

Effective viscosity and tearing rate of polymer networks in the presence of cross-link slip

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Abstract

We are trying to describe long timescale rates of deformation in semi-flexible filament networks with transient cross-links. We have addressed the problem using a simplified model in which cross-links are allowed to slip past one another in a linear drag-like manner. This model gives a prediction for the long timescale effective viscosity of the medium that depends on network architecture and effective drag coefficient between filaments. We predict different modes of anomalous behavior when the network undergoes large strains that drive it into an anisotropic configuration. We have verified our solution using computational models of filaments undergoing shear and extensional stress. In addition, we attempt to derive a time constant for network tearing. In this model, we find that the inclusion of slip gives rise to varying levels of connectivity even when network architecture alone would predict a fully connected structure.

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I. INTRODUCTION

Cross-linked networks of semi-flexible polymers are a class of materials with poorly understood but highly interesting properties. They are often studied in conjunction with their important role in the formation of the cell cytoskeleton, the structural underpinning of eukaryotic life. In addition to the important role of these types in living organisms[7, 24], *in vitro* studies have revealed interesting mechanical properties of these composite networks on their own, which has spurred much interest in soft-matter physics[4].

A. Rheological Observations of Cross-link Dominated Response

A key area of inquiry in soft matter physics concerns the long timescale motion of cross-linked polymer networks as this may govern physiologically important behaviors[27].

The mechanics of semi-flexible polymer networks at short timescales have been studied in depth in a multitude of *in vitro* studies[10, 13, 29]. And many studies using more compliant cross-linkers showed that cross-links mechanics governed many short timescale rheological properties[8, 9, 14, 18, 19, 33]. More recent work has begun to highlight the long-timescale importance of transient cross-linking dynamics.[16, 17, 20, 36].

In these studies, long timescale creep behaviors are thought to arise predominantly from the transient nature of binding in most biologically relevant cross-linkers[17]. The importance of cross-link dynamics in determining the mechanical response of semi-flexible polymer networks has been known for at least 20 years[31]. Nevertheless, the dependence of network rheology on cross-link compliance and binding rates is still a subject of much research[22].

B. Overview of Theoretical Descriptions of Cross-linked Networks

A number of successful theoretical descriptions of cross-linked networks have been introduced into the literature[2, 11, 12, 28, 35]. For a comprehensive but straightforward introduction to this field we recommend [4]. We wish primarily to expand upon current theories of composite semi-flexible polymer and cross-link networks to incorporate both nonlinear extension and transient binding of cross-links.

Many theoretical methods have sought to model cross-link binding and unbinding directly[3, 22], and previous modeling work often take cross-links as extended springlike structures [15] separate from the main semi-flexible filament constituents. We simplify our approach to coarse-grained filaments which are able to slide past each other as molecular bonds form and rupture, akin to coarse-grained models of molecular friction[6, 26, 30]. This drag-like coupling has been shown to be an adequate approximation in the case of ionic cross-linking of actin[5, 32], and can be found in the theoretical basis of force-velocity curves for myosin bound filaments[1]. We propose that it will form a suitable bulk approximation in the presence of super molecular cross-links as well.

Importantly, this simplification allows us to extend our single polymer models to dynamical systems of larger network models for direct comparison between theory and modeling results. This level of coarse graining will therefore make it easier to understand classes of behavior for varying compositions of cross-linked filament networks. In addition, it gives concrete predictions for behaviors in widely different networks with measurable dependencies on molecular details.

II. EXPLANATION OF MODEL

A. Composite Cross-link & Filament Representation

We consider individual semi-flexible polymers as chains of springs of relaxed length l_s , whose orientations are linearly coupled to their neighbors. Filaments can be represented as a sequence of nodes with positions \mathbf{x}_i and nearest neighbor interactions of the form

$$|F_{i,i+1}|_{\parallel} = -\mu \cdot \frac{|\mathbf{x}_{i+1} - \mathbf{x}_i| - l_s}{l_s} \quad (1)$$

$$|F_{i,i+2}|_{\perp} = -\frac{\kappa}{l_s^2} \cdot \text{acos} \left(\frac{\mathbf{x}_{i+2} - \mathbf{x}_{i+1}}{|\mathbf{x}_{i+2} - \mathbf{x}_{i+1}|} \cdot \frac{\mathbf{x}_{i+1} - \mathbf{x}_i}{|\mathbf{x}_{i+1} - \mathbf{x}_i|} \right) \quad (2)$$

This is essentially a discretized equivalent to a model of filaments with separable extensional and bending moduli like the type in [11] with a potential defined by

$$\mathcal{H} = \frac{1}{2}\kappa \int ds (\nabla^2 \mathbf{u})^2 + \frac{1}{2}\mu \int ds \left(\frac{dl(s)}{ds} \right)^2 \quad (3)$$

where, μ represents an extensional modulus of a filament, and κ represents a bending modulus.

Here, the extensional and bending moduli are taken as composite quantities related to both filament and cross-linker compliance in a manner similar to a recently proposed effective medium theory[2]. We provide a molecular level derivation of this composite compliance in Appendix B, but for now we wish to highlight the main features.

In the limit of highly rigid cross-links and flexible filaments, our model clearly reduces to the pure semi-flexible filament models of [11, 34]. In the opposite regime of nearly rigid filaments and highly flexible cross links, our method is still largely similar to the model of [2] in small strain regimes before any nonlinear cross link stiffening. However, in departure from those models, the magnitude of the force on interior cross-links in our model is still the same as those on the exterior. This is a simplification of the varying levels of strain that would actually be present in these cross-linkers, but we choose to ignore the slight variation in favor of an approximate, universal approach. Finally, in the event that the induced strain of the filament and the cross-linker are of comparable scales, our composite stiffness can be expressed by the approximation *huh* as we shown in Appendix B.

In our simulations we explore the role that nonlinear stiffening of filaments or cross linkers would play, and what complications arise.

B. 2D Network Formation

We choose to focus our attention on 2D networks both for their tractability as well as their relevance in the quasi-2D cytoskeletal cortex of many mammalian cells[21]. In addition, recent developments in 2D *in vitro* systems[23, 25], make 2D models all the more interesting as a renewed focus of study.

We are using a minimal network (Mikado model) of connected unstressed linear filaments in a rectangular 2D domain. We generate 2D networks of these semi-flexible filaments by laying down straight lines of length, L , with random position and orientation. We then assume that some fixed fraction of overlapping filaments become cross-linked (defined in IIC) at their point of overlap.

Although real cytoskeletal networks may form with non-negligible anisotropy, we choose to focus our attention on isotropically initialized networks for simplicity. We define the density using the average distance between cross-links along a filament, l_c . A simple geometrical argument can be used to derive the number of filaments filling a domain as a function of L and l_c [11]. However, for our purposes we take the approximation that the number of filaments needed to tile a rectangular domain of size $W \times H$ is $2WH/Ll_c$, and that the length density is therefore $1/l_c$.

In the absence of cross-link slip, we expect the network to comprise a connected solid with a well defined elastic modulus[11, 34]. These networks are only well-connected when the ratio of filament length to intercrosslink spacing, L/l_c is greater than ~ 6 . Near this percolation threshold, there are only locally connected domains, and discussions of global network properties becomes less reasonable. Additionally, as the filament density is increased beyond this point, there is another transition between non-affine bending and affine stretching of filaments, which changes the dominating term of the network's elastic modulus.

C. Drag-like Coupling Between Overlapping Filaments

In departure from the previous models, we wish to incorporate relaxation of the network's stored stress by letting the attachment points slip. We do this by replacing the fixed with a drag-like coupling between filaments.

$$\mathbf{F}_{\text{drag}} = \xi \cdot \int ds (\mathbf{v}(\mathbf{s}) - \mathbf{v}_0(\mathbf{s})) f(s) \quad (4)$$

Where $f(s)$ represents the locational distribution of cross-link points (equal to 1 at locations of cross-links and 0 elsewhere). This model assumes a linear relation between applied force and the velocity difference between attached filaments. Obviously, non-linearities can arise in the presence of force dependent detachment kinetics as well as non-linear force extension of cross-links. In particular, we address non-linear effects of stress induced unbinding

in Appendix A. Assuming inhomogeneities from non-linear effects are of second order, the motion for the entire network is governed by a dynamical equation of the form

$$\int ds (\zeta \mathbf{v}_i(\mathbf{s}) + \xi \sum_j (\mathbf{v}_i(\mathbf{s}) - \mathbf{v}_j(\mathbf{s})) p_{ij}(s)) = \nabla \mathcal{H}_i \quad (5)$$

Here, the first term in the integral is the filament's intrinsic drag through its embedding fluid, ζ , while the second comes from the drag-like coupling between filaments, ξ .

D. System of Equations for Applied Stress

We model our full network as a coupled system of differential equations satisfying 5. Although the general mechanical response of this system may be very complex, we wish to focus our attention on low frequency deformations and the steady-state creep response of the system to an applied stress. To do this we introduce a fixed stress, σ along the midline of our domain. This stress will point in the direction, $\hat{\mathbf{u}}$, producing either shear ($\hat{\mathbf{u}} = \hat{\mathbf{x}}$) or extensional ($\hat{\mathbf{u}} = \hat{\mathbf{y}}$) stress.

Finally, we add a 0 velocity constraint at the far edges of our domain of interest. We assume that our network is in the "dry," low Reynold's number limit, where inertial effects are so small that we can equate our total force to 0. Therefore, we have a dynamical system of wormlike chain filaments satisfying

$$\int ds (\zeta \mathbf{v}_i(\mathbf{s}) + \xi \sum_j (\mathbf{v}_i(\mathbf{s}) - \mathbf{v}_j(\mathbf{s})) p_{ij}(s)) = \nabla \mathcal{H}_i + \sigma \hat{\mathbf{u}} \cdot \delta(x - D) \quad (6)$$

subject to constraints such that $\mathbf{v}_i(\mathbf{x})$ is 0 with $x = 0$. This results in an implicit differential equation for filament segments which can be discretized and integrated in time to produce a solution for the motion of the system.

III. ANALYTICAL DERIVATIONS

A. Low Strain Approximation of Effective Viscosity

We would like to begin with a calculation of a low-strain estimate of the effective viscosity for a network described by our model. We carry this out by assuming we have applied a

constant stress along a transect of the network. With moderate stresses, we assume the network reaches a steady state affine creep. In this situation, we would find that the stress in the network exactly balances the sum of the drag-like forces from cross-link slip. So for any transect of length D , we have a force balance equation.

$$\sigma = \frac{1}{D} \sum_{\text{filaments}} \sum_{\text{crosslinks}} \xi \cdot (\mathbf{v}_i - \mathbf{v}_0) \quad (7)$$

where $\mathbf{v}_i(\mathbf{s}) - \mathbf{v}_j(\mathbf{s})$ is the difference between the velocity of a filament at it's cross-link point and the velocity of the filament to which it is attached. We can convert the sum over cross-links to an integral over the length using the average density of cross-links, $1/l_c$ and invoking the assumption of (linear order) affine strain rate, $\mathbf{v}_i - \mathbf{v}_0 = \dot{\gamma}x$. This results in

$$\begin{aligned} \sigma &= \frac{1}{D} \sum_{\text{filaments}} \xi \cdot \int_0^L ds (\mathbf{v}(\mathbf{s}) - \mathbf{v}_0(\mathbf{s})) \frac{1}{l_c} \\ &= \sum_{\text{filaments}} \frac{\xi \dot{\gamma} L}{l_c} \cos \theta \cdot (x_l + \frac{L}{2} \cos \theta) \end{aligned} \quad (8)$$

Here we have introduced the variables x_l , and θ to describe the leftmost endpoint and the angular orientation of a given filament respectively. Next, to perform the sum over all filaments we wish to convert this to an integral over all orientations and endpoints that intersect our line of stress. We assume for simplicity that filament stretch and filament alignment are negligible in this low strain approximation. Therefore, the max distance for the leftmost endpoint is the length of a filament, L , and the maximum angle as a function of endpoint is $\arccos(x_l/L)$. The linear density of endpoints is the constant $D/l_c L$ so our integrals can be rewritten as this density over x_l and θ between our maximum and minimum allowed bounds.

$$\sigma = \frac{1}{D} \int_0^L dx_l \int_{-\arccos(\frac{x_l}{L})}^{\arccos(\frac{x_l}{L})} \frac{d\theta}{\pi} \frac{\xi \dot{\gamma} L}{l_c} \cdot \frac{D}{L l_c} \cdot (x_l \cos \theta + \frac{L}{2} \cos^2 \theta) \quad (9)$$

Carrying out the integrals and correcting for dangling filament ends leaves us with a relation between stress and strain rate.

$$\sigma = \frac{(L - 2l_c)^2 \xi}{4\pi l_c^2} \dot{\gamma} \quad (10)$$

We recognize the constant of proportionality between stress and strain rate as a viscosity. Therefore, our approximation for the effective viscosity, η_{eff} , at steady state creep in this low strain limit is

$$\eta_{eff} = \frac{(L - 2l_c)^2 \xi}{4\pi l_c^2}. \quad (11)$$

B. Effect from Large Strains

This derivation is only valid in the limit of moderate strains. If filaments have significant compliance or the network is allowed to deform far from its initial conditions various . The general .

However, it isn't immediately clear how large these components of the strain will be or over what timescale they might take effect. We next aim to determine the dependence of these effects on the microscopic parameters of our model.

Assuming that no drastic filament alignment has occurred, then phase c will closely resemble our linear approximate, aside from a relatively minor geometrical factor due to the correlations between orientation and filament extension (See Appendix C). The effects of phase b and phase d are quite drastically different so we approach their separately.

1. Transient Softening from Slow Filament Stretch

Our approximation was derived in the limit of rigid rods. In the case of filaments with non negligible compliance, we might expect there to be some interplay between filament stretching and effective viscosity. Indeed, our simulations below uncover a noticeable decrease in the short-term viscosity even beyond the timescale of relaxation to mechanical equilibrium. This decrease in the observed viscosity actually decays away as filaments relax due to cross-link slip. This leads to a less than linear time dependence in the creep response curves that lasts for long times.

In general, filament stretch gives rise to two dominant mechanical responses. First we see a rapid stretching of the filaments, corresponding roughly to the degrees of freedom allowed in the irreversibly cross linked case such as [11]. In the second phase, filaments are able to stretch farther as constraints from the cross-linking points relax too.

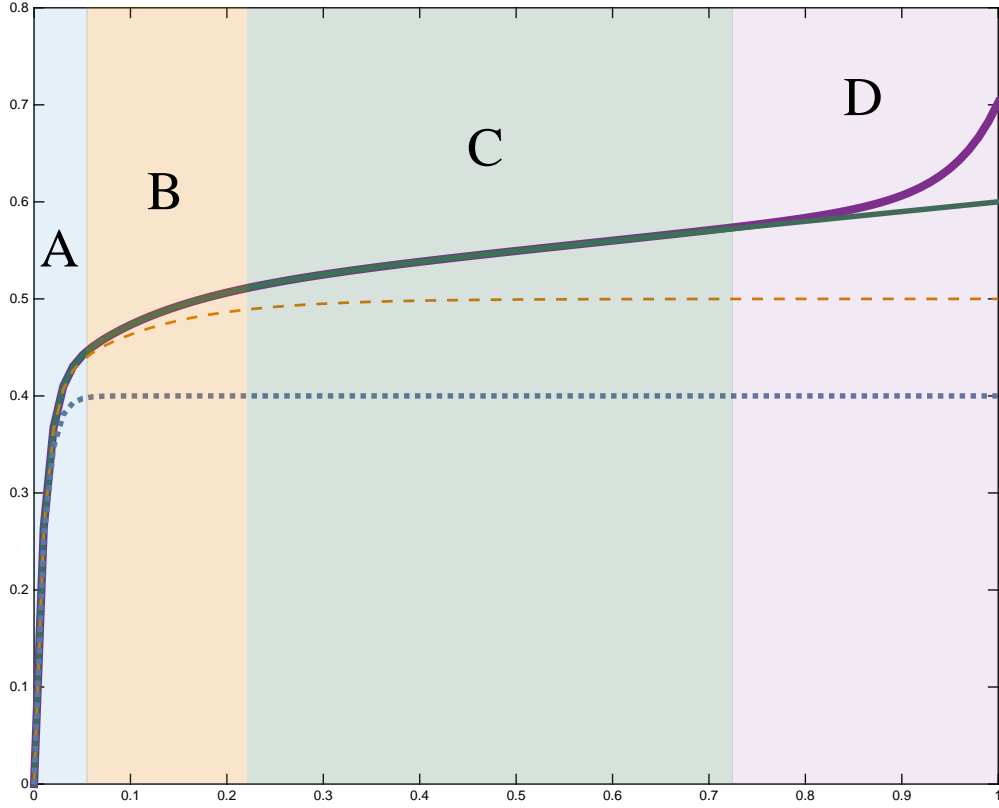


FIG. 1. General schematic of the creep response of compliant filament networks illustrating the 4 phases of deformation: a) rapid mechanical response, b) combination of slow filament stretching and cross-link slip, c) cross-link slip dominated, d) network tearing from filament alignment.

We see a broadening of the filament stretch distribution away from the affine approximation $\langle \delta L/L \rangle_0 = \gamma_{xy} \sin(\theta) \cos(\theta)$. Assuming for the time being that this broadening is randomly distributed throughout the network we have $\delta L/L = \langle \delta L/L \rangle_0 + \epsilon(t) \cdot \mathcal{N}$. This has an effect on the total mechanical energy stored in the network $\mathcal{H} \sim \langle \delta L/L \rangle^2 = \langle \delta L/L \rangle_0^2 + \epsilon(t)^2$. Thus, the network will appear less viscous while some strain energy is being stored in the further stretching filaments.

Eventually the strain rate from slow filament stretching will become negligible to that due to pure cross-link slip on rigid rods. We wish to define the timescale of the relaxation between the distribution of network strain at purely mechanical equilibrium and that in the equilibrium with network slip. This will set the timescale over which the creep response of

the network will be sub-linear.

2. Filament Alignment And Network Tearing

This will remain valid as long as any ordering by the induced strain is still relatively small. However, if the network reorganization is relatively fast due to high stresses, we find that our approximation no longer holds.

At this point even the mean filament stretch becomes anisotropic $\langle \delta L/L \rangle \neq \gamma_{xy} \sin(\theta) \cos(\theta)$, with a larger number of filaments undergoing extension than compression. This results in a tighter peak around $-pi/4$ and a broader one at $pi/4$.

C. Extensional Thinning and Network Tearing

For moderate extensional stresses, the rigid filament approximation of the effective viscosity simply picks up a different geometrical factor out front.

However, at higher stress and in the presence of different things happen.

$$\frac{\partial l_c}{\partial t} = l_c \dot{\gamma} = \frac{l_c \sigma}{\eta} \sim l_c^3 \frac{\sigma}{L^2 \xi} \quad (12)$$

We can see that the rate of network thinning accelerates as we would expect. When the network reaches some minimum connectivity we assume that it stops behaving as a continuum material and the network tears irreversibly.

$$\tau_{break} = \frac{\eta_{eff}}{2\sigma} \cdot \left(1 - \frac{l_c^2}{l_{break}^2} \right) \quad (13)$$

This provides us with an estimate of the timescale of catastrophic breakdown for a network with a given initial architecture and molecular drag.

D. Frequency Dependence of Elastic Modulus

Finally, we wish to extend this analysis to have some idea of the frequency dependence of the complex elastic modulus. I sure hope there is some way to do this.

IV. ANALYSIS OF SIMULATION RESULTS

Next, we wanted to test our analytical conclusions on a computational model. The technical details of the model can be found in the Appendix, but we summarize the main modeling points here.

A. Computational Simulation Method

We discretize the filaments such that the equations of motion becomes a coupled system of equations for the velocities of filament endpoints, \mathbf{x} . The drag-like force between overlapping filaments results in a coupling of the velocities of endpoints.

$$\mathbf{A} \cdot \dot{\mathbf{x}} = \mathbf{f}(\mathbf{x}) \quad (14)$$

where \mathbf{A} represents a coupling matrix between endpoints of filaments that overlap, and $\mathbf{f}(\mathbf{x})$ is the spring force between two endpoints. We can then numerically integrate this system of equations to find the time evolution of the positions of all filament endpoints.

We generate a network by laying down filaments with random position and orientation within a domain of size $2D$ by D with periodic boundaries. The external stress (shear or extensional/compressional) is applied to all filament endpoints falling within a fixed x -distance from the center of the domain. Finally, filament endpoints falling within a fixed x -distance from the edges of the domain are constrained to be nonmoving.

The nominal units for length, force, and time are μm , nN , and s , respectively. We explored parameters space around an estimate of biologically relevant parameter values, given in Table X.

For computational simplicity in these models, unless otherwise mentioned we assume that the bending rigidity, κ , is infinite. This allows us to model filaments as non-bending springs of rest length, L , and spring modulus μ . In the appendix, we show that our result is not significantly different from the result for semi-flexible polymers.

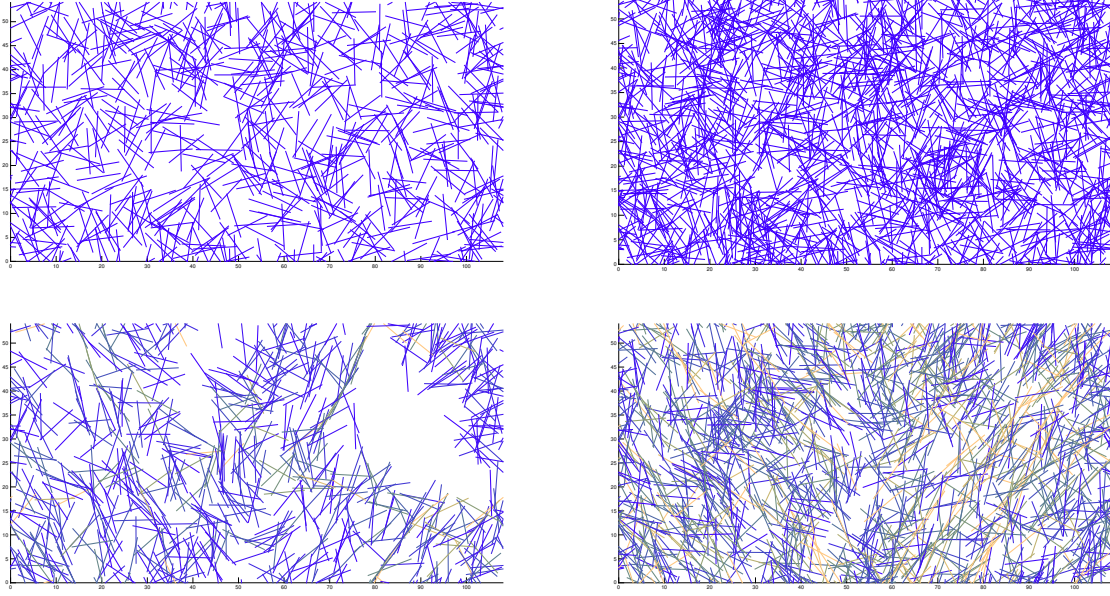


FIG. 2. Two Simulation setups with $L = 9\mu m$, $D = 54\mu m$ before and after 1000s of applied stress.
a) low density $l_c = 2\mu m$, b) moderate density $l_c = 1\mu m$

B. Behavior at Moderate Strains above Percolation Limit

Beginning with moderate strains ($\gamma < 0.2$), our computational simulations show that in the high density limit, our theoretical approximation from Eqn 11 is highly accurate at explaining the network behavior. Aside from a geometrical factor, our approximation is valid for both shear and extensional stresses applied to the network.

As the density of the network approaches the breakdown limit, the effective viscosity diverges from our expected value. At the low connectivity limit, our expected viscosity goes to 0, but the medium viscosity begins to take over as we cross the percolation threshold.

In addition to changing the architecture and effective drag coefficient, we also validated the generality of our approximation by varying simulation size, medium viscosity, filament stiffness, and applied stress. We were able to find a slight trend that depended on filament stiffness as indicated in the difference between blue and red data points in Figure 3. This deviation from our approximation manifested itself more strongly when filaments were highly compliant, and we therefore interpret this as a higher order manifestation of the strain induced reordering of our networks which we focus on in the next section.

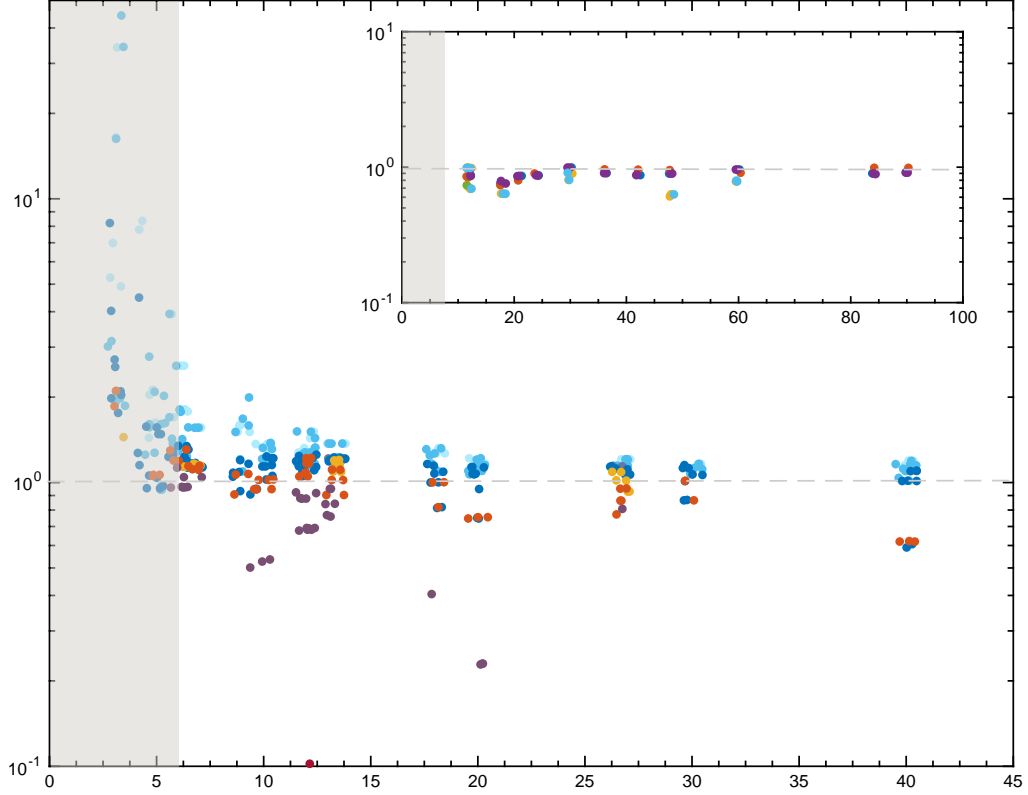


FIG. 3. Ratio of effective viscosity measured by shear simulation to predicted effective viscosity as a function of connectivity, L/l_c . Inset: Same measurement for extensional simulations

C. Behavior at High Shear Strain

Large scale reorientation of cross-linked filaments under strain is a subject of active research at the moment [Sayantan]. Therefore, we wished to use our computational approach to extend our understanding of generic filament networks in the regime of shear induced anisotropy. Because we allowed variation in both the extensibility of individual filaments and the drag-like coupling between filaments, we found that we could induce reordering from high strain via either large filament compliance or large slip (or both). As the implications for each context are different, we address both cases separately.

1. *Compliant Network, Low Cross-link Slip*

If the elastic deformation of the network is larger than the deformation from cross-link slip, the system enters a regime of high network anisotropy. Under these conditions, large scale reordering drives the network into a highly anisotropic distribution of filament orientations and lengths long before significant cross-link slip begins to occur. This gives rise to a more complex relationship determining the number of effective cross-links that are connecting the network.

Perhaps more interestingly, we also see the interplay between filament stretching and cross-link slip give rise to a long timescale deviation from purely viscous behavior. When cross-link slip is slow, we find this leads to two phases of filament stretching. In the first phase, filaments undergo large-scale dilation or contraction depending on their orientation relative to the direction of shear. However, some portion of the filament is still "under-stretched" due to local constraints from cross-links. As the cross-link slip allows these constraints to be removed, the filament is able to stretch more slowly.

This occurs on a timescale similar to that of cross-link slip and causes the effective viscosity to decay back toward the rigid limit over a time on the scale of τ_{slip} . This gives rise to a less-than-linear creep response during times after the initial elastic relaxation but before full filament relaxation from cross-link slip. In Figure 3, we show that the time dependent effective viscosity increases back toward the isotropic estimate as the slip-derived strain becomes of the same scale as that from pure mechanical stretching.

We are still investigating how the mechanics that allows this reorientation to give rise to a transient softening.

2. *Rigid Network, High Cross-link Slip*

If, on the other hand, our filament stiffness is very high, large scale strains manifest themselves in a very different way.

We find that over time, the orientational distribution of the filaments begins to peak around 45 degrees as the induced alignment $\langle \cos^2 \theta \rangle$ increases. This reorientation causes there to be fewer bonds bridging the network perpendicular to the line of strain, which results in a lower connectivity. In addition, we see formation of subdomains

and the deformation from cross-link slip begins to approach the order of the systems size As diagrammed in Figure,

D. Tearing Events During Extensional Strain

This behavior is caused primarily by the low density network undergoing tearing events that interrupt global connectedness.

E. Subnetwork Formation from Nonlinear Filament Extension

F. Nonlinear Extension and Long-Lived Strain Memory

Finally, we found an interesting behavior when we introduced non-linear extensional stiffness into our filaments. This behavior mimics recent experiments in filamin cross-linked networks. Filamin provides a high level of compliance to a network ($\gamma_0 > 0.5$) without substantial cross-link unbinding. This allows large scale rearrangements to take place without driving very much cross-link slip, similar to the conditions in section IV C 1. However, if we force individual filaments to undergo a strongly nonlinear stiffening at strains above 10%, we find an interesting long term "strain storage."

For moderate strains, this result is largely the same as the result for extensional stress. However, at larger deformations, extensional networks tear apart To further explore the occurrence of tearing events ...

We explored a mode of deformation highly relevant to cortical mechanics in vivo. Under this deformation stress was applied between a region of extension and region of compression. Interestingly, until nearing the point of breakdown, the network did not experience a significant change in effective viscosity. This was due to the between the diminishing viscosity of the thinning domain and the increasing viscosity of the thickening domain.

Finally, we wished to explore the non-linear effects of reorientation of the filaments and non-linear network thinning/thickening. To do so, we applied oscillatory shear

V. DISCUSSION

Finally I wax philosophical, but who is going to pay for the ink?

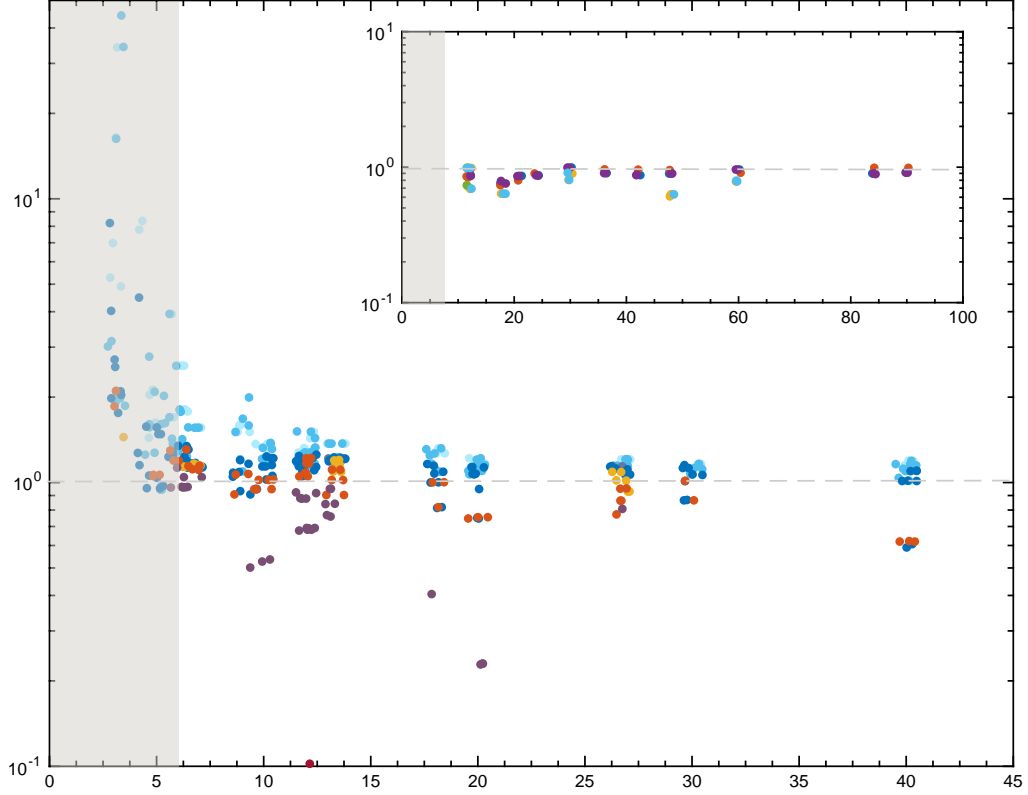


FIG. 4. Analysis of viscosity softening under large shear strain. Inset: time scaling of creep response vs relative filament compliance

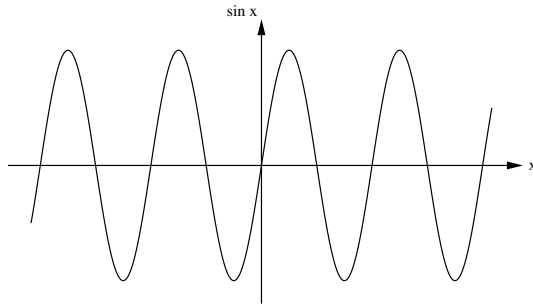


FIG. 5. Tearing rate as a function of L/l_c .

Appendix A: Deriving Molecular Drag Coefficients

Thus far, the idea of a molecular drag coefficient was taken as a phenomenological, measured parameter for a given experimental setup. While this is a sufficient pragmatic

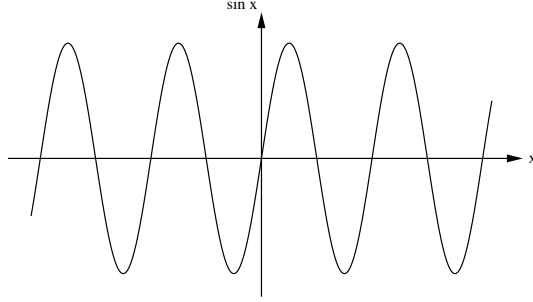


FIG. 6. Frequency dependence of elastic moduli.

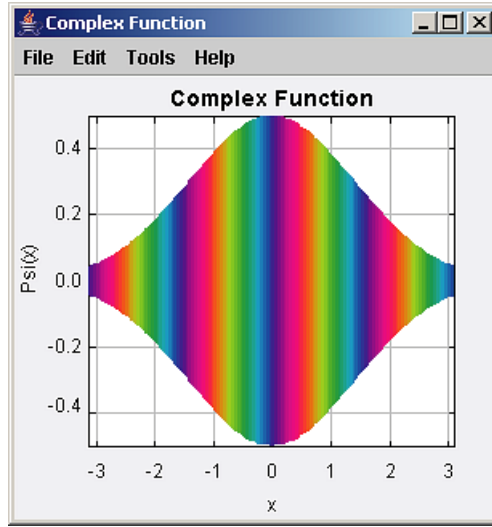


FIG. 7. Phase diagram of network connectivity.

justification, it's useful to try to motivate the quantitative value of this drag coefficient by connecting it to the underlying cross-link properties of binding affinity, concentration, and extensibility.

To do this we'll imagine the simplified case of two cross linkers sliding past each other in one dimension. In this case, imagine that we have an equilibrium number of bound cross-linkers, n_B , each of which can be displaced from its equilibrium length by some distance x . Each cross linker unbinds with rate k_{off} and rebinds at it's relaxed position ($x = 0$) with rate k_{on} . At the same time, all the cross linkers are being pulled from their relaxed position at a rate, v , which is simply the rate at which the filaments are sliding past each other.

We can right down an equation for the change in the density of cross-links as they are pulled upon, bind, and unbind.

$$\frac{\partial \rho}{\partial t} = -k_{off}\rho(x) - v\frac{\partial \rho}{\partial x} + k_{on}\delta(x) \quad (A1)$$

Recognizing that $\int \rho(x) = n_B$ implies $k_{on} = k_{off}n_B$, we can find the steady state solution

$$\rho(x) = \frac{n_B k_{off}}{v} \cdot \exp\left(-\frac{k_{off}}{v}x\right) \quad (A2)$$

If each cross-link has a spring constant μ_c , then we can equate the force on all cross-links to the applied force that is sliding the filaments past each other. Realistically, the spring constant and binding affinity would be functions of the cross-link stretch, but here we are taking them as approximately constant.

$$\int_0^\infty \rho(x)\mu_c x dx = v \frac{\mu_c n_B}{k_{off}} = F_{app} \quad (A3)$$

a

Therefore, the term next to v, (i.e. $\frac{\mu_c n_B}{k_{off}}$) would be equal to our molecular drag coefficient, ξ . assuming, approximately 1-5 cross links per filament overlap, and using the following table of estimates pulled from Ferrer et al., we can chart the accuracy of this simple predictive model.

cross-linker type	α -actinin	filamin-A
dissociation constant (s^{-1})	0.4	0.6
spring constant ($nN/\mu m$)	455	820
drag coefficient ($\frac{nN \cdot s}{\mu m}$)	200-1000	500-2500

This molecular description assumed both a constant off-rate and linear force extension of cross-links. In the event that binding kinetics are regulated by the state of extension, we would expect (based on Rf) to find a region that exhibits a stick-slip behavior instead of the smooth. Based on the coupling between cross-links this could either manifest itself as a global stick-slip behavior or as a heterogenous mixture of jammed and sliding cross-links. It would be interesting to explore this topic further in the future, but as neither phenomenon is observed experimentally, we choose to ignore these nonlinear effects for the time being.

Appendix B: Deriving Filament and Cross-Link Composite Extensional Modulus

Section describing how you derive the extensional modulus.

Appendix C: Rigid Rod Approximation with Non-Uniform Distributions of Length and Orientation

Section describing how you derive the extensional modulus.

Appendix D: Semiflexibility

Brief section showing that the results are not thoroughly flummoxed by semi flexibility.

Appendix E: Simulation details

All changes in the force felt by an endpoint are made smooth to allow integration of the differential equation (i.e. moving between stress domains, constraint domains, and overlap coupling occurs smoothly to prevent discontinuities). Parameter conditions that cause instabilities are excluded, and the endpoint trajectories are integrated out to at least 1000 seconds. In addition, because we wish to probe the behavior of large scale network deformations, we are neglecting the sub-dominant effects from small thermal fluctuations.

And I think I'll probably include all the gory details of how my simulations work since I'll be wanting to have direct references to the code.

```
double y0 = 10; // example of declaration and assignment statement
double v0 = 0;  // initial velocity
double t = 0;   // time
double dt = 0.01; // time step
double y = y0;  // solved all problems
```

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