Bulk turbulence in dilute polymer solutions

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By tracking small particles in the bulk of an intensely turbulent laboratory flow, we study the effect of long-chain polymers on the Eulerian structure functions. We find that the structure functions are modified over a wide range of length scales even for very small polymer concentrations. Their behaviour can be captured by defining a length scale that depends on the solvent viscosity, the polymer relaxation time and the Weissenberg number. This result is not captured by current models. Additionally, the effects we observe depend strongly on the concentration. While the dissipation-range statistics change smoothly as a function of polymer concentration, we find that the inertial-range values of the structure functions are modified only when the concentration exceeds a threshold of approximately 5 parts per million (p.p.m.) by weight for the 18×10^6 atomic mass unit (a.m.u.) molecular weight polyacrylamide used in the experiment.

1. Introduction

Minute amounts of long-chain, flexible polymers added to a fluid can strongly modify its flow properties. In a turbulent wall-bounded flow, for example, polymer additives lead to the extraordinary phenomenon of drag reduction (Toms 1948; Virk et al. 1967; Lumley 1973; Sreenivasan & White 2000; L'vov et al. 2004; Bonn et al. 2005). In slow, laminar flows, elastic turbulence strongly enhances fluid mixing (Groisman & Steinberg 2000, 2001). These effects may be qualitatively attributed to the stretching of polymer molecules by the velocity gradients in the flow. Although progress has recently been made in understanding drag reduction at a wall, comparatively little is known about the action of polymers far from the boundaries of a turbulent flow (Liberzon et al. 2005, 2006; Crawford et al. 2002, 2008; Crawford 2004). In particular, the role of the polymer concentration is poorly understood, though threshold concentrations for the onset of elastic effects have previously been observed in elastic turbulence (Groisman & Steinberg 2001), using the same polymer used in the present experiments.

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In a Newtonian fluid, viscosity provides the sole mechanism for the dissipation of energy. In this case, the classical Richardson–Kolmogorov cascade hypothesis (Kolmogorov 1941) states that energy is injected into the flow at large length and time scales, transferred through the inertial range of scales without loss and finally dissipated at the smallest scales at which viscosity acts. The addition of long-chain polymer molecules to the fluid, however, provides a new mechanism for energy dissipation. As suggested by Tabor & de Gennes (1986), when the polymers are stretched by the flow, they store elastic energy. In a turbulent flow, the polymer molecules continually stretch and recoil in the fluctuating flow field, dissipating turbulent kinetic energy through the interactions between the monomers of a single polymer molecule and between the polymers and the fluid. The turbulent energy cascade may therefore be modified in a polymer solution, irrespective of any drag reduction at the walls.

Newtonian turbulence is characterized solely by the Reynolds number Re. When Re is large, eddies spanning a wide range of length and time scales participate in the energy cascade. A polymer solution is additionally characterized by both the polymer concentration ϕ and the Weissenberg number Wi, which compares τ_p , the longest relaxation time of a single polymer molecule, to the fastest flow time scale. For turbulent flows, therefore, $Wi = \tau_p/\tau_\eta$, where τ_η is the Kolmogorov time scale. We note that Wi is sometimes defined using either the flow Lyapunov exponent or the root-mean-square gradient; since we cannot measure either of these quantities, however, we use τ_η to characterize the flow. When Wi is less than a critical value (of order unity), the polymer molecules are generally in their coiled state and are passively advected by the flow. When Wi is larger than this value, however, the polymers will, on average, be stretched by the flow and may modify it (Lumley 1973).

The effects of polymers on the properties of bulk turbulence have been investigated previously, though questions still remain. Several studies have found that polymers damp turbulence in the bulk of the flow, particularly at small scales (McComb, Allan & Greated 1977; Tong, Goldburg & Huang 1992; Bonn et al. 1993; van Doorn, White & Sreenivasan 1999). In these experiments, however, turbulence was forced through boundary-layer interactions: McComb et al. (1977) and van Doorn et al. (1999) studied grid-generated turbulence; Tong et al. (1992) used a Couette cell; and Bonn et al. (1993) used a counter-rotating-disc flow apparatus with smooth discs. In each of these cases, polymer drag reduction at the forcing element may have played a role in the observed dynamics. In contrast, in a counter-rotating-disc system with baffled discs, which forces the turbulence inertially, Cadot, Bonn & Douady (1998) found that the energy injection rate was no different for pure water and a dilute polymer solution. By using Lagrangian particle tracking in a low-Re inertially forced experiment, Liberzon et al. (2005, 2006) found that the stretching rates of fluid elements are lower in polymer flows (Liberzon et al. 2005), and through the same method they also obtained direct evidence for an additional energy dissipation term due to the polymers (Liberzon et al. 2006). At much higher Re and in the same apparatus used in the present work, polymers were found to decrease the Lagrangian acceleration variance (Crawford et al. 2002, 2008; Crawford 2004).

Isotropic turbulence in polymer solutions has also been numerically studied using model equations for the polymers. Such simulations are, however, very challenging, due both to the nature of the equations and to numerical instabilities (Vaithianathan *et al.* 2006). De Angelis *et al.* (2005) found that polymers both reduced the energy dissipation rate with respect to an identical Newtonian fluid and interrupted the

energy cascade. Similar results were also found in the simulations of Perlekar, Mitra & Pandit (2006). In very highly resolved simulations, Davoudi & Schumacher (2006) studied the stretching of model polymers in a turbulent shear flow. Using similar model equations, Casciola & De Angelis (2007) recently extended the traditional Kármán–Howarth and Kolmogorov equations to polymeric turbulence.

We have studied the second-order Eulerian velocity structure functions, simple probes of the energy cascade, in an inertially forced counter-rotating-disc flow, as described in § 2. We investigate the flow far from the walls of our chamber; therefore, we study not drag reduction but rather the effects of polymer additives on the turbulent bulk. As shown in § 3, we find that even very small polymer concentrations change the turbulence statistics over a wide range of scales. In § 4, we show that the concentration plays a key role in determining the effect of the polymers on the energy cascade: for small ϕ , the inertial-range statistics are unchanged, and only the smallest scale statistics are modified, while for larger ϕ , both the dissipation-scale and inertial-range statistics change. In § 5, we summarize our results and offer some suggestions for further research.

2. Experimental methods

As described previously (La Porta et al. 2001; Voth et al. 2002; Bourgoin et al. 2006; Ouellette et al. 2006b), we generate turbulence in a water flow between two counter-rotating baffled discs. Flow properties are measured by tracking (Ouellette, Xu & Bodenschatz 2006a) the simultaneous motion of hundreds of nearly neutrally buoyant 33 µm fluorescent polystyrene tracer particles. The particles are excited by a high-power pulsed Nd:YAG laser delivering up to 90 W, and their images are recorded with three Phantom v7.1 CMOS cameras from Vision Research (Wayne, NJ).

The polymer used was an 18×10^6 atomic mass unit (a.m.u.) molecular weight polyacrylamide (Polysciences 18522) with an equilibrium radius of gyration $R_g = N^{3/5}a = 0.5 \,\mu\text{m}$, where N is the number of monomers and a is the length of a monomer, a fully stretched length of 77 μ m and a relaxation time of $\tau_p = R_p^3 \mu/(k_B T) = 43$ ms, where μ is the dynamic viscosity, k_B Boltzmann's constant and T the temperature (Crawford 2004). We make the assumption that the polymers do not interact with our tracer particles under the experimental conditions. While our apparatus allows us to reach very high Reynolds numbers, the size and flexibility of the polymer molecules make them prone to mechanical degradation in intense turbulence (Groisman & Steinberg 2004). We therefore only consider Reynolds numbers at which our results are not affected by polymer degradation (Crawford et al. 2002; Crawford 2004). In our experiments, Wi ranges from 1.2 to 6.0; the Taylor-microscale Reynolds number $R_{\lambda} \equiv \sqrt{15u'L/\nu}$ ranges from 200 to 350 (measured in pure water), where u' is the root-mean-square turbulent velocity, L the integral velocity scale and ν the kinematic viscosity. We find that $L \approx 7 \,\mathrm{cm}$ does not change over the range of R_{λ} considered here. In our experiments Wi and R_{λ} are coupled, since $R_{\lambda} = \sqrt{15}T_L/\tau_{\eta} = \sqrt{15}T_LWi/\tau_{p}$, where T_L is the large-eddy turnover time. We varied the polymer concentration ϕ from 0 (pure water) to 20 parts per million (p.p.m.) by weight. At such low concentrations, the kinematic viscosity changes only weakly from its water value (Arratia, Voth & Gollub 2005). We also find that u' varies only weakly with ϕ , as shown in figure 1. Additionally, the inherent large-scale anisotropy of our flow is not significantly affected by the polymers.

It is possible that R_{λ} may change upon the addition of polymers, since they modify the small scales of the flow. We find, however, that the energy injection

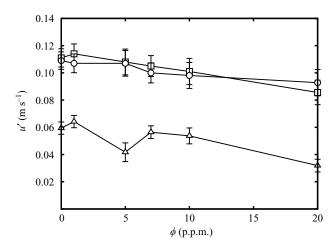


FIGURE 1. The velocity u' as a function of ϕ for $R_{\lambda}^w = 350$. All three components of velocity are shown: u'_x (\square) and u'_y (\bigcirc) are the two in-plane components, while u'_z (\triangle) is the axial component.

(estimated from the large-scale velocity) changes only weakly with such small polymer concentrations, as was also observed by Cadot *et al.* (1998) in a similar apparatus. Additionally, Liberzon *et al.* (2005) found that R_{λ} changed only weakly at the polymer concentrations we use. We therefore choose to base R_{λ} on the large scales and report its value as measured in pure water for a given rotation rate of the discs (Voth *et al.* 2002; Ouellette *et al.* 2006b). Additionally, since we cannot measure the Kolmogorov scales directly, we infer them from scaling arguments based on statistics obtained in pure water solutions. As a mnemonic, we therefore write the Kolmogorov length scale as η_w and the Reynolds number as R_{λ}^w .

Eulerian velocity structure functions have traditionally been used as scale-local probes of the energy cascade in Newtonian turbulence. They are defined to be the moments of the spatial velocity differences over a separation r, $\delta u(r) = u(x+r) - u(x)$. These velocity differences may be thought of as coarse-grained, scale-local gradients. In isotropic turbulence, the second-order structure function tensor can be parameterized by two scalar functions: a transverse structure function $D_{NN}(r)$, measuring the difference in the velocity component orthogonal to r, and a longitudinal structure function $D_{LL}(r)$, where the velocities are taken parallel to r. Here, we show only measurements of $D_{NN}(r)$; our results for $D_{LL}(r)$ are similar.

In isotropic Newtonian turbulence, we expect three distinct scaling ranges for $D_{NN}(r)$. In the dissipation range $(r \ll \eta)$,

$$D_{NN}(r) = \frac{2}{15} \frac{\epsilon r^2}{\nu} \quad (r \ll \eta). \tag{2.1}$$

In the inertial range, according to the Kolmogorov (1941) theory,

$$D_{NN}(r) = \frac{4}{3}C_2(\epsilon r)^{2/3} \quad (\eta \ll r \ll L),$$
 (2.2)

where C_2 is expected to be a universal constant (Sreenivasan 1995). Finally, at large scales $(r \gg L)$, $D_{NN}(r)$ saturates at twice the velocity variance. There may, of course, be intermittency corrections to this scaling law, but they are weak for second-order statistics (Frisch 1995) and do not appreciably change our results. We note that even

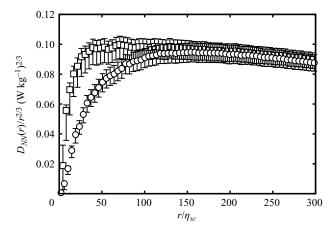


FIGURE 2. The values of $D_{NN}(r)/r^{2/3}$ for both water (\square) and a 5 p.p.m. polymer solution (\bigcirc) at $R_{\lambda}^{w}=290$ (Wi=3.5); r is scaled by η_{w} , the Kolmogorov scale computed from the water data. The polymer data reach an inertial-range plateau with the same value as the water data but at a larger length scale.

though our flow is not isotropic, we observe pseudo-isotropic scaling by spherically averaging our structure functions (Eyink 2003). Additionally, as we have a complex large-scale mean flow in our apparatus, we subtract a time-averaged velocity field from our measured instantaneous velocities before computing structure functions from the velocity fluctuations.

3. Eulerian structure functions

The scaling relations presented above were derived for Newtonian turbulence; we expect, however, that the scaling of the structure functions may be similar for dilute polymer solutions. In figure 2, we plot $D_{NN}(r)/r^{2/3}$ at $R_{\lambda}^{w}=290$ for both pure water and a $\phi=5$ p.p.m. polymer solution. Plotted in this way, we observe the expected inertial-range plateau, where $D_{NN}(r) \sim r^{2/3}$. Both data sets show such a plateau, providing an a posteriori confirmation of our hypothesis that the scaling of the structure function does not change in the polymer solution. We observe, though, that the plateau occurs at a larger length scale in the polymer solution. At larger scales, the polymer data collapse onto the water data, suggesting that the large scales of the turbulence are not significantly modified by the presence of the polymer, at least at such a low concentration.

The scale r_p at which the polymer data in figure 2 returns to the water data is very large compared with the size of the polymer molecule. For $R^w_\lambda=290$ and $\phi=5$ p.p.m., we find that $r_p\approx 150\eta_w=16.6$ mm, more than 200 times the fully stretched length of a polymer molecule. This result suggests that r_p is a dynamical scale rather than one associated with the physical size of the polymers. In pioneering work, Lumley (1973) suggested that the length scale of any polymer effect should be determined purely by the time scale on which the polymer recoils. This (inertial-range) length scale $r^*=(\epsilon\tau_p^3)^{1/2}$ is the scale at which the local Wi is unity and at which we expect the polymers to begin to be stretched by the flow; r^* may also be obtained by using a Kolmogorov-style argument and assuming that the polymer couples with the flow only through its time scale. Since $r^*\sim\sqrt{\epsilon}$ and therefore scales with R^w_λ , we tested Lumley's (1973) 'time criterion' hypothesis by varying R^w_λ while fixing ϕ ; r^* is expected to

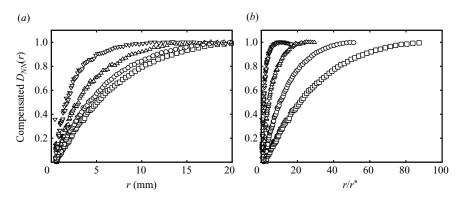


FIGURE 3. Reynolds number effects at fixed concentration; $D_{NN}(r)$ (compensated so that it is unity in the inertial range) is shown for four values of R_{λ}^{w} (\square : $R_{\lambda}^{w} = 200$; \bigcirc : $R_{\lambda}^{w} = 240$; \triangle : $R_{\lambda}^{w} = 290$; ∇ : $R_{\lambda}^{w} = 350$) with $\phi = 5$ p.p.m. (a) r_{p} (in physical units) decreases with increasing R_{λ}^{w} . (b) Scaling by the Lumley scale r^{*} does not collapse the data for different R_{λ}^{w} . Data are shown for the same range of r in each panel.

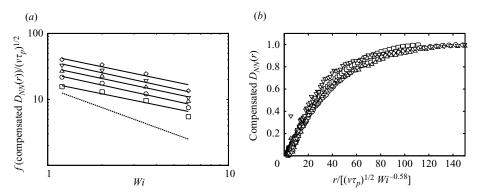


FIGURE 4. (a) Fractions f of the compensated $D_{NN}(r)$ scaled by $(\nu\tau_p)^{1/2}$ as a function of Wi. From bottom to top, $f=0.2,\ 0.3,\ 0.4,\ 0.5$ and 0.6. The solid lines are fits of a Wi^n power law to the data. Averaging over different values of f, we find that $n=-0.58\pm0.09$. The dashed line shows a reference n=-1 power law, for comparison with the model of Fouxon & Lebedev (2003). (b) The compensated structure functions scaled by $(\nu\tau_p)^{1/2}Wi^{-0.58}$. The collapse is significantly better than that shown in figure 3(b).

increase with R_{λ}^{w} . As shown in figure 3(a), however, r_{p} appears to *decrease* (in physical units) as R_{λ}^{w} increases. Therefore, as shown in figure 3(b), $r_{p} \neq r^{*}$; that is Lumley's (1973) theory does not appear to apply to polymer effects in the bulk of the flow.

Instead, since the typical size of the polymer molecules lies in the dissipation range, we propose a length scale that includes viscous effects to capture the behaviour of the structure functions. We note that $(\nu\tau_p)^{1/2}$ is the viscous length scale for the polymers, just as $(\nu\tau_\eta)^{1/2} = \eta_w$ is the viscous length scale for the turbulence. We expect that a combination of this scale with Wi, which describes the mean stretching by the flow, will capture the effect we see. We assume that $r_p \sim (\nu\tau_p)^{1/2}Wi^n$. As shown in figure 4(a), by fitting many fractional cuts of the compensated structure function scaled by $(\nu\tau_p)^{1/2}$, we find that $n=-0.58\pm0.09$. Figure 4(b) shows the compensated structure functions with r scaled by $(\nu\tau_p)^{1/2}Wi^{-0.58}$; the data collapse much better than in figure 3(b).

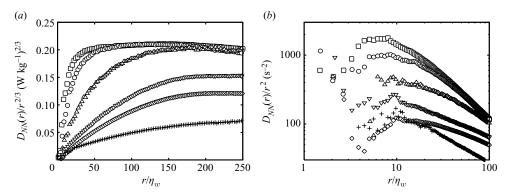


FIGURE 5. The effect of polymer concentration on $D_{NN}(r)$ at $R_{\lambda}^{w}=350$ (Wi=6.0, $\eta_{w}=84$ µm). Data are shown for $\phi=0$ p.p.m. (\Box ; pure water), 1 p.p.m. (\bigcirc), 5 p.p.m. (\triangle), 7 p.p.m. (∇), 10 p.p.m. (\triangle) and 20 p.p.m. (+). In (a), $D_{NN}(r)$ is scaled by $r^{2/3}$, its inertial-range scaling prediction. In (b), we scale instead by r^{2} , as predicted for the dissipation range.

Our result is not predicted by current theory. Fouxon & Lebedev (2003) have predicted that turbulence in a polymer solution supports elastic waves, similar to Alfvén waves in a plasma, for the range of scales $\eta^* \ll r \ll r^*$, where $\eta^* = \eta W i^{-1/2}$. Rewriting our result, we have $r_p = \eta W i^{n+1/2}$. For r_p to be equivalent to η^* , we would need n = -1; since instead we find $n = -0.58 \pm 0.09$, r_p is different from η^* . It is unlikely that the Fouxon & Lebedev (2003) theory applies to our system; our result therefore awaits theoretical explanation.

4. Concentration effects

We now explore the concentration dependence of the polymer effects by fixing R_{λ}^{w} and varying ϕ . In figure 5(a), we show $D_{NN}(r)/r^{2/3}$ at $R_{\lambda}^{w}=350$ (Wi=6.0) for ϕ ranging from 0 p.p.m. (pure water) to 20 p.p.m. For small ϕ (\leq 5 p.p.m. in our experiments), the shape of $D_{NN}(r)$ does not change in the presence of polymers: the curves reach an inertial-range plateau with a value that is independent of ϕ , though r_{p} does change with ϕ . At higher ϕ (\geq 7 p.p.m.), however, the apparent inertial-range plateau is decreased relative to the water data, though the data still appear to scale as $r^{2/3}$ above r_{p} . These results suggest that there is a critical concentration ϕ_{c} below which the Newtonian scaling constant $(4/3)C_{2}\epsilon^{2/3}$ in unaffected by the polymers. Above ϕ_{c} , either C_{2} must change, which would imply a breakdown of turbulent universality, or ϵ must be decreased, implying that less energy is transferred to small scales than in the traditional energy cascade. Determining which of these possibilities is occurring would require an independent measurement of ϵ . Unfortunately, however, our current experimental method does not allow us to make such a measurement. We must therefore leave this point as an open question.

It may also be seen in figure 5(a) that r_p increases with ϕ , which suggests that the dissipation-range statistics of the flow may also vary with ϕ . We therefore show in figure 5(b) D_{NN}/r^2 , where we now expect to see a plateau in the small-scale dissipation range. Due to the finite spatial resolution of our measurement system, these very small scales are only partially resolved. This effect is more pronounced for the pure water case, since the small scales appear to increase in the presence of the polymer. Even so, for the water case, we find that the values of ϵ measured from the dissipation-range scaling and the inertial-range scaling of $D_{NN}(r)$ agree to

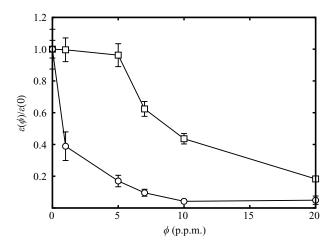


FIGURE 6. Concentration effects on the value of ϵ extracted from the inertial-range scaling of $D_{NN}(r)$ (\Box) and the dissipation-range scaling of $D_{NN}(r)$ (\bigcirc) at fixed Reynolds number ($R_{\lambda}^{w}=350$; Wi=6.0). While the value of ϵ extracted from the dissipation range decreases smoothly as ϕ increases, the value extracted from the inertial range changes only when $\phi>5$ p.p.m.

within our experimental accuracy, suggesting that our dissipation-range statistics are resolved well enough to draw conclusions from. We note that in figure 5(b), the slope of the curves in the range $10 < r/\eta_w < 100$ varies with ϕ . We interpret this result to be due to smooth interpolation of the data between the small-scale r^2 scaling and the inertial-range $r^{2/3}$ scaling over intervals that grow with ϕ .

The polymer effect we see in the dissipation range is qualitatively different from our results in the inertial range. While we found evidence of a critical concentration in the inertial range, figure 5(b) shows no such sharp change with ϕ . Instead, the damping of the small scales by the polymers appears to increase smoothly with increasing ϕ . We note that, just as for the inertial range, the structure functions in the dissipation range appear to show the same scaling (i.e. $D_{NN}(r) \sim r^2$) in both the water and polymer cases; only the scaling constant $(2\epsilon)/(15\nu)$ differs.

To determine if a simple renormalization of ϵ while holding C_2 and ν constant could consistently capture our results, we extracted a value of $\epsilon(\phi)$ from both the dissipation-range and inertial-range scaling ranges of $D_{NN}(r)$, as shown in figure 6. The two curves are very different, suggesting that the polymer effects are more complex than a simple damping of ϵ as the concentration is increased. Our observed critical concentration in the inertial range is also indicative of more complex physics.

The physical basis for the observed transition at $\phi \approx 5$ p.p.m. remains unclear. One possibility is that it may result from polymer–polymer interactions that occur only above the so-called overlap concentration. For the polymer used in our experiments, the overlap concentration based on the maximum extension length is $\sim 10^{-4}$ p.p.m., while it is approximately 200 p.p.m. if based on the radius of gyration. Neither of these estimates coincides with the critical concentration observed in our experiments. The critical concentration proposed by Balkovsky, Fouxon & Lebedev (2001) also does not match our results. We note that $\phi \approx 7$ p.p.m. was found to be the threshold for the onset of efficient mixing in elastic turbulence (Groisman & Steinberg 2001), using the same polymer as in our work.

We note that Tabor & de Gennes (1986) suggested a qualitative framework for polymer dynamics in turbulence that could account for a critical concentration. In their model, they defined a length scale r^{**} based on balancing the turbulent kinetic energy and the elastic energy stored by the polymers as a function of scale. Since each polymer molecule can store some elastic energy, increasing the number of polymers increases the total energy in the polymer phase, so that r^{**} is a function of ϕ . They then defined a critical concentration ϕ^* , determined by $r^{**}(\phi^*) = \eta_w$, for the onset of polymer effects. There are, however, both undetermined parameters in their qualitative theory and questions about the validity of their assumptions (Ryskin 1987). Further development is therefore needed to make a quantitative experimental test.

5. Summary

We have investigated the effect of very small concentrations of long-chain polymers on the dynamics of turbulence in the bulk of the flow. Our main findings are (a) that the length scale r_p at which the structure functions in the polymer solutions begin to scale as $r^{2/3}$ decreases with R_{λ}^w , and $r_p \sim (\nu \tau_p)^{1/2} W i^n$ with $n = -0.58 \pm 0.09$; (b) that there is a critical concentration ϕ_c below which the structure functions reach the same inertial-range values as in Newtonian turbulence, though at a larger length scale that increases with ϕ ; and (c) that the dissipation-range scales are depressed smoothly as a function of ϕ . These results cannot be explained by current theory.

A potential interpretation of these results is that the introduction of polymers into the flow modifies the energy cascade so that the rates of energy injection, transfer and dissipation *for the turbulence* are no longer all equal, as they must be in Newtonian turbulence where viscosity provides the only mechanism of energy dissipation. In order to test this hypothesis, new experimental techniques that can accurately measure these three energy rates independently are required.

Our results suggest several challenges for future research. The qualitative change in the polymer effect above the critical concentration must be explained, and the exact ways in which the polymers affect the energy cascade must be clarified. And finally, if Lumley's (1973) time criterion does not hold in the bulk, a physical mechanism by which the polymers can affect scales much larger than their size must be identified.

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