Complex dynamics of knotted filaments in shear flow

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Abstract – Coarse-grained simulations are used to demonstrate that knotted filaments in shear flow at zero Reynolds number exhibit remarkably rich dynamic behaviour. For stiff filaments that are weakly deformed by the shear forces, the knotted filaments rotate like rigid objects in the flow. But away from this regime the interplay between shear forces and the flexibility of the filament leads to intricate regular and chaotic modes of motion that can be divided into distinct families. The set of accessible mode families depends to first order on a dimensionless number that relates the filament length, the elastic modulus, the friction per unit length and the shear rate.

The interaction between a shear gradient and suspended objects can generate fascinating dynamical behaviour. G. Jeffrey [1] showed in 1922 that rigid bodies trace out complex periodic orbits that depend in detail on their shapes. For deformable objects [2] even richer behaviour is possible. Red blood cells, for example, change shape with increasing shear rate [3]. Furthermore, the coupling to shear can lead to either unstable tumbling or to a steady state mode where the cells remain at a fixed angle to the flow while their outer membrane rotates like the treading of a tank [4]. A similar crossover to tank treading is predicted for star polymers [5]. Experimental advances in single molecule techniques have made it possible to directly observe the stretching and tumbling behaviour of individual DNA molecules [6]. This work inspired a great deal of theoretical research on the way that the polymer flexibility, shear and Brownian noise interact [7]. Several decades earlier it had been shown that filaments in the non-Brownian regime exhibit at least five distinguishable regimes of motion as stiffness, length, and shear rate are varied [8]. These results are still the subject of active investigation by theorists [9]. The crossover between the Brownian and non-Brownian regimes has also been recently considered [10].

In this paper we use coarse-grained computer simulations to study the behaviour of knotted non-Brownian filaments in shear flow. Knots are a generic possibility for any long elastic objects and occur naturally in biologically active DNA [11,12]. In the limit of strong bending modulus A or weak shear rate ˙γ the knotted filament will take on its equilibrium shape [13], and rotate in a manner similar to that first predicted by Jeffrey [1]. Chiral knots should also migrate in the vorticity direction [14,15], a hydrodynamic effect that has recently been observed for other objects including helical bacteria [16].

The focus of this paper, however, is what happens for stronger shear forces and/or for more flexible filaments, i.e. the regime where the knots can be tightened by the flow. To estimate where this crossover occurs we consider the following argument: in the limit of small local bond deformations, the bending energy of a knot of length L scales as ∼A/L so there is a force opposing knot tightening ∼A/L. To tighten the knot, strands which are close to each other must be moved in opposite directions. The typical velocity difference will be ∼σ ˙γ, where σ is the filament width.

Combining this with the drag and comparing to the bending force results in a dimensionless knot deformation number, α = A/ησ ˙γL^3. (At the crossover, the knot length ≈ total filament length, so in calculating α we take L to be the total filament length.) This resembles the sperm number used with microscopic swimmers [18] but instead of determining when the filament as a whole may be deformed, it indicates when the knot will be tightened. For larger α we expect the stiff knot regime, but for lower α the shear should cause significant deformation.

Indeed, as illustrated in fig. 1 for the case of the simple trefoil knot, lowering α leads to a crossover from knots that remain close to their equilibrium shape, to a

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regime of surprisingly rich dynamical behaviour where the whole filament exhibits intricate shape oscillations in time. These orbits may be grouped into a few distinct families comprising very similar types of motion (modes). Some modes show regular, and others chaotic, motion. Different modes show distinct rates and directions of drift along the vorticity axis. A few families are accessible at each mode: they show distinct rates and directions of drift along the vorticity axis. The series spans approximately one period, the time between successive configurations is 1028.7τ0 and migration is in the −z-direction. (b) Example configurations for different mode families over a range of α. Note that the orientation is different for the VII mode. Full animations of the modes are available online (see table 1 in the appendix).

Fig. 1: Predicted modes of behaviour as a function of the knot deformation number α. x is the flow direction, y is the direction of the shear gradient, and z is the vorticity direction. All configurations are for filaments with a $3_1(-)$ trefoil knot. (a) Time series for filament in a II mode at $\alpha = 0$. Time increases from left to right. The knot rotates clockwise around the z-axis. The series spans approximately one period, the time between successive configurations is 1028.7τ0 and migration is in the −z-direction. (b) Example configurations for different mode families over a range of α. Note that the orientation is different for the VII mode. Full animations of the modes are available online (see table 1 in the appendix).

We apply a coarse-grained bead-spring model [19]. The natural input units for the simulation are $\sigma$, $\eta$, and $k$, from which a natural time unit of $\tau_0 = \sigma \tau / k$ follows. We used $\gamma = (150\tau_0)^{-1}$, and verified that the stretching of individual bonds relaxes much more quickly than the characteristic time for shear induced motion and so should not influence the dynamics [22].
although for some parameter choices, for example high $\kappa$, it was necessary to reduce this for numerical stability. We checked that using $\Delta t = 10^{-3}$ gave equivalent results.

Mode families were identified by visual inspection of their motion (see, e.g., the supplementary videos in table 1 in the appendix) and by measuring their drift velocity in the vorticity direction, which distinguished well between different modes. The identity was confirmed by two additional order parameters which measure the direction of maximum extension and the asymmetry under a $\pi$ rotation about $z$. Detailed definitions, as well as plots of average values for individual runs, are given in the appendix.

The elastic modulus of our bead-spring model $A = \kappa \sigma$ [18], so that the knot deformation number defined earlier takes the form $\alpha = \kappa / \eta^2 L^3$. In fig. 1 we consider 10 different values of $\kappa$ corresponding to $\alpha = 0 - 2.56 \times 10^{-3}$. For the largest $\alpha$, the filament remains in braid-like configurations [13] that characterise family VII in fig. 1(b). The motion is composed partly of rotation of the configuration and partly of tank-treading—a particular point moves around the contour. As flexibility is increased ($\alpha$ is lowered) there is a change from modes which rotate in the $x-y$ plane to modes which have relatively large extensions in the $z$-direction. Interestingly, a similar shift was seen in experiment with linear filaments [8]. The first family in which the knot is significantly tightened is $V$.

Some mode families show both regular and chaotic modes, sometimes at the same $\alpha$, for example family II. Others showed only regular (VI), or only chaotic (IV), motion. To distinguish regular and chaotic modes, we calculated the largest Lyapunov exponent, $\sigma_1$ [25]: a second system was created with the bead positions $\vec{r}_i$ randomly displaced to $\vec{s}_i$ and constrained so that $d^2 = \sum_i |\vec{r}_i - \vec{s}_i|^2 / \sigma^2 = 1$. Both systems were integrated forward in time. After each $1.5t_0$, $\vec{r}_i - \vec{s}_i$ were rescaled by changing $\vec{s}_i$ so as to make $d = 1$. The Lyapunov exponent is then given by [25]:

$$\sigma_1 \equiv \lim_{n \to \infty} \sigma_n = \lim_{n \to \infty} (2/3nt_0) \sum_{j=1}^{n} \ln(d_j),$$

where $d_j$ is the distance after the $j$-th evolution. If the measured $\sigma_1$ tended to zero or a positive constant as a function of $n$ the motion was identified as regular or chaotic, respectively. This behaviour is illustrated in the inset of fig. 2.

Fig. 2: Comparing regular and chaotic runs with modes in family II at $\alpha = 0$. The average $z$-position of the filament beads with the average drift subtracted is plotted as a function of time. The inset shows the estimate of the largest Lyapunov exponent as a function of time for the two runs, plotted on a log-log scale.

Fig. 3: Migration velocity in the $z$-direction, averaged over all runs belonging to each family. The labels indicate families and are subdivided into regular and chaotic modes, denoted by the subscripts $r$ and $c$, respectively. Error bars show the standard deviations. The figures in brackets indicate the percentage of runs at each $\alpha$ that were observed to fall into each group.

Data points. Figure 3 also shows that at some $\alpha$ there exist modes that migrate in opposite directions. The period of rotation of regular modes varies with $\alpha$, approximately in the range $2 - 6 \times 10^4 t_0$. The shortest periods were seen for the highest $\alpha$. For example at $\alpha = 0$ the average period of modes in the II family was $6220 \pm 400 t_0$ and that of family VII at $\alpha = 2.56 \times 10^{-3}$ was $2040 \pm 90 t_0$. Figure 3 also shows the percentage chance that a run with random initial conditions ends up in the particular mode family.

We also considered simulations for other knot types at $\alpha = 0$. For $3_1$ the same modes are seen but the migration, and the orientation, is as expected, in the opposite $z$-direction. We see a similar behaviour — regular and chaotic modes with migration — for more complex knots such as $4_1$ and $5_1$. For achiral $4_1$ the distribution of migration velocities is symmetric about zero: all migrating modes have
a partner with opposite migration direction. Depending on initial conditions, the $4_1$ knot may thus migrate in the $\pm z$-direction, but that the average migration velocity over many runs would be zero.

Finally, we consider the sensitivity of our results to changes in parameters and changes in simulation details. It should be kept in mind that for these dynamical systems with behaviour that may depend sensitively on initial conditions, one would expect quantitative changes when simulation details are changed. The main thrust of our paper is qualitative, and so the most important tests will be whether the overall behaviour, i.e., the mode families, are robust to these changes.

Firstly, we consider the effect of hydrodynamic interactions by setting $H_{ij} = 0$ for $i \neq j$ in eq. (2). We find, as expected, that the motion in the vorticity direction is a consequence of off-diagonal hydrodynamic interactions. Modes that resemble those of families $V$ and $VII$ were seen but none with large $z$-extensions.

Secondly, to force hydrodynamics simply align in the $x$-$z$ plane without access to different shear velocities. Simulations must therefore explicitly take the finite thickness into account by considering the torque on individual beads [9]. We tested this sensitivity by using algorithms that include the torque, and find that, in contrast to unknotted filaments, similar mode families are observed. The knot forces the system out of the plane so that it always accesses different shear velocities, and this dominates.

Thirdly, we checked how the behaviour is affected by changing $\kappa$, $\eta$ and $\dot{\gamma}$ in such a way as to keep $\alpha$ fixed: in the absence discretisation effects, such changes of parameters should lead to descriptions of the same physical system and so the same behaviour is expected. We ran two sets of simulations where $\dot{\gamma}$ was reduced by a factor of 10 and either $\kappa$ was decreased or $\eta$ increased to compensate. We obtained very similar results.

Finally, most of our results are for a fixed length filament with $N = 50$ beads. It is interesting to investigate how sensitive our results are to the length $L = N\sigma$. For example, for $N = 50$ significant tightening first occurs in family $V$ at $\alpha = 0.64 \times 10^{-3}$. We ran additional simulations with $N = 40, 70$ and 100 at $\alpha = 0.64 \times 10^{-3}$. In each case the majority of runs show a mode very similar to those in family $V$, see fig. 4. The migration velocities are similar but decrease with $L$. The measured velocities are $1.60, 1.51, 1.35$ and $1.27 \times 10^{-4}\sigma/t_0$, respectively (in each case all runs in the mode had exactly the same velocity to the accuracy given). At other $\alpha$'s we checked the results at different $N$ were also qualitatively similar, although the agreement worsens at lower $\alpha$. For example at $\alpha = 1.28 \times 10^{-3}$ modes like families VI and VII were seen for all $N$ but for $\alpha < 0.16 \times 10^{-3}$ we observed families for $N = 100$ that were qualitatively different to any seen for other $N$. In fact, we expect substantial differences at small $\alpha$ because the shape of the tighter knot is then fixed by the excluded volume of the chain and not just by the physics that enters into the derivation of $\alpha$. In that regime, for fixed filament thickness, we expect the influence of the knot to become progressively smaller as $L/\sigma \to \infty$. While it would be interesting to explore these effects further, at fixed $\dot{\gamma}$, changing $N$ while fixing $\alpha$ means that $\kappa$ must be increased as $N^3$ and the integration timestep correspondingly decreased for stability. Combining this with $N^3$ time for calculation of $H_{ij}$ gives a prohibitive $\sim N^9$ scaling of simulation time.

To summarise, we have demonstrated that knotted filaments in shear exhibit a rich dynamical behaviour with modes which can be divided into families. Some families have both regular and chaotic modes. Mode families migrate in different directions along the vorticity axis. The crossover from a stiff knot to the regime where multiple modes are possible can be described by a dimensionless number. In future work it may be interesting to consider more sophisticated treatments of the hydrodynamics that include effects such as lubrication. It may also be interesting to consider the effect of noise: initial simulations suggest that fluctuations may alter the stability of modes leading to a variation or even flipping of migration velocity as function of noise strength.

Experimentally, this behaviour would be most easily observable with macroscopic filaments in highly viscous solutions [8]. However, it may also be visible for DNA. For example, the $P4$ phage genome (common in knotting experiments [12]) is about 77 thermal persistence lengths long. We estimate a crossover ($\alpha = 10^{-3}$) at $\dot{\gamma} \approx 2 \times 10^3 s^{-1}$ in water. The Weissenberg number $\approx 10$ so shear should be reasonably strong compared to thermal effects.

**APPENDIX**

We present a range of animations of modes (see supplementary movies in table 1) from the families described in the main text. The filenames, along with additional information, are listed in table 1. All animations are of duration 9000$t_0$ and of simulations at a shear rate of $\dot{\gamma} = (150t_0)^{-1}$ with a $3_1(-)$ knot. The green sections of the filaments are markers to allow the motion to be more easily followed.

We next briefly discuss the two order parameters that were used to help group runs into mode families. The first, $\phi$, was the angle of the direction of maximum extension to the $z$-axis, allowed to vary between 0 and $\pi/2$. $\phi$
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Table 1: Examples of animations of modes belonging to the various families described in the main text. Animations were created using VMD [26].

| Filename            | Mode family | Regular/Chaotic | N | $\alpha \times 10^3$ |
|---------------------|-------------|-----------------|---|----------------------|
| fam1r.mpg           | I           | r               | 50 | 0                    |
| fam2r.mpg           | II          | r               | 50 | 0                    |
| fam2c.mpg           | II          | c               | 50 | 0                    |
| fam3r.mpg           | III         | r               | 50 | 0.04                 |
| fam3r2.mpg          | III         | r               | 50 | 0.16                 |
| fam4c.mpg           | IV          | c               | 50 | 0.16                 |
| fam5r.mpg           | V           | r               | 50 | 0.64                 |
| fam6c.mpg           | VI          | c               | 50 | 1.28                 |
| fam7r.mpg           | VII         | r               | 50 | 0.64                 |
| fam5N40.mpg         | V           | r               | 40 | 0.64                 |
| fam5N70.mpg         | V           | r               | 70 | 0.64                 |
| fam5N100.mpg        | V           | r               | 100| 0.64                 |

Fig. 5: The values of the order parameters, averaged over single runs for $N=50$ filaments at different values of $\alpha$. (a) The angle of the direction of maximum extension to the $z$-axis, $\phi$. The averages of $\phi$ for each run are plotted for a given $\alpha$ in an arbitrary order. The labels indicate the modes to which the different groups of points correspond, (b) The same as (a) but for $C_2$, an order parameter to detect two-fold symmetry about the z-axis. Lower values indicate more symmetric configurations. It should be emphasised that all the points within two consecutive vertical lines correspond to different runs at the same $\alpha$ — the positions along the $x$-axis within each section are irrelevant.

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