 DOI: 10.1002/masy.201450304

Single Polymer Dynamics in A Random Flow

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Summary: The dynamics and conformations of a single fluorescently stained T4 DNA molecule are studied in a random flow of elastic turbulence created by the same unlabeled molecules. The explored polymer concentration covers the dilute and semi-dilute unentangled regimes, according to the measurement of the longest relaxation time by extension relaxation of single polymer chains. The criterion of the coil-stretch transition was found to be close to the theoretically predicted value. Using measured polymer stretching statistics and its known elastic properties, the elastic stress in elastic turbulence is obtained over a broad range of Weissenberg number and polymer concentration. The existing theory of elastic turbulence is disproved by the measurements of the elastic stress and the degree of polymer stretching. The role of increasing shear rate on polymer extension and angular statistics in a random flow is also studied and compared with theory and numerical simulations.

Keywords: conformational analysis; fluorescence; imaging; rheology; single polymer chain

Introduction

Dynamics and conformations of single polymer molecules in flow is considered to be the key ingredient in hydrodynamics of dilute polymer solutions and have remained an outstanding problem in polymer physics for several decades.[1] It is also directly related to the occurrence of drag reduction in turbulent flows, in which the addition of small quantity of high molecular weight polymers to a turbulent pipe flow drastically decrease the drag.[2] The dynamics of single polymer chains in a turbulent flow was widely discussed[3] but complete understanding and in particular its experimental verification are still lacking.

Pioneering by the seminal studies of Chu and collaborators, the dynamics and statistics of single polymer chains in stationary elongation,[4] shear,[5] and mixed[6] flows were extensively studied experimentally. A good agreement with theory[7] and numerical simulation[8–10] was found. In contrast, it is difficult to study single polymer dynamics in a general 3D random flow, due to the difficulty to create a random flow in a microscopic-size volume. A general way to create chaotic or turbulent flow is to work at high velocities, V, and in a large-size vessel, L, to reach large Reynolds numbers, $Re \equiv VL/\nu$, where ν is the kinematic viscosity. Large Re flow cannot be achieved in a hundred micron cell for single molecule experiment. The recently discovered elastic turbulence, a random flow at low Re,[11] opens the door to study the dynamics and conformation of single polymer molecules in a random flow. Long polymer chains added to a fluid make it elastic and can store non-linear elastic energy when they were stretched in a flow. An elastic instability shows up when the elastic energy overcomes the dissipation due to polymer relaxation. The ratio of the elastic term to the relaxation term is defined by the Wissenberg number, $Wi \equiv \lambda V/L$, where λ is the longest polymer relaxation time. In flows of dilute polymer solutions with

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curvilinear trajectories, at Wi above a threshold Wi_c, elastic turbulence, a spatially smooth and temporally random flow, had been observed.[11] The main features of elastic turbulence are: sharp growth in flow resistance, algebraic decay of velocity power spectra over a wide range of scales, and orders of magnitude more effective way of mixing than in an ordered flow.[12] These properties are analogous to hydrodynamic turbulence and make elastic turbulence an ideal system to study single polymer dynamics in a random flow at arbitrarily low Re. The underline mechanism of elastic turbulence is the stretching of polymer by the flow[13] and the feedback of the associated elastic stress to the flow.[14] Recent single polymer experiments proved that there is coil-stretch transition in a random flow,[15–17] which has also been discussed in a number of theoretical[18,19] and numerical studies.[20–22]

Another important case of polymer dynamics and statistics in a random flow with a shear component is studied theoretically and numerically.[23–25] In this case the polymer stretching is described by the following equation:[26]

$$\partial_t x_i = x_j \nabla_j v_i - \lambda^{-1} x_i + \zeta_i \quad (1)$$

where x_i is the polymer end-to-end vector, and ζ_i is the thermal Langevin force. The velocity gradient tensor $\nabla_j v_i$, includes contributions from both the shear component and the random part. In a random flow with a strong shear component, theory predicts that the polymer are mostly stretched in the shear-preferred direction and more complex features of polymer stretching than in the case of isotropic velocity statistics due to molecule tumbling shows up.[24,25]

In this paper, we present the dynamics of single fluorescently stained DNA molecules in a random flow created by a sea of the same unlabeled molecules. Polymer relaxation times by a chain retraction of stretched single polymer molecules are studied over a broad concentration region. Coil-stretch transition above the elastic instability is investigated. The elastic stress in elastic turbulence is obtained over a broad range of Wi and polymer concentration c by using measured polymer stretching statistics and its known elastic properties. Polymer stretching and angular statistics in a random flow with a shear component are studied and compared with theory and numerical studies.

Experimental Part

A stock polymer solution of ~0.3-0.4 mg/mL T4GT7 (T4) DNA with molecular weight of 1.08×10^8 (165.6 kbp) was obtained from Nippon Gene. A T4 DNA solution was prepared by diluting the stock solution by the mixed solvent, a pH8 buffer containing 10 mM tris-HCl, 2 mM EDTA, 10 mM NaCl, 4% β-mercaptoethanol, glucose oxidase (~50 μg/mL), and catalase (~10 μg/mL), with saccharose of different concentration.[15–17] The solution viscosity η as a function of a shear rate $\dot{\gamma}$ was measured on the stress-controlled rheometer AR 1000N (TA Instruments). T4 DNA solutions with concentration c ranged from 6.29 to 100.64 μg/mL (overlap concentration $c^* = 19.2$ μg/mL) were used, and their properties were listed in Table 1.

The experiment was conducted in a setup similar to that used in our previous experiment to study the coil-stretch transition in a random flow of a dilute polymer solution.[15–17] A von Karman swirling flow was created in a gap of $d = 675$ μm between the flat butt of the rotating delrin rod of radius $R = 2.25$ mm ($d/R = 0.3$) and a cover slip. The plastic rod was glued into the

Table 1.
Properties of polymer solutions used in this study.

c (μg/mL)	c (ppm)	% sucrose [a]	η_s (cP) [b]	λ (s)
100.64	87.44	35.0	4.2	8.2
50.32	41.86	45.0	9.0	13.2
25.16	20.47	50.0	14.7	15.5
12.58	9.87	58.0	40.4	37.5
6.29	4.64	71.5	230.0	185.0

[a] In solution of $c = 6.29$ μg/mL, sucrose/sorbitol at a proportion of 1:2 was used. [b] η_s is the solvent viscosity.

metal shaft that was driven via a belt by an optically encoded dc mini-motor with less than 1% rms of velocity variations. Beating of the polished delrin surface due to misalignment was less than 1 μm on a radius of 300 μm. The delrin-made cell had an inner radius $R_c = 6$ mm. The experiments were carried out on area of $267 \times 267\ \mu m^2$ at three radial locations: at $r_1 = 300$ μm, $r_2 = 900$ μm, and $r_3 = 1700$ μm and $h = 100$ μm above the cover glass.

To study the statistics of polymer stretching in a flow of a solution of T4 DNA, the same fluorescently stained molecules with YOYO-1 (Molecular Probes) at dye/base-pair ratio of 1:4 were added into the solution in concentration ~ 1 ng/mL. At equilibrium, the coiled T4 DNA has $x_0 \approx 3$ μm, while its entire contour length is $L \approx 71.7$ μm with roughly 1100 persistence lengths.[27,28] The longest relaxation times λ were measured via relaxation dynamics of stretched polymer molecules for all polymer solutions. Fluorescently stained T4 DNA were imaged using a home-built epi-fluorescent microscope by a 40×, 1.3 NA oil-immersion objectives (Zeiss) with 0.4 μm depth of focus. The fluorescent dye was excited by a 488 nm argon-ion laser reflecting on a 505 nm long-pass dichroic mirror. Fluorescence emanating from the fluorophore went through a 515 nm long pass emission filter before it was recorded with a PhotonMax 512B EMCCD camera (Princeton Instruments) of spatial resolution down to 0.5 μm at 20 or 25 fps. In order to reduce photo induced degradation we stroboscopically illuminated the molecules using a chopper which was synchronized with the camera. The data were recorded in a wide range of angular velocity Ω from 0 up to 15.7 s^{-1}, or in the Weissenberg numbers, $Wi \equiv s\lambda$ from 0 up to about 1000, where $s \equiv \Omega r/d$ is the shear rate. Due to the narrow focal depth, 3D motion of a molecule was traced only during its location in 2D imaging plane, and only those molecules remained totally in a focus were analyzed. For the analysis we used the data below and above the coil-stretch transition up to the highest values of *Wi*. By tracking individual molecules, we measured probability distribution function (PDF) of the polymer extensions x and the out-of-shear-plane inclination angle θ, i. e. the angle between a main polymer axis and a tangent to a polymer trajectory in 2D plane. For sufficiently large number of molecules analyzed, the statistics is representative for all molecules in a 3D random flow. For each *Wi* the statistics of the polymer stretching and orientation angle were based on about 1000 molecules.

The structure and fluctuations of velocity and velocity gradient field in 3D were characterized by using particle image velocimetry (PIV) measurements. The fluorescent particles with diameter of 0.5 μm at a concentration of ~500 ppm were added to polymer solutions as seeding particles.

Results and Discussion

Longest Relaxation Times

The longest relaxation times λ of polymer solutions at different concentration were measured by extension relaxation of single T4 DNA molecules. T4 DNA solution was allowed to relax for more than 10 times of the relaxation time before it was subjected to a shear flow with sufficiently high shear rate, then the flow was abruptly stopped and the conformations of the molecules were recorded as a function of time (inset of Fig. 1). The polymer extension is measured from its conformation and averaged for about 20 molecules.

Figure 1 shows the extension relaxation of T4 DNA in a solution of $c = 25.16$ μg/mL with 50% sucrose. It can be well fitted with a single exponential decay in the region of a relative extension less than 0.3~0.4, which gives the longest polymer relaxation time $\lambda = 15.5 \pm 0.3$ s. The molecule extension relaxation measurement can be performed at extremely low concentration of ~ 1 ng/mL fluorescently labeled T4 DNA in a sucrose solvent, which provides a unique way to measure λ_0, the longest polymer relaxation time at infinite dilution. It should be noted that the relaxation times obtained

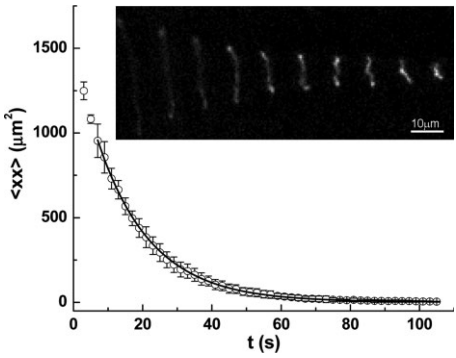

Figure 1.
Extension relaxation of 14 individual T4 DNA molecules averaged together in a solution of $c = 25.16\ \mu g/mL$ with 50% sucrose. The solid line is the fitting to a single exponential decay with $\lambda = 15.5 \pm 0.3$ s. The inset shows the conformations of a single T4 DNA molecule at 5 s intervals.

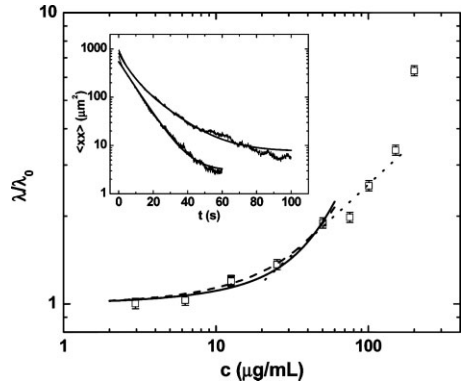

Figure 2.
Concentration dependence of the normalized longest relaxation time of T4 DNA molecules. The solid line is the best fitting for the data at $c < 60\ \mu g/mL$ by $\lambda/\lambda_0 = 1 + cA - \sqrt{2}(cA)^{1.5} + 2(cA)^2$ with $A = 14570$ mL/g, the dash line is the best fitting for the data at $c < 60\ \mu g/mL$ by the exponential growth $\lambda/\lambda_0 = \exp(cA)$ with $A = 12730$ mL/g, and the dot line is the best fitting for the data at $20 < c < 160\ \mu g/mL$ with an exponent 0.48 ± 0.03. The inset shows the extension relaxation of stretched single T4 DNA molecules in T4 DNA solutions with $c = 151\ \mu g/mL$ (bottom curve) and $c = 200\ \mu g/mL$ (top curve) in 30% sucrose; the solid lines are the fittings by a single exponential decay for $c = 151\ \mu g/mL$ and a double exponential decay for $c = 200\ \mu g/mL$, respectively.

by molecule extension relaxation is in good agreement with those obtained from stress relaxation of the bulk solution with a rheometer,[29,30] which shows the inherent correlation between single polymer conformation and its macroscopic solution property.

Studying the concentration dependence of relaxation time of polymer solution provide considerable insight into the polymer dynamics and the role of polymer interactions in different concentration regimes such as dilute, semi-dilute unentangled, and semi-dilute entangled.[31] Figure 2 shows the concentration dependence of the longest relaxation time of T4 DNA molecules after normalization by λ_0. In the dilute regime, the data at T4 DNA concentration $c < 60\ \mu g/mL$ can be well fitted according to the theoretical prediction $\lambda/\lambda_0 = 1 + cA - \sqrt{2}(cA)^{1.5} + 2(cA)^2$ or the exponential growth $\lambda/\lambda_0 = \exp(cA)$ with $A \approx 0.5[\eta]$, where $[\eta] = 29000$ mL/g is the intrinsic viscosity of T4 DNA.[30] At T4 DNA concentration higher than c^*, λ/λ_0 increases nonlinearly. In the semi-dilute unentangled regime of $20 < c < 160\ \mu g/mL$, λ/λ_0 shows a power law dependence with exponent of 0.48. While for the point $c = 200\ \mu g/mL$, an additional slow relaxation mode in the extension relaxation experiment shows up (inset of Fig. 2), indicating it lies in the semi-dilute entangled regime. In the following, the dynamics and conformations of single fluorescently labeled T4 DNA molecules were studied in solutions of T4 DNA concentration c ranged from 6.29 to 100.64 μg/mL, which located in the dilute to semi-dilute unentangled regimes.

Coil-Stretch Transition in a Random Flow

Recent theory indicated that the coil-stretch transition of polymer in a random flow is characterized by the transformation of the PDF of molecular extension as a function of Wi_{loc}, where $Wi_{loc} = \lambda\gamma$ with γ the largest Lyapunov exponent of a random flow and $\gamma \approx (dV_\theta/dr)^{rms}$ in an isotropic random flow.[18,19] According to the theory, the tail of PDF of molecular extension is described by the power law $P(x/L) \sim (x/L)^{-\alpha-1}$ in the vicinity of the

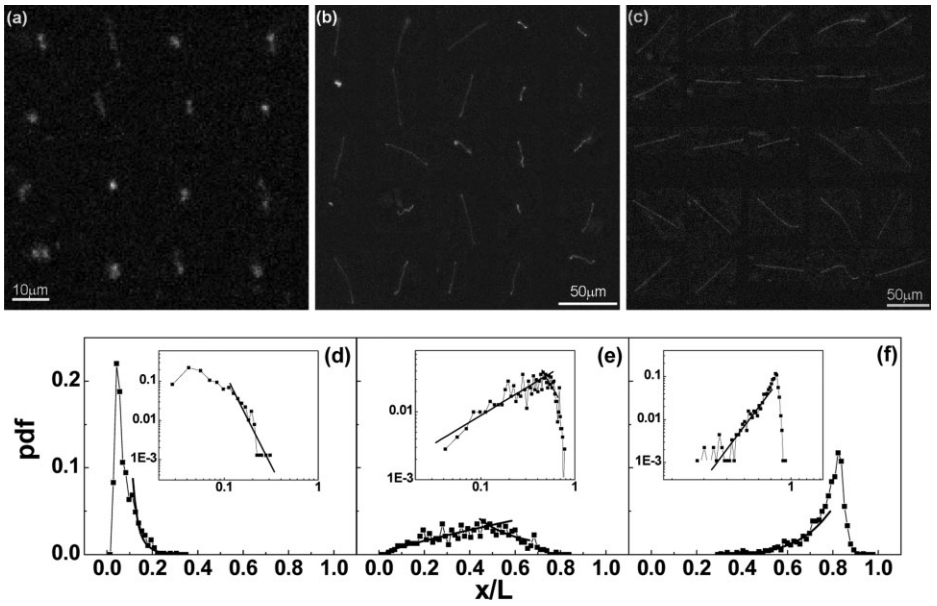

Figure 3.
Typical conformations of single T4 DNA molecules at different $Wi = 2.9$ (a), 8.7 (b), and 108 (c), in a solution of $c = 25.16$ μg/mL with 50% sucrose. The corresponding $P(x/L)$ were shown in (d), (e), and (f), respectively. The inset of (d)-(f) is the same as in the main plot but in log-log presentation. The thick solid lines are algebraic fittings to the PDF tails $P(x/L) \sim (x/L)^{-\alpha-1}$ with $\alpha = 3.8$ at $Wi = 2.9$, $\alpha = -1.8$ (tail on the left side) and 1.3 (tail on the right side) at $Wi = 8.7$, and $\alpha = -5.0$ at $Wi = 108$.

transition.[19] A positive α corresponds to the majority of the polymer molecules being in coiled state, and a negative α is the case when the majority of the molecules is strongly stretched. The condition of $\alpha = 0$ is the criterion for the coil-stretch transition. To study the coil-stretch transition of T4 DNA, the molecular extension in a polymer solution of $c = 25.16$ μg/mL with 50% sucrose were measured at different Wi below and above the elastic instability threshold $Wi_c = 5.8$. Figure 3 shows the typical polymer conformations of single T4 DNA molecules at $Wi = 2.9$, 8.7, and 108, and the obtained PDF of the relative molecular extension x/L. At $Wi = 2.9$, T4 DNA molecules are weakly stretched with most of the molecules in coiled state, and a positive α of 3.8 is obtained. At $Wi = 108$, T4 DNA molecules are strongly stretched with a negative α of -5.0. At $Wi = 8.7$ close to the coil-stretch transition, the PDF has a broad distribution and α approaches 0.

The value of the rms of the velocity gradient fluctuation, $(dV_\theta/dr)^{rms}$, is obtained by PIV measurement of the same polymer solution at different Wi. In Figure 4, we plot the obtained α as a function of $1/Wi_{loc}$. The linear fitting to the data intersects with $\alpha = 0$ at the critical value of $Wi_{loc,cr} = 0.70 \pm 0.20$. This criterion is close to the theoretical predicted value and the numerical simulation,[18–22] as well as the previous experiments on λ DNA.[15]

Polymer Stretching as Elastic Stress Sensor in Elastic Turbulence

Having measured the PDF of polymer stretching at different Wi, an attempt to quantify the elastic stress in elastic turbulence was made. From the measured PDF of fractional extension x/L (Fig. 3 and left inset of Fig. 5), the peak value of the PDF is obtained and plotted as a function of Wi/Wi_c for 5 T4 DNA concentrations, see Figure 5. The value of x_p/L increases with Wi and saturates at about 0.7–0.9

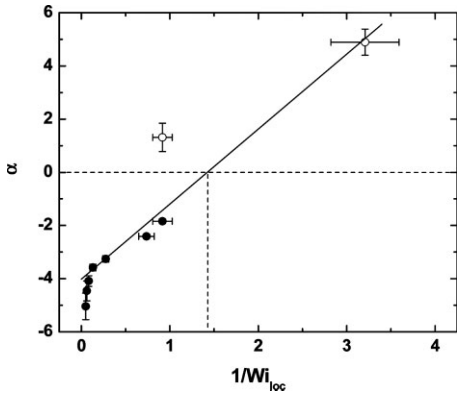

Figure 4.
Exponent α in $P(x/L) \sim (x/L)^{-\alpha-1}$ as a function of $1/Wi_{loc}$. Open points are the data before coil-stretch transition, and solid points after it. The solid line is the fitting to the data and the dash lines shows the transition at $\alpha = 0$.

depending on c at $Wi/Wi_c > 10$. At the same value of Wi/Wi_c, the molecular extension becomes higher for lower polymer concentration (right inset of Fig. 5). At $c = 6.29$ μg/mL and extremely high $Wi = 1033$, due to very high solvent viscosity and the longest relaxation time, T4 DNA is significantly overstretched (up to 30%) that is rather unexpected in a flow. The overstretched transition had been observed for DNA molecules up to 1.7 times of its contour length at the longitudinal external force of ~65 pN.[32,33] In a random flow, two mechanisms limiting the polymer stretching above the coil-stretch transition were analyzed theoretically.[18,19] The first one is based on the nonlinear elasticity of a polymer, where polymer can be stretched up to L with a stationary state at each Wi.[18] The second mechanism is based on the feedback reaction of the stretched polymers with linear elasticity on the flow.[19] The dynamic balance between polymer stretching and the flow results in $x_{back}/L << 1$. The observed $x_{back}/L \approx 0.7$-0.9 in elastic turbulence seems to disapprove the mechanism based on feedback of polymers with linear elasticity (we will discuss this point further below).

To calculate the elastic stress generated in elastic turbulence by stretched DNA molecules, we used an improved approximation of the worm-like chain model:[34,35]

$$\frac{FA}{kT} = \frac{1}{4(1-x/L)^2} - \frac{1}{4} + \frac{x}{L} + \sum_{i=2}^{7} a_i \left(\frac{x}{L}\right)^i \quad (2)$$

where F is the force, $A \approx 66$ nm is the persistence length of fluorescently stained DNA,[36] k is the Boltzman constant, T is the temperature, and a_i are the coefficients of the seven-order polynomial.[35] Figure 6

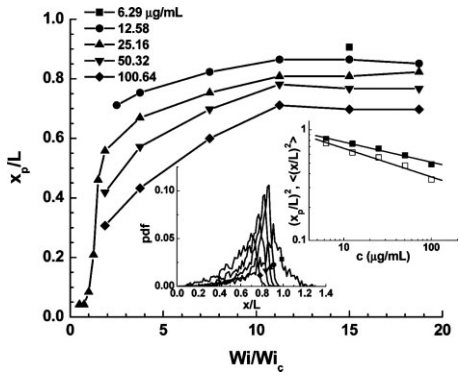

Figure 5.
Peak fractional extension of T4 DNA, x_p/L, as functions of Wi/Wi_c for 5 T4 DNA concentrations. Left inset: $P(x/L)$ at the highest Wi for 5 concentrations. Right inset: square of the peak fractional extension, $(x_p/L)^2$ - solid square, and average of the square of the fractional extension, $\langle (x/L)^2 \rangle$ - open square, as functions of c.

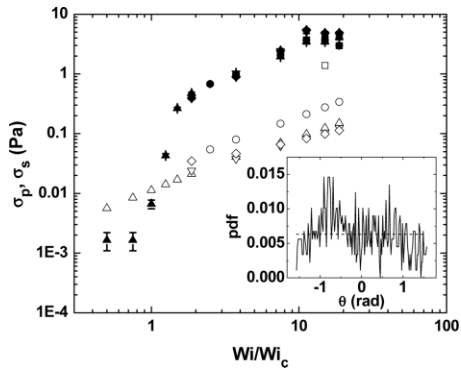

Figure 6.
Elastic σ_p (full symbols) and viscous σ_s (open symbols) stresses as functions of Wi/Wi_c for 5 polymer concentrations (symbols as in Fig. 5). The inset is the PDF of inclination angle θ at $Wi/Wi_c = 7.5$ and $c = 25.16$ μg/mL. The dash line is the average uniform PDF.

presents the elastic stress calculated from the peak of the PDF of molecular extension x_p averaged over a uniform distribution of an in-plane inclilation angle (see inset of Fig. 6) $\sigma_p = <cN_A F x_p/M_w>$ as a function of Wi/Wi_c for all c, where N_A is Avogadro number, M_w is the molecular weight. For comparison, the viscous stress $\sigma_s = \Omega\eta r/d$ is also plotted, where η is the solution viscosity. It is found that σ_p increases by 2-3 orders of magnitude over the studied Wi and reaches up to ~ 5 Pa, which is up to 60 times of σ_s in elastic turbulence. The saturated value of elastic stress σ_p^s is about the same for all c. This is in good agreement with the macroscopic mechanical measurement where polymer stress in elastic turbulence is found to be ~ 170 times higher than in a laminar flow.[13]

To compare with the theory of elastic turbulence, the normalized elastic stresses $\sigma_p/(\eta/\lambda)$ are plotted as functions of Wi_{loc} for five values of c in Figure 7. The same linear relation $\sigma_p/(\eta/\lambda) = 130 Wi_{loc}$ in a wide range of Wi_{loc} from 2 to 30 was found for all c. At the highest values of Wi_{loc}, $\sigma_p/(\eta/\lambda)$ saturates for each c at the same value, as shown in the inset of Fig. 7. The theory of elastic turbulence, based on polymers with linear elasticity and the feedback reaction on the flow, provides an estimation of the Wi_{loc} and σ_p in a bulk flow.[14,19] The theory is based on two assumptions: i) the statistics of the velocity field and polymer stretching in elastic turbulence is stationary due to the feed back reaction of the polymers on the flow that results in $(dV_\theta/dr)^{rms}$ of the order of λ^{-1} in the bulk of elastic turbulence; ii) both the elastic stress σ_p/λ and the viscous stress $\eta[(dV_\theta/dr)^{rms}]^2$ are of the same order.[14] From ii) one gets $\sigma_p/(\eta/\lambda) \sim Wi_{loc}^2$, whereas i) gives $Wi_{loc} \sim 1$. Then both assumptions leads to $\sigma_p/(\eta/\lambda) \sim 1$, resulting $\sigma_p \approx 0.01$ Pa at the most. These are not consistent with the experimental results presented in Figs. 6 and 7: both Wi_{loc} and σ_p increase with Wi and saturate at much higher values in elastic turbulence than theory predicted. This is a consequence of polymer being stretched into the nonlinear regime and the current theory[14] of elastic turbulence based on feedback reaction of stretched polymers with linear elasticity on the flow does not work any more. It is also supported by the dependence of either $(x_p/L)^2$ or $<(x/L)^2>$ on c (right inset of Fig. 5). Since in the case of a polymer with linear elasticity, $\sigma_p \propto c<(x/L)^2>$, one gets $<(x/L)^2> \propto c^{-1}$ in the elastic-turbulence regime with the same σ_p. However, in the right inset of Fig. 5, the exponent in this relation is found to be about -0.26. By varying c one varies the degree of polymer stretching, which is nonlinearly related to the polymer elastic force. So to get the same σ_p, the smaller value of the exponent is sufficient, which is a consequence of the polymer being stretched into the nonlinear regime. Therefore, a new theory of elastic turbulence based on feedback reaction on the flow of stretched polymers with nonlinear elasticity should be developed.

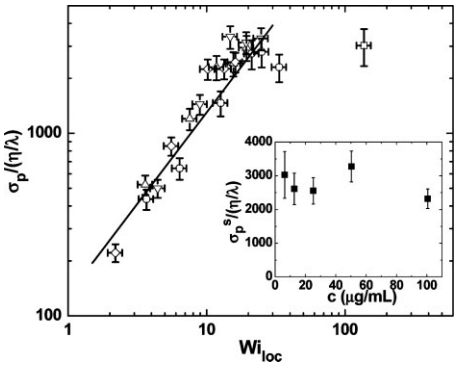

Figure 7.
Normalized elastic stress as functions of Wi_{loc} for 5 values of c (symbols as in Fig. 5). The inset is the saturated normalized elastic stress as a function of c.

Effect of Shear Rate on Polymer Extension and Angular Statistics

We have characterized the structure and fluctuations of velocity and velocity gradient field of elastic turbulent flow in 3D by PIV measurements. It is found that $\Lambda \equiv (dV_\theta/dz)^{rms}/s$, the ratio between the random part to the shear component in elastic turbulence, changed from an order of unity at $r_1 = 300\,\mu$m till small values at $r_2 = 900\,\mu$m and $r_3 = 1700\,\mu$m. Therefore by changing r besides Wi, one can study the

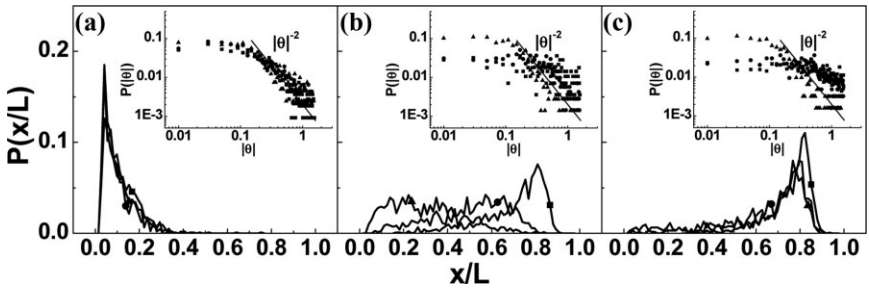

Figure 8.
$P(x/L)$ at various Wi for three locations r_1 (square), r_2 (circle), and r_3 (up-triangle). (a) $Wi = 4.3$ for $r_1$and r_2, and $Wi = 4.1$ for r_3; (b) $Wi = 65$ for r_1 and r_2, and $Wi = 61$ for r_3; (c) $Wi = 87$ for r_1, $Wi = 260$ for r_2, and $Wi = 491$ for r_3. The insets are the corresponding $P(|\theta|)$.

effect of shear rate on polymer extension and angular statistics in a random flow.

Figure 8 shows the evolutions of PDFs of polymer extension $P(x/L)$ and the angular PDFs $P(|\theta|)$ at three locations r_i for various Wi in a solution of $c = 25.16\ \mu g/mL$ with 50% sucrose. At low Wi =4.3 below the elastic instability threshold (Fig. 8 (a)), both $P(x/L)$ and $P(|\theta|)$ at three locations are very close. This is the case for a simple shear flow with observed $P(|\theta|) \sim |\theta|^{-2}$, which agrees with the theory.[25]

At higher $Wi = 65$ (Fig. 8(b)), $P(x/L)$ and $P(|\theta|)$ are significantly different at three locations. At $r_1 = 300\ \mu m$, a well-defined peak in $P(x/L)$ at $x_p/L \sim 0.8$ is observed with a uniform distribution of the orientation angle θ, which is expected for a random flow with neglected shear contribution. At $r_3 = 1700\ \mu m$, $P(x/L)$ is still peaked close to the coiled state with $P(|\theta|) \sim |\theta|^{-2}$, which is a result from the strong shear component than random part at this position. At $r_2 = 900\ \mu m$, both $P(x/L)$ and $P(|\theta|)$ show behaviors in between: $P(x/L)$ has a peak $x_p >> x_0$, and $P(|\theta|)$ is constant in a wide range of angles with the decay of $\sim |\theta|^{-2}$ in a narrow range at high angles. Therefore, depending on the contribution from the shear and random part of the flow, the difference in PDFs of polymer extension and angular statistics is obvious.

At the highest values of Wi (Fig. 8(c)), T4 DNA molecules are stretched at all three locations. However, a flat plateau at r_3 is evident, which is the sign of deterministic dynamics (tumbling) due to shear. The strong shear component at r_3 significantly broadens the polymer extension distribution compared with the cases without or with small shear. A decay of $P(|\theta|) \sim |\theta|^{-2}$ at r_3 is also resulting from the tumbling of molecules at strong shear, in contrast to the random distribution of θ in a random flow at r_1. These experimental findings are in good agreement with theory and numerical simulations.[23–25]

To compare further a polymer extension in a mixed flow and a random flow with a shear component, the reduced averaged polymer extension $<x>/L$ as a function of Wi at the three locations are shown in the inset in Figure 9. By assuming that the random flow with the shear component is

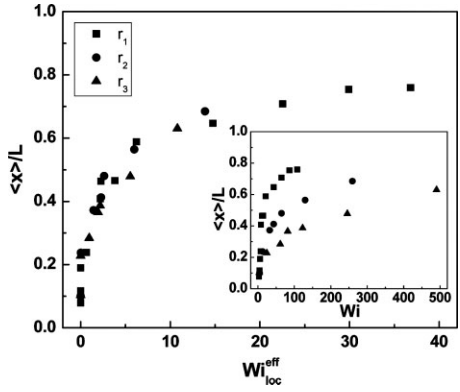

Figure 9.
$<x>/L$ as a function of Wi_{loc}^{eff}. Inset: $<x>/L$ as a function of Wi at three positions.

similar to the mixed flow with $(dV_\theta/dz)^{rms}$ representing the elongation rate E_{el}, and with the shear rate $s \equiv E_s + \omega = <dV_\theta/dz>$ as in a shear flow part. One can introduce $\beta = \Lambda/(1+\Lambda)$ analogous to the mixed flow,[10] then for a random flow with a shear component, the rescaled $Wi_{loc}^{eff} = Wi_{loc}\beta^{\delta}$ instead of Wi or Wi_{loc} should be used to collapse the data for $<x>/L$. As shown in Figure 9, the data are collapsed on the universal plot with $\delta = 0.65 \pm 0.01$ obtained by minimization of the rms deviations of the data points from the fitting function. However, the explanation for the value of δ to the flow properties is still lacking, in contrast to the mixed flow.[10]

Conclusion

To summarize, the dynamics and conformations of single fluorescently stained T4 DNA molecules in a random flow created by the same unlabeled molecules are studied in details. The explored polymer concentration covers the dilute and semi-dilute unentangled regimes, as confirmed by the concentration dependence of the longest relaxation time determined by extension relaxation of single polymer chains. Coil-stretch transition above the elastic instability is investigated and the criterion of the transition was found to be in good agreement with theoretical studies and numerical simulations. The elastic stress in elastic turbulence is obtained over a broad range of Wi and c, by using measured polymer stretching statistics and its known elastic properties. The existing theory of elastic turbulence is disproved by the measurements of the elastic stress and the degree of polymer stretching. The role of increasing shear rate on polymer extension and angular statistics in a random flow is also studied and found to agree with theory and numerical simulations. The results on the conformations and dynamics of single polymer chain and the associated elastic stress in a spatially smooth and random-in-time flow, provide important information to develop new theory based on different assumptions and should help us to understand the long-standing turbulent drag reduction problem.

Acknowledgement: This work is partially supported by the grants of the Israel Science Foundation, the Minerva Foundation, the Lower Saxony Ministry of Science and Culture Cooperation Grant, and the Minerva Center for Nonlinear Physics of Complex Systems. YL is supported by the grants of the MPG-CAS Partner Group Program, the National Natural Science Foundation of China (21274147), and the Natural Science Foundation of Jilin Province, China (201215093).

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