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Polymer stretching in laminar and random flows: entropic characterization

Stefano Musacchio,¹ Victor Steinberg,² and Dario Vincenzi^{3,*}

¹*Dipartimento di Fisica and INFN, Università di Torino, Via P. Giuria 1, 10125 Torino, Italy*

²*Department of Physics of Complex Systems, Weizmann Institute of Science, Rehovot 76100, Israel*

³*Université Côte d'Azur, CNRS, LJAD, Nice, France*

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Polymers in non-uniform flows undergo strong deformation, which in the presence of persistent stretching can result in the coil–stretch transition. The statistics of polymer deformation depends strongly on the nature and the properties of the flow. Sultanov *et al.* [Phys. Rev. E **103**, 033107 (2021)] have characterized the coil–stretch transition in an elastic turbulence of von Kármán flow by measuring the entropy of polymer extension as a function of the Weissenberg number. The entropic characterization of the coil–stretch transition is here extended to a set of laminar and random velocity fields that are benchmarks for the study of polymer stretching in flow. In the case of random velocity fields, a suitable description of the transition is obtained by considering the entropy of the logarithm of the extension instead of the entropy of the extension itself. Entropy emerges as an effective tool for capturing the coil–stretch transition and comparing its features in different flows.

I. INTRODUCTION

The configuration of a polymer in a moving fluid drastically changes from coiled to fully stretched when the Weissenberg number Wi , *i.e.* the product of the characteristic velocity gradient and the polymer relaxation time, exceeds a critical threshold. This phenomenon is known as the coil–stretch transition [1] and is observed in both laminar [2, 3] and random flows [4–6], even though with partially different features in the two cases. Several observables have been used to characterize the coil–stretch transition. A natural quantity is the steady-state distribution of polymer extensions [2–6], which changes dramatically near to the critical Wi : the mean increases rapidly, the coefficient of variation attains its maximum value, and the peak shifts from the equilibrium extension R_{eq} to the maximum length L (here R_{eq} is the polymer root mean square extension in the absence of flow). Another characterization considers the equilibration time of the statistics of polymer extension [7, 8] or alternatively the autocorrelation time of the extension [9]; near the coil–stretch transition these properties are strongly amplified that results in a critical slowing down of the stretching dynamics. Furthermore, the transition is characterized by a maximum dispersion of the work done by the flow to stretch polymers [10].

Recently, Sultanov *et al.* [11] have proposed to study the coil–stretch transition by measuring the entropy of the polymer extension. This quantifies the “randomness” of the extension within an ensemble of polymers. By imaging fluorescently stained T4 DNA molecules of maximum length $L = 71.7\mu m$ and radius of gyration $R_g = 1.5\mu m$ in an elastic turbulence of von Kármán flow [12, 13], Sultanov *et al.* [11] have found that the entropy displays a maximum near the transition. This result has a clear interpretation in terms of information. In the coiled and stretched states the information concerning the polymer elongation reaches a maximum because the distribution of the polymer extensions is peaked around a single value (R_{eq} and L , respectively). These states are hence minima of entropy. Conversely, the broadening of the probability distribution of polymer elongations at the transition corresponds to a loss of information and therefore a maximum of entropy.

Here we pursue the entropic characterization of the coil–stretch transition by examining a set of analytical and numerical flows. Since it concentrates information on the statistics of polymer stretching in a single scalar quantity, entropy emerges as an effective tool for comparing polymer stretching in different flows.

II. POLYMER MODEL AND FLOW CONFIGURATIONS

The polymer is modelled as a finitely extensible nonlinear elastic (FENE) dumbbell [14–16]. The evolution equation for the polymer end-to-end vector \mathbf{R} is

$$\frac{d\mathbf{R}}{dt} = \boldsymbol{\kappa}(t) \cdot \mathbf{R} - f(R) \frac{\mathbf{R}}{2\tau} + \sqrt{\frac{R_0^2}{\tau}} \boldsymbol{\xi}(t), \quad (1)$$

* Also Associate, International Centre for Theoretical Sciences, Tata Institute of Fundamental Research, Bangalore 560089, India

where $\kappa_{ij}(t) = \nabla_j u_i(t)$ is the velocity gradient at the centre of mass of the polymer, τ is the polymer longest relaxation time, $R_0 = R_{\text{eq}}/\sqrt{3}$, $f(R) = (1 - R^2/L^2)^{-1}$, and $\boldsymbol{\xi}(t)$ is three-dimensional white noise. Within this model, the radius of gyration is $R_g = R_{\text{eq}}/2 = \frac{\sqrt{3}}{2}R_0$ and the extensibility parameter is defined as $b = (L/R_0)^2$ [14]. The dumbbell model can in principle be refined to include effects such as hydrodynamic interactions or a conformation-dependent drag force [14, 15]. Given that our work is focused on the entropic characterization of the coil–stretch transition, rather than on the properties of dumbbell model itself, for the sake of simplicity we restrict to the basic version of the model, which in any case has proved useful for a qualitative, and sometimes even quantitative, understanding of the coil–stretch transition, in both steady [1–3, 17] and random [18–21] flows.

Calculating the entropy requires obtaining the probability density function (PDF) of the extension, $P(R)$, from Eq. (1), analytically or numerically. We shall consider the following set of model flows, which have been widely employed in the study of polymer stretching and are representative of more complex situations.

Extensional flow The uniaxial extensional flow $\mathbf{u} = \gamma(-x/2, -y/2, z)$ is the first configuration in which the coil–stretch transition has been predicted [1] and observed experimentally [2]. It consists of a direction of pure stretching and two directions of compression with magnitudes that ensure incompressibility. The Weissenberg number is defined as $\text{Wi} = \gamma\tau$ and its critical value is $\text{Wi}_{\text{cr}} = 1/2$. If the rescaled end-to-end vector $\boldsymbol{\rho} = \mathbf{R}/L$ is expressed in spherical coordinates as $\boldsymbol{\rho} = \rho(\sin\theta \cos\phi, \sin\theta \sin\phi, \cos\theta)$, then the stationary PDF of $\boldsymbol{\rho}$ is

$$P(\boldsymbol{\rho}) \propto (1 - \rho^2)^{b/2} \exp\left\{\frac{b\text{Wi}}{2}\rho^2[3\cos^2(\theta) - 1]\right\}, \quad (2)$$

where $b = (L/R_0)^2$ is the extensibility parameter [14]. An integration over the angular variables yields

$$P(\rho) \propto \rho e^{-\frac{b\text{Wi}}{2}\rho^2} (1 - \rho^2)^{b/2} \text{erf}\left(i\frac{3b\text{Wi}}{2}\rho\right), \quad (3)$$

where erf is the error function.

Shear flow In a linear shear flow $\mathbf{u} = (\sigma y, 0, 0)$, the coil–stretch transition is not observed [22]. Owing to thermal fluctuations, the dynamics of the polymer indeed consists of a sequence of tumbling events which in turn correspond to as many coiling and stretching events, so that persistent stretching is never realized [23–25]. Nevertheless, it will be instructive to study the entropy of polymer extension also in this configuration and compare its behaviour with that observed in other flows. The Weissenberg number is $\text{Wi} = \sigma\tau$, and the PDF of R is now calculated numerically by means of Brownian Dynamics simulations of Eq. (1), where the nonlinearity of the elastic force is resolved by using Öttinger’s rejection algorithm [26].

Batchelor–Kraichnan (BK) flow In random flows, it is convenient to define the Weissenberg number as $\text{Wi} = \lambda\tau$, where λ is the Lyapunov exponent of the flow, *i.e.* the average stretching rate of line elements. A general theory of the coil–stretch transition in random flows has been developed by Balkovsky *et al.* [19] for linear polymer elasticity (Oldroyd-B model) and by Chertkov [20] for nonlinear polymer elasticity (FENE model). For intermediate extensions $1/\sqrt{b} \ll \rho \ll 1$, the PDF of ρ behaves as $\rho^{-1-\alpha}$ with α decreasing as a function of Wi and crossing zero at $\text{Wi} = 1/2$. Therefore, in the limit $L \rightarrow \infty$ the PDF of ρ is not normalizable if $\text{Wi} \geq 1/2$. This is interpreted as an indication that the coil–stretch transition also exists in random flows and the critical Wi is again $\text{Wi}_{\text{cr}} = 1/2$. For finite L , the measured slope may be affected by the nonlinearity of the elastic force, but the theory still implies an analogous strong modification of $P(R)$ at Wi_c [20].

The BK flow has been used extensively in the analytical study of turbulent transport below the viscous-dissipation scale (see Ref. [27] and, for applications to polymer dynamics, Ref. [28] and references therein). The velocity gradient is an isotropic tensorial white noise with correlation $\langle \kappa_{ij}(t)\kappa_{kl}(t') \rangle = \lambda\delta(t-t')(4\delta_{ik}\delta_{jl} - \delta_{ij}\delta_{kl} - \delta_{il}\delta_{jk})/3$, where $i, j = 1, 2, 3$. The properties of this stochastic flow allow an exact calculation of $P(\rho)$ (see Refs. [20, 29]):

$$P(\rho) = c\rho^2 \left(1 + \frac{2\text{Wi}b}{3}\rho^2\right)^{-\beta} (1 - \rho^2)^\beta \quad (4)$$

with $\text{Wi} = \lambda\tau$, $\beta^{-1} = 2(b^{-1} + 2\text{Wi}/3)$, and

$$c^{-1} = \frac{\sqrt{\pi}\Gamma(\beta+1)}{4\Gamma(5/2+\beta)} {}_2F_1(3/2, \beta; 3/2 + \beta + 1; -2b\text{Wi}/3). \quad (5)$$

Here Γ and ${}_2F_1$ denote the Gamma and hypergeometric functions, respectively. In this case, the exponent of the power-law region of the PDF is $\alpha = 2\beta - 3 \approx -3(1 - 1/2\text{Wi})$ for $b \gg 1$.

Isotropic turbulence Although useful for a qualitative study of the coil–stretch transition, the BK flow is Gaussian and has zero correlation time. It therefore cannot capture all features of a fully turbulent flow. Thus, we also consider polymers in homogeneous isotropic turbulence. To this end, we use a database of Lagrangian trajectories from a direct numerical simulation (DNS) of the Navier–Stokes equations in a periodic cube at Taylor–microscale Reynolds number $R_\lambda = 111$ (see Refs. [30, 31] for the details). The velocity gradient $\kappa(t)$ is evaluated along 10^4 trajectories and is then inserted in Eq. (1), which is again solved by using Öttinger’s rejection algorithm [26]. The values of the parameters of the dumbbell model in the DNS are $R_0 = 1$ and $L = 18$. The extensibility parameter is $b = (L/R_0)^2 = 18^2$. The effect of thermal noise on the position of the centre of mass is disregarded, since thermal fluctuations are negligible compared to the fluctuations of the turbulent velocity field. The Weissenberg number is again defined in terms of the Lyapunov exponent. Numerical simulations of isotropic turbulence [9, 32] have shown that the core of $P(\rho)$ behaves has a power of ρ , in agreement with the theory of Balkovksy *et al.* [19].

III. RESULTS

Following Sultanov *et al.* [11], we introduce the entropy of the rescaled polymer elongation $\rho = |\mathbf{R}|/L$ as

$$S_\rho = - \int_0^1 P(\rho) \log[P(\rho)] d\rho. \quad (6)$$

The PDF of the extension is normalized as follows: $\int_0^1 P(\rho) d\rho = 1$. The entropy S_ρ is plotted in Fig. 1 (left panel) as a function of Wi for the different flows described in the previous section. In all cases (except for the experimental data) the extensibility parameter is set to a representative value of $b = 18^2$.

In the extensional flow, S_ρ displays a narrow maximum at Wi near critical, *i.e.* the coil–stretch transition is marked by a strong amplification of the entropy of ρ . This behaviour reflects the fact that, at both small and large Wi, the PDF of ρ is dominated by a peak (near to either $1/\sqrt{b}$ or 1), whereas only in a narrow range of Wi around Wi_{cr} the PDF has a broader shape. A large variety of polymer configurations is thus observed at the coil–stretch transition, as can be appreciated by direct inspection of the time series of ρ [8, 33].

In the shear flow, S_ρ starts growing in an appreciable way only when Wi is significantly greater than Wi_{cr} . However, it eventually reaches values higher than for the extensional flow. This is consistent with the distributions of the extensions that have been observed in experiments [22, 23] and numerical simulations [34, 35]. The aforementioned tumbling events indeed entail continuous recoiling and restretching of the polymer. Therefore, fairly large Wi are required to stretch polymers appreciably, and since the tumbling frequency increases with Wi [23–25], the distribution of the extensions becomes broader and broader as Wi grows. A pronounced maximum at extensions comparable to L only forms for Wi as large as 200 [35], and only then is S_ρ expected to start decreasing.

Coming to the random case, S_ρ displays a maximum for both the BK flow and isotropic turbulence. At small and moderate Wi, the two curves are remarkably close despite the idealization of the BK flow. It has indeed been shown in Ref. [36] that the shape of $P(R)$ and the exponent of the power-law intermediate region $P(R) \sim R^{-1-\alpha}$ for $R_0 \ll R \ll L$ are largely insensitive to the correlation time of the flow up to correlation times of the order of λ^{-1} . At large Wi, the behavior differs: S_ρ saturates in the BK flow, whereas it decreases in isotropic turbulence. The reason for this is that if the flow is turbulent and Wi is sufficiently large, $P(R)$ displays a power-law intermediate region together with peak near to L [9]. The development of this sharp peak causes the reduction of S_ρ at increasing Wi. In contrast, such a peak is absent in the BK flow, because a time-decorrelated velocity field is less effective in stretching polymers up to their maximum length [29].

Figure 1 (left panel) also shows a qualitative comparison with the experimental data of Sultanov *et al.* [11]. This comparison requires some caveats. First of all, the experimental points have been translated vertically, which corresponds to using the extensibility parameter b of the dumbbell model as fitting parameter [2, 17]. Indeed, the entropy S_ρ defined from the PDF of the rescaled elongation $\rho = R/L$ can be expressed in terms of the entropy of $P(R/R_0)$ as $S_\rho = S_{R/R_0} - \log(b)/2$, where $S_{R/R_0} = \int P(R/R_0) \log(P(R/R_0)) d(R/R_0)$, therefore a vertical translation of the entropy is equivalent to a change of b . In particular, the observation that $S_\rho^{(dumb)} \simeq S_\rho^{(exp.)} + \Delta S_\rho$ corresponds to fitting the experimental data with a dumbbell with equivalent extensibility $b^{(fit)} = b^{(exp.)} [\exp(-\Delta S_\rho)]^2$. Thanks to this simple relation, the comparison of the entropy curves provides a useful tool to determine the parameter b of the dumbbell model which fits the experimental data. A precise, quantitative comparison between the experiment and the theory is not possible because the Weissenberg number was defined in a different way in the two cases. However, the analysis shows that the experimental data are qualitatively compatible with the entropy of a dumbbell in a random flow with extensibility parameter $b \approx 30^2$. The latter estimate is obtained from the entropy shift $\Delta S_\rho = 0.33$. The corresponding value of the ratio $(L/R_g)^{(fit)} \approx 34.5$ is not far from the experimental value $(L/R_g)^{(exp.)} = 47.8$.

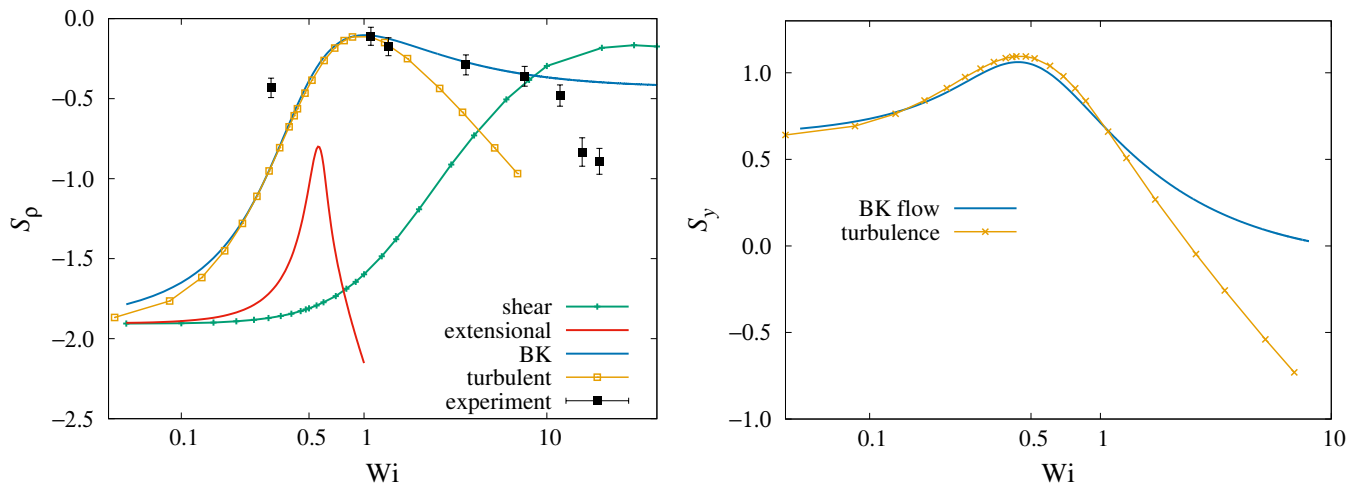


FIG. 1. Left: Entropy of ρ vs Wi for different flows. In all cases (except for the experimental data) the extensibility parameter is set to $b = 18^2$. The experimental data have been translated vertically by $\Delta S_\rho = 0.33$, which corresponds to a fit to a dumbbell with $b \approx 30^2$. Right: Entropy of $y = \ln \rho$ for the BK and turbulent flows and for the same parameters as in the left panel.

Let us now come back to the comparison between the entropy curves in random flows and extensional flows. In both cases, the maximum of S_ρ is an indication of an increased randomness of the polymer configuration in the transitional regime. However, there are some important differences in the behavior of S_ρ observed in random flows with respect to that of extensional flows. First, for a comparable value of Wi the entropy is always greater in random flows. This is because in random flows $P(\rho)$ has a power-law intermediate region and is therefore broader. Second, the maximum of S_ρ is much wider, since in random flows the transition from the coiled to the stretched state is much less sharp [4]. Third, the maximum of S_ρ is located at a value of Wi larger than $Wi_{cr} = 1/2$. To understand this latter point, it is necessary to examine the power-law behaviour of $P(\rho)$.

As mentioned earlier, in random flows the $P(\rho)$ displays a power-law in the intermediate region $1/\sqrt{b} \ll \rho \ll 1$ which scales as $P(\rho) \sim \rho^{-1-\alpha}$, where the exponent α turns from positive to negative at Wi_{cr} . Therefore, at the transition $P(\rho) \sim \rho^{-1}$. Given that α decreases monotonically with Wi , it is rather at $Wi > Wi_{cr}$ that $P(\rho) \sim \rho^0$ and the PDF of ρ is the broadest [9, 29]. Since S_ρ is a measure of the randomness of ρ , it is therefore natural that in random flows S_ρ reaches its maximum value at $Wi > Wi_{cr}$. This fact explains the behavior of S_ρ . However, it also raises the issue of an apparent discrepancy between the critical Wi for the coil–stretch transition and the value of Wi at which S_ρ is maximum. How to reconcile these two different thresholds?

In a random flow, the time-dependent PDF $P(\rho, t)$ satisfies the diffusion equation

$$\frac{\partial P}{\partial T} = \frac{\partial}{\partial \rho} [\rho f(L\rho)P] + \frac{\partial}{\partial \rho} \rho^2 \mathcal{K}(\rho) \frac{\partial P}{\partial \rho}, \quad (7)$$

where statistical isotropy has been assumed, time has been rescaled as $T = t/2\tau$, and the stretching term has been modelled *à la* Richardson via the eddy diffusivity $\mathcal{K}(\rho) = K\rho^2 + b^{-1}$. The coefficient K depends on the the Reynolds and Weissenberg numbers in a way that is specific to the particular random flow. However, its explicit expression is not needed for the discussion below.

Eq. (7) can be recast as a Fokker–Planck equation with drift coefficient $D_1(\rho) = 4K\rho - \rho f(L\rho) + 2/b\rho$ and diffusion coefficient $D_2(\rho) = K\rho^2 + b^{-1}$. The associated Itô stochastic equation is

$$\dot{\rho} = D_1(\rho) + \sqrt{2D_2(\rho)} \xi(t), \quad (8)$$

where $\xi(t)$ is white noise. Note that, for the BK flow, Eqs. (7) and (8) hold exactly with $K = 2Wi/3$ [29]. One important property of Eq. (8) is that the amplitude of the noise depends on ρ . This follows from the fact that if the flow is random, the velocity gradient in Eq. (1) plays the role of a multiplicative noise. However, to be able to use Wi as a control parameter for the coil–stretch transition, it is desirable to move to a representation where the amplitude of the noise is independent of the stochastic variable, *i.e.* a stochastic equation with additive noise only. This is achieved by considering a transformation of variable of the form [37]:

$$y \propto \int \frac{d\rho}{\sqrt{D_2(\rho)}} = \frac{1}{\sqrt{K}} \ln[K\rho + \sqrt{K(K\rho^2 + b^{-1})}] + \text{const.} \quad (9)$$

Around the coil–stretch transition, the coefficient K is $O(1)$. For $\rho \gg 1/\sqrt{b}$ Eq. (9) thus gives

$$y \sim \ln \rho. \quad (10)$$

Now note that the PDF of y is related to that of ρ via the relationship $P(y) \propto \rho P(\rho)$. Therefore, according to the theory of Balkovsky *et al.* [19], at $Wi = Wi_{cr}$ the core of $P(y)$ is flat and the entropy of y ,

$$S_y = - \int P(y) \log[P(y)] dy, \quad (11)$$

is expected to reach its maximum value. This suggests that, for random flows, it may be more appropriate to characterize the coil–stretch transition by measuring the entropy of y rather than that of ρ .

Figure 1 (right panel) shows S_y vs Wi for the BK flow and isotropic turbulence. The experimental data have not been included because calculating $P(y)$ from $P(\rho)$ would require a higher resolution of the small extensions than that available in the experiment [recall that $P(y) \sim \rho P(\rho)$]. As expected, S_y is maximum at $Wi = Wi_{cr}$, which confirms that in random flows S_y provides a convenient characterization of the coil–stretch transition. The differences between the BK flow and isotropic turbulence that have been discussed earlier obviously also manifest themselves in the behaviour of S_y .

IV. SUMMARY AND CONCLUSIONS

In a non-uniform flow, polymers can be highly deformed by the local velocity gradients. However, the statistics of the deformation and the way it varies with Wi depend very sensitively on the properties of the flow. In particular, substantial differences are observed between laminar and random velocity fields. An entropic characterization of the coil–stretch transition has been recently proposed by Sultanov *et al.* [11] for an elastic-turbulence von Kármán flow. We have further developed this approach by examining a set of flows that have been regarded as benchmarks for the study of polymer stretching, in both the laminar and the random case.

This study confirms that the dependence of entropy on Wi provides a useful characterization of the change in the statistics of polymer extension that occurs near the coil–stretch transition. Moreover, it allows a quantitative comparison between flows with different stretching properties. This characterization is particularly relevant to practical situations where limited statistics is available. Entropy is indeed less sensitive to statistical fluctuations than quantities such as the slope of $P(\rho)$ or the correlation time of $\rho(t)$ which have been used previously to describe the coil–stretch transition.

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