R_0 = 0.1$, $\ell_K = 1$, $L = 10^3$, $\xi = 4/3$, and Wi = 2.75 (black dotted line), Wi = 5.0 (black dashed line), Wi = 7.0 (black dot-dashed line), Wi = 14.5 (black solid line), Wi = 30 (gray solid line), Wi = 60 (gray dashed line). The PDF is normalized as follows: $\int_0^\infty P_{\rm st}(R) dR = 1$. Right: Maximum of the PDF of R rescaled by L as a function of Wi. The dashed line is proportional to Wi^{3/2}.

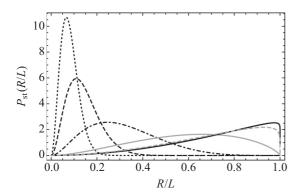


FIG. 2. Stationary PDF of R/L for $R_0 = 0.1$, $\ell_K = 1$, $L = 10^3$, Wi = 30, and $\xi = 1/3$ (black dotted line), $\xi = 2/3$ (black dashed line), $\xi = 1$ (black dot-dashed line), $\xi = 4/3$ (gray solid line), $\xi = 5/3$ (gray dashed line), $\xi = 2$ (black solid line). The gray solid line is the same as described in the caption of Fig. 1. For $\xi = 2$, the stationary PDF of R/L is calculated by using Eq. (4); its explicit form can be found in Ref. [20].

Thus, the structure functions under consideration have the asymptotic behaviors given in Eqs. (4) and (9).

By inserting Eq. (13) into Eq. (3) and by using statistical isotropy, we obtain the following expressions for the drift and diffusion coefficients of the Fokker-Planck equation for P(R) [see Eq. (5)]:

$$C_1(R) = \frac{4\tau d_{LL}(R)}{R} + 2\tau d'_{LL}(R) - Rf(R) + \frac{2R_0^2}{R}, \quad (22)$$

$$C_2(R) = 2\tau d_{LL}(R) + R_0^2. (23)$$

The stationary PDF of R is then calculated by substituting $C_1(R)$ and $C_2(R)$ from Eqs. (22) and (23) into Eq. (6). Figures 1 to 2 illustrate the behavior of the PDF of R/L for different values of Wi and ξ . The equilibrium size and the maximum length of the dumbbell are chosen in such a way as to emphasize the range of extensions $\ell_K \ll R \ll L$, in which the dumbbell experiences spatially rough velocity fluctuations. As Wi increases, the distribution of polymer extensions broadens considerably, and its maximum progressively translates toward larger extensions, until nonlinear elastic effects become important (Fig. 1). Thus, increasing Wi has the effect of stretching polymers beyond ℓ_K , even though fairly large values of Wi are required for the PDF of extensions comparable to L to become appreciable. Moreover, the emergence of the stretched state is gradual as a function of Wi (R_{\star} only grows as a power law of Wi) and, even for the largest values of Wi, a significant fraction of polymers deforms only in part. Finally, Fig. 2 shows that the ability of the flow to stretch polymers beyond ℓ_K is rather weak for small ξ and strengthens as the spatial regularity of the flow improves. In particular, both the mean and the standard deviation of $P_{\rm st}(R)$ increase with increasing ξ .

IV. CONCLUSIONS

We have studied the statistics of polymer extension in a homogeneous and isotropic turbulent flow in the case in which the contour length of the polymers lies in the inertial range. By using the elastic dumbbell model and a stochastic velocity field with nonsmooth spatial correlations, we have analytically calculated the PDF of polymer extension as a function of the Weissenberg number and of the scaling exponent of the velocity structure functions.

The behavior of $P_{\rm st}(R)$ can be summarized as follows:

$$P_{\rm st}(R) \propto \begin{cases} R^{d-1}, & 0 \le R \ll R_0, \\ R^{-1-\alpha}, & R_0 \ll R \ll \ell_K, \\ R^{d-1} \exp\left(-\frac{b_{\xi}}{\text{Wi}} \frac{R^{2-\xi}}{\ell_K^{2-\xi}}\right), & \ell_K \ll R \ll L, \end{cases}$$
(24)

where α is defined in Eq. (7) and b_{ξ} is a positive constant. For sufficiently large values of Wi, $P_{\rm st}(R)$ consists of a power-law left tail with an exponent close to d-1 and a stretched exponential right tail, which falls fast to zero near to L. The PDF of R is very broad; hence, polymers with very different end-to-end extensions coexist in the solution.

Our study shows that even a spatially rough random flow can stretch polymers. However, the way the stretched state emerges is different from that found in laminar flows and in smooth random flows $(L < \ell_K)$. In particular, for a smooth random flow with short correlation time, $P_{st}(R)$ displays a power-law behavior for $R_0 \ll R \ll L$ with a slope that changes from negative to positive as Wi increases [9]. Hence, when Wi exceeds a critical value, the maximum of $P_{\rm st}(R)$ abruptly moves from near to R_0 to near to L [20]. If the contour length of polymers exceeds the dissipation scale of a turbulent flow, increasing the Weissenberg number results in a gradual translation of the maximum of $P_{\rm st}(R)$ from R_0 to $R_\star \propto \ell_K {\rm Wi}^{1/(2-\xi)}$, until nonlinear elastic effects dominate the dynamics. Contrary to the case $L < \ell_K$, the emergence of the stretched state is therefore not abrupt and there is not a coil-stretch transition. Moreover, compared to the case $L < \ell_K$, much larger values of Wi are required to obtain comparable fractional extensions if $L > \ell_K$. The stretching effect of the flow also weakens as the flow becomes rougher in space.

The scale R_{\star} can be identified with Lumley's scale in the theory of turbulent drag reduction [8,26]. The scale R_{\star} is such that the eddy turnover time associated with R_{\star} is comparable to the polymer relaxation time τ . At scales smaller than R_{\star} , the stretching effect of the flow dominates the elastic relaxation, whereas the latter is stronger than the former at larger scales.

Our study can be generalized to the case of dumbbells whose maximum extension exceeds the integral scale ℓ_0 of a turbulent flow. By definition of integral scale, the velocity field is indeed spatially uncorrelated at scales greater than ℓ_0 . Hence, the behavior of the structure functions for $r\gg\ell_0$ can be captured by setting $\xi=0$ in Eq. (9). From Eq. (12), the PDF of R for $\ell_0\ll R\ll L$ is therefore of the form $P_{\rm st}(R)\propto R^{d-1}\exp[-b_0R^2/\ell_0^2{\rm Wi}]$ with $b_0>0$, which indicates that the probability of polymer extension decreases very rapidly for $R\gtrsim\ell_0{\rm Wi}^{1/2}$.

It was shown in Refs. [13,20,30] that in a smooth random flow the coil-stretch transition is characterized by a critical slowing down of polymer dynamics. The time required for the distribution of polymer extension to reach equilibrium indeed shows a peak near to the coil-stretch transition. This behavior is due to the large breadth of the PDF of R for values of Wi near

to the critical one. It would be interesting to study whether or not this phenomenon also persists when polymers can extend into the inertial range. In view of the breadth of the PDFs shown in Figs. 1 and 2, we expect that an analogous peak of the equilibriation time will be associated with the emergence of the stretched state. However, the peak will be much broader than for a smooth flow, because for polymers longer than ℓ_K the emergence of the stretched state is slower as a function of Wi. A second interesting question concerns the effects of a potential preferential sampling of the flow [37,38], which

is not present when the correlation time of the velocity field vanishes [44].

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