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The structure of turbulence in a dilute polymer solution

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Abstract

The structure of turbulence is studied and quantified in terms of the higher order moments of the distribution of the velocity differences in the system. These moments are experimentally accessible with the technique of photon correlation spectroscopy. We have developed a new analysis scheme where the distribution of the velocity differences is calculated directly from the higher order spectral moments of the measured correlation functions.

We studied, with this method, the effect of a drag reducing polymer on the structure of a turbulent flow. We observe that the polymer causes a large suppression of small velocity fluctuations accompanied by a large reduction of the turbulent volume fraction. The stretching of the linear, flexible polymer seems to cause a stabilization of the highly fluctuating turbulent core.

Keywords: Drag reducing solutions; Photon correlation spectroscopy; Turbulent structure

1. Introduction

The subject of this paper is a class of apparently Newtonian fluids which exhibit a very remarkable flow anomaly. When turbulence is generated in these solutions, the dissipation of turbulent kinetic energy is enormously reduced relative to the energy dissipation in the pure solvent [1,2]. A maximal reduction can be obtained of about 80%. In a turbulent pipe-flow, this is equivalent to an 80% reduction of the
pressure drop along the pipe. The solutions which exhibit this so-called drag reduction are solutions of linear and flexible polymers. These polymers are very large (a few million a.m.u.) and they are effective at minute concentrations (a few weight ppm). The simple shear viscosity of these solutions is typically only a few percent larger than the viscosity of the solvent alone, which is the reason why a laminar flow remains practically unaffected upon addition of a drag reducing polymer. The extensional viscosity of these solutions however is a few orders of magnitude larger than the solvent viscosity. Due to the elongational motion of the fluid, the polymer molecules tend to stretch and align along the streamlines of the flow [3], thereby inducing an extra tension. Comparing the magnitudes of both effects on the viscosities and taking into account the fact that turbulence is a rapidly varying system of stretching and shearing motions, it is probable that stretching of flexible polymers is responsible for the phenomenon of drag reduction.

As yet, the mechanism by which the reduction of turbulent drag occurs has not been firmly established. Lumley [4] and Landahl [5] consider drag reduction to be an effect related to the turbulent boundary layer. According to Lumley, the reduction of turbulent drag is the result of the enhancement of the viscosity of the turbulent core while the viscosity of the boundary layer remains practically unchanged. Landahl considers the large extensional viscosity of the polymer solutions to stabilize small scale secondary motions in the boundary layer, thereby inhibiting the production of turbulent bursts. However, the experiments of McComb and Rabie [6] and Bewersdorf [7] indicated that, after injecting the polymer in the core of the turbulent flow, reduction of turbulent drag occurred before the polymer had reached any boundary. This led Tabor and de Gennes [8] to try a completely new approach, based on a viscoelastic polymer eddy interaction in the core of the turbulent flow, far from any boundary. In the turbulent cascade, the kinetic energy is subtracted from the mean flow by the large eddies and is transferred successively to smaller scales, until the dissipation scale is reached. At this scale, the energy is dissipated by viscosity. At large scales the hydrodynamic stresses are much larger than the internal stresses of the polymer and the polymer is only advected passively. At smaller scales, the Reynolds stress becomes of the same order of magnitude as the elastic stress in the polymer. At this point, a truncation of the turbulent cascade is expected, which should lead to the reduction in the amount of kinetic energy dissipated in the system.

With the usual experimental technique of laser Doppler velocimetry, which measures the flow velocity $v(t)$, it is quite difficult to obtain quantitative spatial information on the small scales of turbulence. We therefore use the technique of PCS (photon correlation spectroscopy), which measures the instantaneous Lagrangian velocity difference $\vec{V}_R(t)$ over a distance $R$:

$$\vec{V}_R(t) = \vec{v}[t] - \vec{v}[t + R].$$

This dynamic light scattering technique is very suitable for measurements on turbulence, since one obtains spatial information on the distribution of the small velocity differences, relative to the mean flow.
2. Method

With the technique of PCS [9,10], one measures the time correlation function of the scattered light intensity \( I(t) \):

\[
g(t) = \frac{\langle I(t' + t)I(t') \rangle}{\langle I(t') \rangle^2}.
\]

(2)

The light is scattered by a large number of small scattering particles which are suspended in the fluid and follow the local flow field affinely. The scattered beam is the beating of the Doppler shifted light scattered by pairs of particles present in the scattering volume. Each particle pair contributes a phase factor \( \cos(q t \mathbf{V}_R) \) to the correlation function.

\[
g(t) = 1 + \int_0^L h(R) \langle \cos[\mathbf{q} \cdot \mathbf{V}_R(t)t] \rangle.
\]

(3)

Here, \( h(R) \) is the distribution of particle pairs separated by a distance \( R \) which are present in the scattering volume, and is well described by

\[
h(R) = \frac{2(1 - R/L)}{L},
\]

(4)

where \( L \) is the length of the (one dimensional) scattering volume and \( \mathbf{q} \) the scattering vector whose amplitude is given by

\[
q = \frac{4\pi n}{\lambda} \sin(\theta/2).
\]

(5)

Here, \( n \) is the refractive index of the fluid, \( \lambda \) the wavelength of the incident light and \( \theta \) is the angle under which the scattered light is collected.

Since the velocity differences can arise from the Brownian motion as well as the advective motion of the scattering particles, \( g(t) \) can be written as

\[
g(t) = 1 + A(t)G(qt, L),
\]

(6)

where \( A(t) \) is the factor representing the Brownian motion: \( A(t) \approx \exp(-q^2tD_T) \) (with \( D_T \) the diffusion coefficient). The function \( G(qt, L) \) is due to the advective motion of the scattering particles and has the form:

\[
G(qt, L) = \int_0^L h(R) \int_{-\infty}^\infty dV_R P(V_R) \cos(q t V_R).
\]

(7)

Here, \( V_R \) is the component of \( \mathbf{V}_R(t) \) in the direction of \( \mathbf{q} \) and \( P(V_R) \) is the probability distribution of \( V_R \).

A useful general expression for \( P(V_R) \) is [11]

\[
P(V_R) = \frac{\exp[-u_L^2(R)/2u_G^2(R)]}{\pi \operatorname{erfc}[u_L(R)/\sqrt{2u_G(R)}]} \times \frac{u_L(R) \exp[-V_R^2/2u_G^2(R)]}{[V_R^2 + u_L^2(R)]},
\]

(8)

where \( \operatorname{erfc} \) is the complementary error function. The first term on the right hand side represents the normalization of \( P(V_R) \), while the second term consists of a
Gaussian multiplied with a Lorentzian. The Gaussian represents the distribution of the turbulent velocity fluctuations which take part in the turbulent cascade. These are the regions where the vorticity is localized. They are called the active regions of the flow. The Lorentzian distribution describes the small velocity fluctuations which do not participate in the turbulent cascade and which are called inactive. Consequently, \( u_G(R) \) is the average of the turbulent velocity differences and \( u_L(R) \) is the average of the ‘quiescent’, inactive velocity differences.

These so-called scaling velocities determine the higher order moments of \( P(V_R) \) as [12]

\[
\langle |V_R|^n \rangle \approx u_L(R)u_G(R)^{n-1}.
\]  

(9)

Returning to the experimentally accessible quantity, the correlation function \( G(t) \), the implication of Eq. (9) for the spectrum \( G(\omega) \), which is the real Fourier transform of \( G(t) \), is that the higher order spectral moments \( \mu_n \) should scale like

\[
\mu_n = \langle |\omega|^n \rangle \approx u_L(L)u_G(L)^{n-1}.
\]  

(10)

We use this relation to derive the following equations, where the coefficients are calculated numerically:

\[
\mu_2 = 0.058u_L(L)u_G(L)q^2,
\]  

(11)

\[
F = \mu_4/(\mu_2)^2 = 3 + 1.6 \frac{u_G(L)}{u_L(L)}.
\]  

(12)

Here, \( \mu_2 \) is the second order spectral moment and \( F \) the normalized fourth order spectral moment, often referred to as the kurtosis or flatness of the spectrum \( G(\omega) \).

Due to the self-similarity of velocity fluctuations in turbulence [13], the velocities at scale \( R \) are related to the velocities at the experimental scale \( L \) (the length of the scattering volume) as

\[
u(R) = u(L)\left(\frac{R}{L}\right)^{\alpha}.
\]  

(13)

Following Kolmogorov, \( \alpha = 1/3 \) [14].

Under experimental conditions, turbulence is often not fully developed and not completely space-filling. The volume fraction filled with turbulent fluctuations can be denoted as \( \beta \) [15]. This quantity is inversely proportional to the ratio of the Lorentzian scaling velocity and the Gaussian scaling velocity [12]:

\[
1/\beta \approx u_G(L)/u_L(L).
\]  

(14)

Thus \( F \) can be written as

\[
F = 3 + 1.6\beta^{-1}.
\]  

(15)

Before turning to the experiments, we focus on the physical implication of the above relations. While the flatness has its statistical meaning for the distributions of velocity differences, the cumulant expression for \( F \) describes its physical meaning. In
terms of higher order correlation, \( F \) can be written as

\[
F = \frac{\mu_4}{(\mu_2)^2} = \frac{3(\kappa_2)^2 + \kappa_4}{(\kappa_2)^2} = 3 + \left( \frac{\langle V_{R_1} V_{R_2} V_{R_3} V_{R_4} \rangle_c}{\langle V_{R_1} V_{R_2} \rangle^2} \right),
\]

where \( \kappa_n \) is the \( n \)th order cumulant. The last term on the right hand side of Eq. (16) is the normalized 4-points velocity correlation. This equation implies that when the velocity fluctuations are essentially random, there are, upon the Gaussian correlations, no higher order correlations present in the system. The flatness then has the value of the flatness of a Gaussian. When the velocity fluctuations are not completely chaotic, there are higher order correlations present in the system and the flatness is consequently larger than 3. A high flatness thus implies that the system of velocity fluctuations is structured. When we recall from this point of view that turbulence is essentially random (in our terms unstructured), Eq. (15), where the flatness is expressed in the turbulent volume fraction \( \beta \), becomes evident.

Summarizing, in this new and direct method of analysis, the relations (11) and (12) are used to characterize the measured correlation functions in terms of two scaling velocities and to calculate directly the probability distribution \( P(V_R) \) (with Eq. (8)).

3. Experimental set-up

The experimental set-up is depicted in Fig. 1. The flow is achieved by a large container with water located at about 2 m above the measuring cell. The water-level in the container is kept constant by a variable speed peristaltic pump which pumps the fluid flowing out of the cell back to the height of 2 m. A PVC tube with a diameter of 14 mm leads the flow to the measuring part of the set-up. The measurement cell is made of glass and contains parts of optical quartz where the laser beam comes in and where the scattered beam is detected. The horizontal tube has a length of 10 cm and a diameter of 5 mm. As the length of the tube is short compared to the length of the supply tube, it is probable that the turbulence in the horizontal part is governed by the turbulence in the feeding flow. Another feature of this particular geometry is that the turbulence is probably far from being fully developed. On the other hand, since the effect of drag reducing polymers on turbulence is not restricted to a homogeneous and isotropic case, it is not an essential requirement for the turbulence in our experiments to be fully developed.

The Reynolds numbers \( Re \) which can be achieved in the horizontal tube range from 0 to 25 000. At the highest \( Re \), the Kolmogorov dissipation scale \( l_\eta \) is about 25 \( \mu \text{m} \). The drag reducing polymer we use is polyethylene oxide with an average molecular weight of \( 3.8 \times 10^6 \) a.m.u. (polyox WSR 301, Union Carbide). The polymers are dissolved in water at a concentration of 5 weight ppm. This solution gives, at the highest \( Re \) in the horizontal measuring tube, a reduction of the turbulent drag of about 15%. Simultaneously, the amount of drag reduction in the supply tube is found to be negligible, which indicates that the activity of the polymers predominantly takes place in the horizontal glass tube. At the highest
Reynolds number, a reduction of the polymer's activity due to mechanical degradation is observable after at least four complete recirculations of the fluid, while our light scattering experiments take maximally two recirculations.

In Fig. 1, the incident laser beam is sketched together with the scattered beam. We use an argon laser operating at a wavelength of 514 nm. The scattering is collected by a photomultiplier under very small angles $\theta$ ($0 < \theta < 3^\circ$). In this way, it is possible to measure the almost pure transverse velocity fluctuations (in the configuration as depicted in Fig. 1) and the almost pure longitudinal fluctuations (the measuring cell is then rotated 90° with respect to the incoming laser beam). The scattering volume is located about 8 cm downstream and has a length of about 5 mm. The location of the scattering volume ensures a flat velocity profile without the problem of secondary flows [16]. Since the flush time of the polymer solution is at least one order of magnitude larger than the molecular relaxation time [14], we may assume any entrance effects on the polymers to have died away at the point of measurement.

We will focus in this paper on the longitudinal velocity fluctuations, because both the transverse fluctuations and the stretching of the polymers are too much influenced by the vicinity of the pipe walls. The experiments are performed at a
The scattering collected by the photomultiplier is fed into a 1024-channel correlator. As scattering particles, we use polystyrene latex spheres with a diameter of 0.233 μm and a volume fraction of ±5 x 10⁻².

4. Results

The procedure is as follows: the correlation functions are measured in the flow of pure water at various Reynolds numbers. Knowing the diffusion coefficient from measurements under zero-flow conditions, we eliminate the contribution of the Brownian motion (Eq. (6)) and obtain the correlation function G(t) (for a typical correlation function, glance ahead to Fig. 6). By Fourier transforming G(t) into G(ω), we can calculate the (experimental) higher order spectral moments. These are used to quantify the turbulent structure by calculating the probability distribution of the velocity differences (Eqs. (11) and (12)) and to characterize the structure in terms of the turbulent volume fraction (Eq. (15)). Then, the same scheme is performed for the flow of the drag reducing polyox solution.

In Fig. 2, the second order moments μ₂ measured in water and in the polyox solution are plotted as a function of Re. Polyox seems to cause an enhancement in the amount of kinetic energy in the system. In Fig. 3, the flatness F (see Eq. (12)) measured in water is plotted against Re. The flatness increases until Re ≈ 9000, which is exactly the point where the flow in the feeding pipe becomes turbulent too. At this point, the velocity fluctuations in the measuring part of the flow start to behave in a self-similar fashion, which can be concluded from the fact that the measured correlation functions can be superimposed at Re larger than 9000. In this self-similar range, the flatness decreases gradually with increasing Reynolds number. When we now add polyox, the effect on the measured flatness is very dramatic.

![Fig. 2. The square root of the measured second order spectral moments in water (■) and in the polyox solution (○) as a function of Re. The lines are least square fits to the data.](image-url)
Fig. 3. The flatness $F$ of the spectra $G(\omega)$ in water is plotted at different $Re$. After the transition to self-similar behaviour at $Re \approx 9000$, a decreasing trend in $F$ seems to be present (dotted curve). Some error bars are left out for the sake of clarity.

In this range (Fig. 4). The flatness in the polyox solution increases by about a factor four with respect to the case of pure water.

In order to quantify the above observations, we calculate at a Reynolds number of about 24 000 the probability distributions of the velocity fluctuations in water and polyox (Fig. 5(a)). The values of $\mu_2$ and $F$ needed to calculate the Gaussian and the Lorentzian scaling velocity with Eqs. (11) and (12) are extracted from Figs. 2 and 3. We observe that adding polyox leads to a suppression of the small velocity fluctuations, while there seems to be an increase towards larger velocity fluctuations. Considering that the Reynolds number characterizes the behaviour of the turbulent flow as a whole, we can eliminate any effect of polymers on the boundary layer by plotting both distributions normalized on the same turbulent intensity (see

Fig. 4. The flatness of the spectra $G(\omega)$ in the polyox solution (◊) and in water (■) as a function of $Re$ in the self-similar range. At $Re < 9000$, there is no difference in the flatness of a polyox solution and water. The lines are drawn to guide the eye.
Fig. 5. (a) The probability distribution $P(V_R)$ measured in pure water (solid line) and in the polyox solution (dotted line) is plotted as a function of the velocity differences $V_R$. The $P(V_R)$ are calculated from the correlation functions measured at $Re \approx 24\,000$. By adding polyox, a whole range of small velocity differences is suppressed. (b) In this figure, the distributions are normalized on their standard deviations $\sigma = \langle |V_R|^2 \rangle^{1/2}$. $P(\gamma)$, with $\gamma = V_R/\sigma$, is calculated in water (solid line) and in the polyox solution (dotted line). Relative to the same turbulent intensity, the suppression of the small velocity differences is even more pronounced.

Fig. 5(b)). At identical turbulent bulk conditions, the suppression of the small velocity fluctuations by polyox is even more pronounced, which indicates that the action of the polymers essentially occurs in the core of the turbulent flow.

In order to check the above analysis, we calculated a correlation function with the values of the scaling velocities in the case of pure water at $Re = 24\,000$ (Fig. 6). We observe that this numerical correlation function fits well with the experimental one at the corresponding Reynolds number.

Fig. 6. $G(t)$ measured in the polyox solution ($\diamond$) at $Re = 23\,800$ and $G(t)$ calculated from Fig. 5 (solid line).

Fig. 7. The turbulent volume fraction in water ($\#$) and in the polyox solution ($\diamond$) as a function of $Re$. In water, a trend towards more developed turbulence is observed. The addition of polyox drastically reduces the turbulent volume fraction. The lines are drawn to guide the eye.
Finally, we calculate with eqn. (15) the turbulent volume fraction in the case of pure water and in the case of polyox with the measured flatnesses of Fig. 7. In pure water, the turbulent volume fraction increases with increasing \( Re \). This is the natural development of a turbulent flow towards a more developed, homogeneous state. By adding polyox, the turbulent volume fraction drastically reduces by about one order of magnitude. The increasing trend with increasing \( Re \) which is present in pure water, seems to be absent in the polymer solution. This means probably that, in this range, the effect of drag reduction has not reached its point of saturation, or, in other words, the amount of drag reduction still increases with increasing \( Re \).

In order to verify that all of the above observed changes are only due to the addition of a drag reducing polymer, we added a non-drag reducing polymer; a semi-rigid polymer xanthan. Measurements on a 5 weight ppm solution of xanthan indicated that, after a correction on \( Re \) due to the slightly enhanced viscosity of the xanthan solution, the results are identical to those in pure water.

5. Discussion

Considering the above observations, the effect of a drag reducing polymer on a turbulent flow seems to be described best in terms of the Tabor–de Gennes scenario [8]. According to them, a viscoelastic polymer eddy interaction in the core of the turbulent flow should truncate the energy cascade at small scales, thereby deeply modifying the distribution of turbulent velocities. We indeed observe that the addition of a drag reducing polymer causes a reduction of the amount of small velocity fluctuations (Fig. 5). When we compare the distributions at the same turbulent intensity, i.e. under identical turbulent bulk conditions, we observe that the polymer drastically modifies the form of the distributions. This effect is accompanied by a large reduction of the turbulent volume fraction. It is clear that we cannot rule out any effect of the polymers on the turbulent boundary layer, but the experiments described here suggest the action of the polymer to be predominantly a bulk effect.

The criterion which has yet to be satisfied is the time criterion. For the polymer used in this experiment, a coiled–stretch transition is expected to take place at a characteristic Zimm relaxation frequency \( \Gamma_z \) of about \( 10^3 \text{ s}^{-1} \). The turbulent shear rates in the horizontal measuring section of our system roughly vary between \( 10^3 \text{ s}^{-1} \) at the largest length scales and \( 10^6 \text{ s}^{-1} \) at the smallest (dissipative) scales. The shear rates are consequently large enough to stretch the polymers. The turbulent shear rates are not large enough to interact with the rigid polymer molecules of xanthan. Therefore, addition of xanthan does not influence the structure of the turbulent flow. This was indeed observed experimentally.

We observe that adding polyox to our turbulent system causes a suppression of the small velocity differences in the bulk of the turbulent system and thereby a huge reduction of the fraction of the volume occupied by the turbulent cascade. The polymer seems to act as a kind of bufferstop so that turbulent kinetic energy cannot be transferred to smaller scales where it should be dissipated by viscosity. The fact
that this suppression decreases the turbulent volume fraction by about a factor ten and suggests that the reaction of the flow on the polymer is a redistribution of the velocity fluctuations both in space and time. This implies a dynamical feedback from small to large scales. The effect of drag reducing polymers on the origin of this feedback has recently been demonstrated experimentally by means of a vorticity-visualization technique [18,19].

When we recall Eq. (16), the high flatness measured in the flow of the polymeric liquid is equivalent to the presence of higher order correlations in the system. By suppressing the small scales, the polymer stabilizes the turbulent environment causing a more structured medium where large scale convective motions are able to survive for a longer time.

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References

[14] Corrections which have to be made for possible intermittency in turbulence concern the exact value of $\alpha$. Earlier experiments have indicated that the deviation from the value of $\frac{1}{3}$ under drag reducing conditions is negligible, see, e.g. P. Tong et al., Phys. Rev. A, 45 (1992) 7231.
[17] The Zimm relaxation rate $\Gamma_s = k_B T/\eta_s R_g^2$, where $\eta_s$ is the solvent viscosity, $R_g$ the radius of gyration of polymer coil at rest, $k_B$ the Boltzman constant and $T$ the temperature. The $R_g$ of our polymer is about 250 nm, resulting in a Zimm relaxation rate of about $10^9$ s$^{-1}$.