Practical method for aligning charged filaments predicted by "caterpillar" hydrodynamics

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Abstract

We describe a "caterpillar" hydrodynamic model for accurately calculating the hydrodynamic friction force on microscopic slender cylindrical filaments. Using the method we show that, if the filaments are charged, applying a circularly polarized electric field causes them to align along the axis of the field. The field strengths and frequencies required are easily realizable experimentally. We propose that this is a practical method for aligning filaments such as microtubules and functionalized carbon nanotubes. This is an important requirement for many nanotech applications. (PACS: 87.16.Ka,05.45.a, 46.32.+x, 47.15.Gf)

Many slender microscopic filaments display remarkable properties. Notable examples are carbon nanotubes and biological fibers, such as actin and microtubules. In cells, the latter provide both strength and a means of intra cellular transport. They act as "tracks" along which nano-scale motor proteins process, transporting cargo such as vesicles and large proteins. There is considerable scope for applying molecular motors as components in nano-devices. For many nanotech applications manipulating the orientations of these types of filaments is a requirement. That is, imparting directionality to an otherwise disordered system. This is not straightforward. For example, although microtubules possess a dipole moment the static electric field strength required to align them exceeds that at which they simply disintegrate [1]. A greater degree of success has been achieved on surfaces using kinesin and polarity-specific antibodies for immobilization prior to the application of external fields [2, 3]. However, as well as the fact that they are charged, one can also exploit the fact that filaments are normally dispersed in a viscous environment. This too influences their behavior [4, 5, 6, 1]. Notably, if they are subject to a force that generates translational motion (gravity or an electric field, for example) they will tend to orientate in a plane perpendicular to that force.

The origin of this effect is the interplay between hydrodynamic friction and the bending elasticity. That is, the elasto-hydrodynamics of the problem. In short, when one allows for the motion of the solvent caused by the motion of the filament, the friction it experiences is higher towards the ends than in the middle. This causes it to bend slightly. The bending in turn introduces a force anisotropy in the form of a torque leading to rotation and a re-orientation into the perpendicular plane. Unfortunately, within the plane there is no alignment and uniform translational motion will also induce significant inhomogeneity in the system. The question we address here is: is there some other way of exploiting this effect that can actually align filaments to a particular direction? Specifically, we consider a filament in a circularly polarized field. Such a field conveniently avoids net translation, but does the elasto-hydrodynamic aligning effect persist? If so, how does the filament quantitatively respond to such a field? To answer these questions we use computer simulation. Our first

requirement is therefore a numerical model that captures accurately all the relevant effects.

We begin by considering how one constructs a tractable but sufficiently realistic numerical model to solve the problem. First consider a filament of length L discretized into n beads. The distance between neighboring beads is fixed, meaning that the model filament is inextensible. Consequently, the bead separation is given by $b = L/(n-1)$. As the filament deforms elasticity will penalize deviation from the lowest energy conformation. Assuming there is no intrinsic curvature, according to elasticity theory the elastic energy U_e in the continuum limit is

$$U_e = \frac{\alpha}{2} \int_0^L \kappa(s)^2 ds \tag{1}$$

where α is the bending modulus and $\kappa(s)$ the curvature at a point a distance s along the filament. The Hamiltonian of our model system is derived by introducing a bending potential between all sets of three consecutive beads and then regarding the model as a discretization of Eq. (1). The inextensibilty constraint is imposed using the MILC SHAKE algorithm [7]. Filaments of fixed length are effectively infinitely stiff while retaining flexibility. The reason we chose this approach is that the type of filaments we consider here are structured on the nanometer scale and cannot accommodate significant axial extension. This provides a reasonable description of force extension behavior [8, 9].

The most difficult part of the problem is now determining the force exerted on the filament by the surrounding fluid, given that the movement of the filament itself perturbs the fluid. Since the filaments we are considering are microscopic in length, it is reasonable to neglect inertial effects. In this limit the fluid flow equations are linear. However, modeling the interaction of the fluid and filament at an explicit fluid filament interface would be a computationally daunting task. An approximate approach couples the filament and fluid motion by requiring that beads in the model act as Stokeslets (point forces acting in the fluid)[10]. They experience a hydrodynamic frictional force given by $F_H = -\gamma_0 (\mathbf{v} - \mathbf{v}_H)$, where $\mathbf{v}$ is the velocity of the bead and $\mathbf{v}_H$ is the induced fluid velocity at its location. The parameter γ_0 is then the bead friction coefficient. In this model the local fluid velocity $\mathbf{v}_H$ is now a linear combination of the velocity fields generated at that point by each of the

Stokesletts [5, 11, 12], leading to a hydrodynamic force on bead i

$$\mathbf{F}_{iH} = -\gamma_0 \mathbf{v}_i + \frac{\gamma_0}{8\pi\eta} \sum_{i \neq j} \left(\frac{\mathbf{F}_j}{|\mathbf{r}_{ij}|} + \mathbf{F}_j \cdot \frac{\mathbf{r}_{ij}\mathbf{r}_{ij}}{|r_{ij}|^3} \right). \tag{2}$$

The constant γ_0 can also be written in terms of the friction exerted by a sphere of radius a, $\gamma_0 = 6\pi\eta a$, where the constant η is the shear viscosity. We now have a parameter in the model that is the ratio of the bead spacing to the hydrodynamic radius a. The question is now, what value do we choose for a/b? A popular choice is the "shish kebab" model, $a/b = 1/2$. If the hydrodynamic radius is considered equivalent to a real radius this models a filament of touching spheres. That is, it is an approximation to a filament with aspect ratio $1/(n-1)$. Let us consider this in more detail by taking a filament that is aligned with the x-axis spanning the interval $x = [-1, 1]$, where $x(= s/l)$ is the dimensionless contour length and $L = 2l$. The hydrodynamic force in terms of x for the case of an applied external force density ($f^y = F_j^y/b$) acting in the $-\hat{y}$ direction is

$$F_H(x) = -\gamma_0 v_y(x) - \frac{3af^y}{8\pi} ln \left(\frac{(1-x)^2)}{(\beta b/l)^2} \right), \tag{3}$$

where $\beta = e^{-k}$ and k is the Euler-Mascheroni constant defined by

$$k = \lim_{m\to\infty} \left(\sum_{j=1}^{m} \frac{1}{j} - \ln(m) \right) \approx 0.5772. \tag{4}$$

In the steady state the total external force acting on the body equals the total hydrodynamic force, and the filament moves with some terminal velocity U. Using Eq. (3) to calculate the total hydrodynamic force (F^y) in the $\hat{y}$ direction, the average friction coefficient is

$$\gamma^{\perp} = \frac{F^y}{U} = \frac{4\pi\eta L}{ln\left(L/(\beta b)\right) + \frac{2b}{3a^{\perp}} - 1} + O\left(ln^{-3}(L/(\beta b))\right). \tag{5}$$

The exact result from slender body theory is also of this form, but the constant depends on the shape of the filament [13]. For a cylindrical geometry, the result is

$$\bar{\gamma}^{\perp} = \frac{4\pi\eta L}{ln\left(L/r\right) + ln(2) - 1/2} + O\left(ln^{-3}(L/r)\right). \tag{6}$$

Therefore, only when the filament radius is considered to be $r = \beta b \approx 0.561b$ and when the hydrodynamic radius is chosen to be $a^{\perp} = 4b/(3(2ln2 + 1)) \approx 0.559b$ is this model in

agreement with theory to the order of error in the equations. This value is close to, but not equal to, the shish kebab value.

Now we consider motion parallel to the axis, the average friction coefficient to leading order by taking the continuum limit of our discrete model is

$$\gamma^{\parallel} = \frac{2\pi\eta L}{ln\left(L/(\beta b)\right) + \frac{b}{3a^{\parallel}} - 1}\,. \tag{7}$$

whereas, from theory, for a cylindrical geometry

$$\bar{\gamma}^{\parallel} = \frac{2\pi\eta L}{ln\left(L/r\right) + ln(2) - 3/2}\,. \tag{8}$$

So the model only matches theory when the filament radius is considered to be $r = \beta b \approx 0.561b$, and the hydrodynamic radius is chosen to be $a^{\parallel} = 2b/(3(2ln2 - 1)) \approx 1.726b$. In addition, a similar analysis shows that only with these choices of hydrodynamic radii is the correct *variation* of the friction coefficient along the length of the filament recovered to $O(\ln^{-3}(\epsilon))$ in the slenderness parameter $\epsilon = r/l$. This is crucial, because it is this variation in friction along the length that causes a flexible filament to bend when it is set in motion and this is the origin of the effect we are considering here. We conclude that the agreement of the Stokeslett model with analytical results is conditional upon choosing the hydrodynamic radius and hence bead friction coefficient to be the tensor

$$\begin{aligned} \gamma_0^{\perp} &= 6\pi\eta a^{\perp} = 8\pi\eta b\left(2\ln 2 + 1\right)^{-1} \\ \gamma_0^{\parallel} &= 6\pi\eta a^{\parallel} = 4\pi\eta b\left(2\ln 2 - 1\right)^{-1}. \end{aligned} \tag{9}$$

That is, the hydrodynamic radius is different in the parallel and perpendicular directions. This unique parameterization in terms of Stokesletts, appropriate for a cylindrical filament, gives our model its name – the hydrodynamic shape of the filament is more reminiscent of a caterpillar than a shish kebab (see Figure 1). The final form of the hydrodynamic force is now

$$\mathbf{F}_{iH} = -\left(\gamma_0^{\perp}\hat{\mathbf{n}}\hat{\mathbf{n}} + \gamma_0^{\parallel}\hat{\mathbf{p}}\hat{\mathbf{p}}\right)\cdot\left(\mathbf{v}_i - \mathbf{v}_{iH}\right). \tag{10}$$

The vectors $\hat{\mathbf{n}}$ and $\hat{\mathbf{p}}$ are the unit vectors normal and parallel to the axis, respectively.

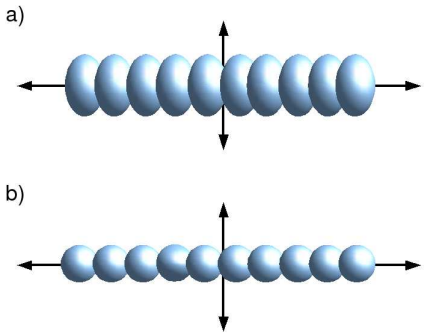

Figure 1: A schematic of a) caterpillar and b) shish kebab hydrodynamic filament shapes.

The caterpillar simulation model is relevant for a broader range problems than the alignment question studied here. Examples range from micro-organism motility [14, 15] to the sedimentation behavior of paper pulp [13]. In the limit that the inhomogeneity of the friction is neglected, Eq. (10) in fact reduces to "resistive force theory" [16]. This accurately predicts the swimming speeds of spermatozoa [15]. Note that the constants from the stokselett model that must be matched to recover the correct result depend on the cross-sectional geometry of the filament. Here we specialize to a cylindrical form, but this is not a necessary restriction. Other geometries can be modeled.

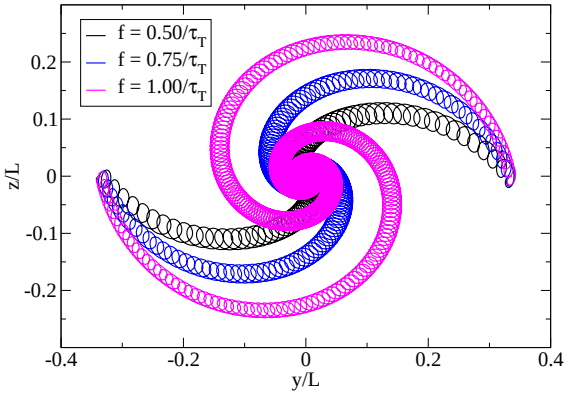

Figure 2: A trace of the ends of the filament during alignment process after subtracting the center of mass motion for three different field frequencies ($B = 1.2$).

We now turn to investigating the possibility of aligning charged filaments using a circularly polarized electric field. Microtubules are charged biofilaments that have been shown to respond to moderate, experimentally accessible electric fields [1]. Carbon nanotubes, on the other hand, are uncharged but can be functionalized to give an effective electrical charge [17]. We carried out a series of simulations using the model described above in experimentally accessible regimes of parameter space. The functional form of the time-dependent applied electric field is

$$\begin{aligned} E_y &= E\cos(2\pi f t)\,\hat{y} \\ E_z &= E\sin(2\pi f t)\,\hat{z}, \end{aligned} \tag{11}$$

where the constants E and f are the field magnitude and frequency, respectively, and t the time. In all cases we set $L = 1$ and the number of beads in the model to 80. Following the argument above, we are considering a cylindrical filament with aspect ratio $\sim 1:80$. For a microtubule (diameter 25 nm), this would typically correspond to a length of a micron. We define a dimensionless force $B = L^3 E\tilde{q}/\alpha$ that characterizes the magnitude of the electric forces to the elastic forces. Here, $\tilde{q}$ is the charge density of the filament. When $B \gg 1$, significant deformation is expected, whereas when $B \ll 1$ elastic forces dominate and the filament will remain predominantly straight. We can estimate experimentally accessible values of B for microtubules. From Ref. [1], the average length L was 5 μm, the flexure α is of the order 10 $pN\mu m^2$, and the effective linear charge density was measured to be $\tilde{q} = 280\ e/\mu m$. The microtubules remained stable in a field of 20 V/cm. Using these values, we calculate that $B \sim 1$ is easily achieved experimentally. Experiments carried out by van den Heuvel *et. al* actually achieved much higher values, and pronounced bending was indeed observed [6] . In our simulations B is near the modest value of unity. We also define a characteristic time $\tau_T = \bar{\gamma}^{\perp} L/E\tilde{q}$, which is the amount of time it takes the filament experiencing an external field of magnitude E to translate transversely a distance of its length.

The simulations predict that a charged body placed in the field given by Eq. (11) gyrates in the yz plane following the direction of the applied field. The center of mass motion occurs

concurrently with a hydrodynamic orientation due to bending of the filament. Traces of the location of the endpoints of the filament after subtracting the center of mass motion for representative simulations are shown in Figure 2. Here, the filament is initially offset at an angle of 45 degrees in the xy plane, but in time it aligns itself with $\hat{x}$, perpendicular to the plane of the polarized field. An animation of the typical dynamics can be viewed in Ref. [1]. Our results therefore suggest that circularly polarized electric fields are a indeed a possible means for aligning charged filaments. Before we can propose that the method is also *practical* there are other things to consider.

The first is as follows. The filament reorientates and, once it is aligned, its motion resembles sedimentation constrained to the surface of a cylinder. If the radius of this cylinder is too large then there is pronounced rotational motion and this is undesirable. The magnitude of this steady state gyration radius R can be estimated using simple scaling arguments. We expect $2\pi R \sim \omega/f$, where ω is the tangential velocity. When the frequency of the electric field is chosen to be $f = \tau_T^{-1}$ and $\omega = L/\tau_T$, $R \sim L/2\pi$, independent of B the field strength. This expression is exact for $B \to 0$, where bending is insignificant. We further confirmed from simulations, with a dimensionless force ranging from $B = 0.08 - 1.6$, that the expression for R remains sufficiently accurate for values of B around unity to provide a reasonable estimate for the spatial extension of the rotation of the filament during the alignment process. From this we can conclude that, so long as the frequency is around τ_T^{-1} or higher, the rotation can be localized to lengths of the order of the length of the filament.

To now quantify the time scale of hydrodynamic alignment, we define the hydrodynamic alignment time τ_H as the time taken for the angle between the filament axis and $\hat{x}$ to change by ten degrees. The first parameter we consider is the magnitude of the frequency of the applied field. In the limit where $f \ll \bar{\tau}_H^{-1}$, where $\bar{\tau}_H$ is the hydrodynamic alignment time in a static field ($f = 0$), we recover the results discussed in Ref. [5]. As we increase the frequency while keeping B constant, the alignment time increases. This dependence is shown in Figure 3. The filament spends an increased amount of time changing orientation

[1] http://www.cmth.ph.ic.ac.uk/people/aimee.bailey/movie.mov (also *.avi)

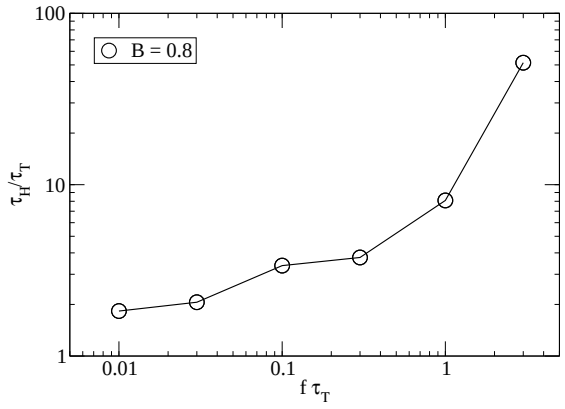

Figure 3: Hydrodynamic alignment time, τ_H, as a function of frequency (B = 0.8).

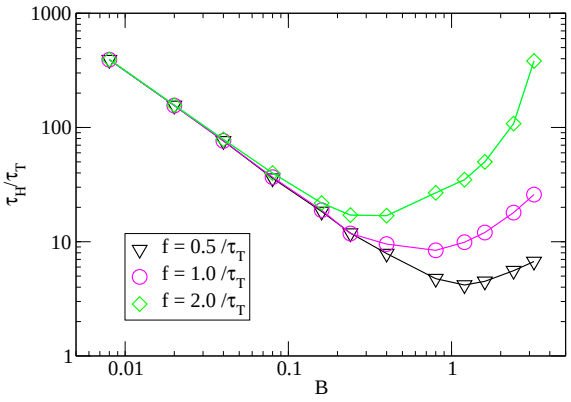

Figure 4: Hydrodynamic alignment time, τ_H, as a function of the dimensionless field strength, B.

to adjust to the alternating direction of the field. The result is that when the frequency is too high, the filament takes an impractically long amount of time to align. So long that the effect of diffusion cannot be ignored and a deterministic simulation is no longer valid (see below). When the frequency is approximately τ_T^{-1} then, for $B \sim 1$, the alignment time is (using values for microtubules reported in Ref. [1]) $\tau_T \sim 1s$. That is, a modest frequency of one cycle per second – an experimental set-up straightforward to implement.

The dependence of τ_H on the dimensionless field strength B is shown in Figure 4. One can observe two scaling regimes. For low B's, the observed relationship is $\tau_H \sim \gamma^{\perp}/\tilde{F}B$,

which is consistent with that observed in Ref. [5] for alignment in a static field. It could be predicted by noting the analytical expression for the torque from Ref. [18] scales with $\sim \tilde{F}B$. For high B's, the hydrodynamic alignment time obeys a different scaling relationship: τ_H actually increases with B. This is because the frequency for these simulations is chosen to be proportional to τ_T^{-1}, so as B increases, so does the frequency and therefore the alignment time, discussed in the previous paragraph. The crossover of scaling behavior therefore occurs where $f^{-1} \sim \bar{\tau}_H$.

Other forces are present that will compete with the hydrodynamic forces driving the alignment. Can these be ignored? As long as the hydrodynamic alignment time is shorter than all other time scales, hydrodynamic alignment will dominate these effects. In reality thermal forces act to randomize its orientation. The time scale for rotational diffusion is roughly $\tau_D \sim \gamma^{\perp} L^2/kT$. We can neglect this when $\tau_H/\tau_D \sim L/B^2\lambda \ll 1$, where λ is the persistence length ($\lambda = \alpha/kT$). For a 5 μm microtubule, this condition is satisfied as long as $B > 0.1$. Additionally, microtubules have a dipole moment along their length, reported in Ref. [1] to be $d_{el} \sim eL$. The time scale associated with the alignment of the dipole with the field is approximately $\tau_d \sim \gamma^{\perp} L^2/d_{el}E$. To ensure that this is negligible requires that $\tau_T/\tau_d \sim d_{el}/\tilde{q}L^2 \sim 1/280L \ll 1$. Consequently, for a microtubule longer than a micron, dipolar reorientation should also be negligible.

A final complication is the presence of other filaments. The analysis up to this point has focused on a single filament whereas, in reality, there will likely be others within close proximity. Do neighbors hinder alignment? We considered the scenario of two filaments separated by a distance d, exposed to the same field given by Eq. (11). At the start of the simulation, one filament is aligned with the $\hat{x}$ direction, and the second is tilted at an angle of 60 degrees in the xy plane. The alignment time was measured for multiple separations. In the regime where $d/L > 1$, the filaments behave as isolated entities. This is consistent with the results from studies of cooperative motion in a static field carried out by Llopis *et al.* [19]. For our purposes, it suffices to say as that as long as the solution is at a low enough concentration, neighboring filaments should not inhibit alignment.

To conclude, the caterpillar hydrodynamic model predicts that it is practical to align charged filaments in a prescribed direction using a circularly polarized electric field. The model also gives the optimal parameters for achieving this alignment. We hope that this will motivate experimental studies and ultimately provide a useful tool for nanotech applications.

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