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Chapter 2

Modeling athermal sub-isostatic fiber networks

Chapter 2 Modeling athermal sub-isostatic fiber networks

Abstract

Models of athermal sub-isostatic fiber networks are highly useful for studying the elasticity and mechanics of stiff biopolymer networks. The underlying network architecture is a key aspect that can affect the elastic properties of these systems, including rich linear and nonlinear elasticity. Existing computational approaches have focused on both lattice-based and off-lattice networks. It is not obvious a priori, if the mechanics of these architectures is fundamentally similar. If they are different, it is also not clear which of these represents a better model for biologically relevant networks. Here, we show that both approaches are equivalent for the same network connectivity, provided the networks are sub-isostatic with respect to central force interactions. Moreover, for a given connectivity, we find that lattice-based networks in both 2D and 3D exhibit identical nonlinear response. We show that with appropriate normalization factors that depend only on the underlying local network geometry, a quantitative comparison of results obtained from different network architectures is possible. In these models, we uncover three distinct elastic regimes. We present a description of the linear mechanics for both architectures in terms of a scaling function. We also show that the nonlinear regime is dominated by fiber bending and that the onset of stiffening is independent of the fiber material properties. In the large strain limit, the network mechanics is completely described by a network of floppy rope-like fibers.

2.1 Introduction

The study of fiber network mechanics is often aimed to elucidate the microscopic origins of viscoelasticity in biopolymer gels. Significant research progress has paved the way for models that highlight the importance and interplay of fiber semiflexibility, cross-link connectivity, and local geometry in the overall elasticity of networks. An important consideration in modeling network elasticity is the inherent instability of the underlying architecture with respect to pure central forces, i.e., tension along the fibers. Whether one looks at intracellular or extracellular biopolymer networks, the constituent fibers usually form either cross-linked or branched architectures [88, 103, 122], and therefore correspond to an average connectivity which is well below Maxwell’s criterion for the isostaticity or marginal stability of spring networks [123]. Below this threshold, the rigidity of a structure governed by central force interactions vanishes. These systems, however, can be stabilized by additional interactions [50] such as fiber bending [113], thermal fluctuations [124], internal stresses generated by molecular motors [125], boundary stresses [88, 107], or even strain [103, 126, 127]. These stabilizing fields give rise to distinct elastic regimes in sub-isostatic networks.
2.2 Network Architecture

The variety of computational models used to understand certain specific aspects of linear or nonlinear elasticity can either be based on off-lattice [70–72, 100, 101, 111, 128, 129] or lattice-based structures [113–117], which can also be combined with a mean-field approach [102, 113, 118, 119]. Indeed, much has been done with lattices to understand linear elasticity, in contrast to nonlinear elasticity often studied on random networks. The advantage of lattice-based models is the computational efficiency as well as the relative ease with which one can generate increasingly larger system sizes. In this chapter, we present an overview of the full range of elastic regimes in a lattice-based network model and compare the results from a random network. We begin with a detailed description of the disordered phantom network [114, 115] as well as the Mikado network [70, 71, 101]. These models allow the independent control of fiber rigidity and cross-link connectivity. We show a unified description of elasticity that allows for a quantitative comparison of simulation results across different network architectures as well as with experiments. Finally we conclude with implications when using these network models to understand the elasticity of athermal sub-isostatic fiber networks.

2.2 Network Architecture

2.2.1 Lattice-based phantom network

The disordered phantom lattice [114, 115] is generated by arranging fibers onto a \( d \)-dimensional space-filling regular lattice of size \( W^d \). The triangular \( (d = 2) \)/FCC \( (d = 3) \) lattice occupies a total volume \( V = v_0 W^d \), where \( v_0 \) is the volume of a unit cell. Periodic boundaries are imposed to reduce edge effects. Freely-hinged cross-links bind intersecting fiber segments permanently at the vertices, which are separated by a uniform spacing \( \ell_0 \). The full lattice has a fixed connectivity of \( z_{\text{max}} = 6 \) for \( d = 2 \) and \( z_{\text{max}} = 12 \) for \( d = 3 \). Typical biopolymer networks have a maximum of 4-fold connectivity, so we randomly detach binary cross-links at each vertex. In 2D, there is one phantom segment for every cross-linked pair of fiber segments at each vertex. In 3D, the six intersecting segments pair up into three cross-linked phantom segments. In either case, the phantom segments move freely through other segments with no interaction except at the cross-links. Up until this point all fibers span the system size and this leads to unphysical stretching contributions to the macroscopic elasticity when the network is deformed. We therefore cut at least one bond on every spanning fiber. Finally, we dilute the lattice by cutting random bonds with probability \( q = 1 - p \) with \( p \) being the probability of an existing bond to allow tuning of the average connectivity \( z \) continuously over the range \( 2 < z < 4 \). Any remaining dangling ends are removed as they have no contribution to the network mechanics. The resulting lattice-based network is sub-isostatic with average
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Figure 2.1: Lattice-based disordered phantom network in (a) 2D and in (b) 3D. The zoom-in shows the phantom segments in 2D. Three pairs of cross-linked strands are color-coded in 3D and are shown slightly displaced relative to each other in the zoom-in. (c) The 2D Mikado network. The fiber length $L$ and cross-linking distance $l_c$ are shown. In all three models, the average connectivity can be tuned within the range $z \in (2, 4)$.

connectivity $z \approx 4p$, average fiber length $L = \ell_0/q$ [113] and average contour distance $l_c$ between neighboring cross-links. We note that in 2D, because phantom segments skip at least one vertex, the average distance between adjacent cross-links is slightly larger than the lattice spacing. For very large systems, $l_c \lesssim \frac{3}{2} \ell_0$. By contrast, 3D lattice-based networks always have $l_c = \ell_0$.

2.2.2 Mikado network

We generate the off-lattice Mikado networks [70, 71] by sequential random deposition of stiff monodisperse rods of length $L \ll W$ onto a two-dimensional $W \times W$ box, which occupies a volume (i.e., area) $V = W^2$. Each rod’s center of mass and orientation relative to a fixed axis are drawn from a uniform distribution, making for an isotropic and homogeneous network. The box has periodic boundaries and if a rod intersects any boundary, the external segment crosses over to the opposite boundary. A freely-hinged cross-link is assigned at the point where a pair of rods intersect. Every time a rod is deposited, the cross-linking density $L/l_c$ is updated, where $l_c$ is the average contour
2.3 Network Elasticity

2.3.1 Fiber elasticity: the extensible worm-like chain model

Every fiber (i.e., rod) in the network has a finite resistance to both tension and bending. When the network is deformed, any point \( s \) on every fiber undergoes a displacement which induces a local change in length \( \frac{dl}{ds} \) and a local curvature \( \left| \frac{d\hat{t}}{ds} \right| \), where \( \hat{t} \) is a unit tangent along the fiber backbone (Figure 2.2(a)). The elastic energy stored in a fiber \( f \) in mechanical equilibrium with respect to the deformed network is given by a sum of the local stretching and bending contributions integrated along the fiber contour [70]:

\[
\mathcal{H}_f = \frac{1}{2} \int_f \left[ \mu \left( \frac{dl}{ds} \right)^2 + \kappa \left| \frac{d\hat{t}}{ds} \right|^2 \right] ds,
\]

where \( \mu \) and \( \kappa \) are the fiber stretching modulus and bending rigidity, respectively. Treating the fiber as a homogeneous cylindrical elastic rod of radius \( a \) and Young’s modulus \( E \), we know from classical beam theory [60] that \( \mu = \pi a^2 E \) and \( \kappa = \frac{1}{4}\pi a^4 E \). These parameters can be absorbed into a bending length scale \( l_b = \sqrt{\kappa/\mu} = a/2 \), which reflects the relative importance of the bending and stretching terms in Equation 2.1. By normalizing \( l_b \) with a natural geometric length scale such as the average spacing \( l_c \) between cross-links or branch points, we obtain a dimensionless fiber rigidity \( \tilde{\kappa} \equiv (l_b/l_c)^2 \), or

\[
\tilde{\kappa} = \frac{\kappa}{\mu l_c^2} = \frac{1}{4} \frac{a^2}{l_c^2}.
\]

Figure 2.2: (a) Schematic of a fiber showing the local change in length \( dl \) and the unit tangent \( \hat{t} \) at an infinitesimal segment \( ds \). Constraints can exist in the form of cross-links with other fibers or branch points (not shown for clarity). The dashed curve represents the fiber backbone. (b) Discretization of the same fiber showing midpoints to approximate bending modes.

distance between neighboring cross-links. The average number of cross-links per rod is given by \( L/l_c - 1 \). Deposition stops as soon as the desired cross-linking density is achieved, after which all dangling ends are removed.
2.3.2 Affine/non-affine network response

Network mechanics is determined not only by the rigidity $\tilde{\kappa}$ of the constituent fibers but is also governed by the network’s affine/non-affine response to the applied deformation. This is controlled by the average network connectivity $z$ [113] or equivalently the average cross-linking density [70] as quantified by the aspect ratio $L/l_c$ of geometric length scales in the network. A higher $z$ imposes more constraints on the cross-links and thereby on the fiber, as does a higher $L/l_c$. As such, the fibers have less freedom to explore deformation modes beyond those dictated by the affine, i.e., uniform displacement field (see Figure 2.3). By contrast, a lower $z$ and lower $L/l_c$ means that the fiber is less constrained, allowing the exploration of non-uniform displacements. Such non-affine modes lead to a softer network response relative to one wherein the displacement field.
2.3 Network Elasticity

is affine. Network rigidity is therefore determined by the coupling of the parameters $\tilde{\kappa}$ and $z \leftrightarrow L/l_c$. On one hand, one could construct a soft network from rigid rods but with a low density of cross-links. On the other hand, a rigid network can be obtained from highly constrained floppy fibers.

2.3.3 Network elastic energy density

The total elastic energy of the network $\mathcal{H} = \sum_f \mathcal{H}_f$ is obtained from the sum of all the fiber elastic energies $\mathcal{H}_f$ expressed in continuous form by Equation 2.1. In our network of straight fibers with discrete segments, a midpoint node is introduced at every segment (Figure 2.2(b)) to capture the lowest bending mode over the smallest length scale $\ell_0$ ($l_c$ in Mikado). The set of spatial coordinates $\{ r_j \} \in \mathbb{R}^d$ of all nodes $j$ (i.e., cross-links, phantom nodes and midpoints) thus constitutes the internal degrees of freedom of the network. Under any finite macroscopic deformation $\varepsilon$, the nodes undergo a displacement $\{ r_j \} \rightarrow \{ r'_j \}$ which induces the dimensionless local deformations $\lambda_j = \delta \ell_j / \ell_j$ and $\theta_j = | \hat{t}_{j,j+1} - \hat{t}_{j-1,j} |$ on the fiber segments. Here, $\delta \ell_j = \ell'_j - \ell_j$ is the length change of a fiber segment with rest length $\ell_j = | r_{j+1} - r_j |$, deformed length $\ell'_j = | r'_{j+1} - r'_j |$, and $\hat{t}_{ij}$ is a unit vector tangent to segment $(ij)$. The fiber in the deformed conformation stores the elastic energy expressed as a discretized form of Equation 2.1:

$$\mathcal{H}_f = \frac{1}{2} \sum_{j \in f} \left( \mu \ell_j \lambda_j^2 + \frac{\kappa}{l_j} \theta_j^2 \right), \quad (2.3)$$

where $l_j = \frac{1}{2} ( \ell_{j-1} + \ell_j )$. By taking $l_j \approx \ell_j \approx l_c$, we can rewrite this equation that shows an explicit dependence on $\varepsilon$ and $\tilde{\kappa}$, and then obtain the network elastic energy as a sum over all $N$ fibers:

$$\mathcal{H} = \mu l_c \sum_{f=1}^N \tilde{\mathcal{H}}_f (\varepsilon, \tilde{\kappa}, L/l_c), \quad (2.4)$$

where

$$\tilde{\mathcal{H}}_f = \frac{1}{2} \sum_{j \in f} (\lambda_j^2 + \tilde{\kappa} \theta_j^2)$$

is a dimensionless fiber elastic energy. Here, the dependence on the macroscopic deformation $\varepsilon$ is appears microscopically as the $(\lambda_j, \theta_j)$ of the individual fiber segments. As discussed above, the dependence on $L/l_c$ couples to $\tilde{\kappa}$, which will be explored further in this chapter.

The elasticity of a fibrous network is characterized by its internal stresses and stiffness or resistance to deformation, all derived from the total elastic energy density $U = \frac{1}{V} \mathcal{H}$. In the case of simple shear deformation $\varepsilon = \gamma$, we obtain the shear stress $\sigma = \frac{\partial U}{\partial \gamma}$ and stiffness as measured by the tangent shear modulus $K = \frac{\partial^2 U}{\partial \gamma^2}$. By taking the sum of the
energies of all fibers, we obtain the total elastic energy density $U = \mu \rho \langle \tilde{\mathcal{H}}_f \rangle$, where the line density $\rho = NL/V$ is the total fiber length per volume and $\langle \cdot \rangle$ is an average\(^1\) over the total number of fibers. Defining the dimensionless energy density $\tilde{U} = \langle \tilde{\mathcal{H}}_f \rangle$, we have

$$U = \mu \rho \tilde{U} (\gamma, \tilde{\kappa}, L/l_c).$$  \hspace{1cm} (2.5)

Successive differentiations with respect to $\gamma$ lead to

$$\sigma = \mu \rho \frac{\partial \tilde{U}}{\partial \gamma} = \mu \rho \tilde{\sigma} (\gamma, \tilde{\kappa}, L/l_c)$$  \hspace{1cm} (2.6)

and

$$K = \mu \rho \frac{\partial \tilde{\sigma}}{\partial \gamma} = \mu \rho \tilde{K} (\gamma, \tilde{\kappa}, L/l_c).$$  \hspace{1cm} (2.7)

### 2.3.4 Rheology simulation

The elastic response of the network is simulated by applying a volume-preserving simple shear strain $\gamma$ (Figure 2.4a) which is increased in steps over a range that covers all elastic regimes, typically from 0.1\% to 1000\%. At each $\delta \gamma$ strain step, the network reaches mechanical equilibrium after minimizing the total elastic energy density given by Equation 2.5, thereby relaxing the internal degrees of freedom using a conjugate gradient minimization routine [130]. Lees-Edwards boundary conditions [131] ensure that the lengths of segments crossing the system boundaries are calculated correctly. From the minimized total elastic energy density, $\sigma$ and $K$ are evaluated using finite

---

\(^1\)The average is actually to be taken over all $N_s$ fiber segments. However, since $\rho = N_s l_c/V = N L/V$, then averaging over segments or fibers are equivalent.
differences. We also determine the normal stress at every $\gamma$:

$$\tau = \mu \rho \tilde{\tau} (\gamma, \tilde{\kappa}, L/l_c),$$  

(2.8)

where $\tilde{\tau} = \left. \frac{\partial \tilde{U}}{\partial \epsilon} \right|_{\gamma}$ and $\epsilon$ is a small uniform deformation applied normal to the shear boundaries. Measuring these quantities allows us to characterize the elastic regimes of the network.

It is possible that a significant fraction of rather straight fibers are initially oriented along or against the strain direction. This introduces an inherent asymmetry of the network with respect to shear deformation [71], which becomes problematic when probing network elasticity especially in the low strain limit. One way to avoid this artifact is to generate very large networks and calculate an ensemble averaged energy vs strain curve. We describe a more efficient alternative (Figure 2.4b) as follows: around zero strain, $H(\gamma)$ is quadratic in $\gamma$ and can be described by a shifted parabola as a result of the asymmetry. We take a full strain sweep of the network $-\gamma_{\text{max}} \rightarrow +\gamma_{\text{max}}$ to obtain $H(\gamma)$. We then take the reverse strain sweep on the mirror image of the same network to obtain $H(\gamma)$. The average of both sweeps should yield a symmetric $H_0(\gamma)$ curve.

### 2.3.5 Quantitative comparison of measurements

With appropriate normalization of the stresses and moduli, it is possible to quantitatively compare these quantities from simulations of different network architectures as well as those from experiments by accounting for the line density $\rho$ and the stretch modulus $\mu$ of the fibers. Any measured quantity $X$ (e.g. stress or modulus) is normalized in dimensionless form as

$$\tilde{X} = \frac{X}{\mu \rho}.$$  

(2.9)

The line density $\rho$ is specific to the chosen network architecture, i.e., the local network geometry. In lattice-based networks, $\rho_d = \tilde{\rho}_d / l_c^{d-1}$ with $\tilde{\rho}_d = \frac{6 \rho}{\sqrt{3}}$ and $\rho_3D = \frac{12 \rho}{\sqrt{2}}$ (Appendix). For Mikado networks, because of the polydispersity of $l_c$, it is more convenient to express the line density in terms of fiber length $L$ such that $\rho_M = \tilde{\rho}_M / L$, where $\tilde{\rho}_M = n_f L^2$ and $n_f$ is the number of rods per unit area [132]. As for experiments, one could use the relation between $\rho$ and volume fraction $\varphi$, which can be obtained from the concentration of fiber material (i.e., protein concentration). In particular, for an arbitrary 3D network structure of approximately cylindrical fibers with cross-section $\pi a^2$, it is straightforward to show that $\rho = \frac{\varphi}{\pi a^2}$ (Appendix). Thus, the normalization factor for experimental measurements should be $\mu \rho = \pi a^2 E \frac{\varphi}{\pi a^2} = \varphi E$. 

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2.4 Results

2.4.1 Distinct elasticity regimes

Distinct elastic regimes can be identified from the family of stiffening curves shown in Figure 2.5a as a function of strain, fiber rigidity, and connectivity or cross-linking density. We observe: (I) a linear regime for low $\gamma$ where $\lim_{\gamma \to 0} K(\gamma, \tilde{\kappa}, L/l_c) = G_0(\tilde{\kappa}, L/l_c)$; (II) a nonlinear stiffening regime showing a dramatic increase of $K$ over the narrow strain range $\gamma_0 \lesssim \gamma \lesssim \gamma_c$; and (III) an affine stretch regime for $\gamma \geq \gamma_c$ where all curves collapse independently of $\kappa$. In regime I, the linear modulus $G_0$ reveals two regimes shown in the inset: (i) a bend-dominated regime with $G_0 \sim \tilde{\kappa}$ for low $\tilde{\kappa}$, which crosses over into (ii) a stretch-dominated regime at high $\tilde{\kappa}$, where bending is suppressed and the response is primarily due to stretching, i.e., $G_0 \sim \mu$. The crossover appears to be controlled by the cross-linking density and fiber rigidity. In regime II, we observe how connectivity controls the onset and extent of the nonlinear stiffening regime. Here, the threshold strain $\gamma_0$ marks the onset of nonlinearity, which roughly corresponds to the strain at which $2 < \frac{K}{G_0} < 3$ [88]. This nonlinear regime terminates at the critical strain $\gamma_c$, which is defined as the strain at which a fully floppy (i.e., $\kappa = 0$) network develops for the first time a nonzero rigidity. As the connectivity and therefore the density of cross-links increases, both $\gamma_0$ and $\gamma_c$ shift to lower values. Moreover, the width of regime II also decreases as the network shows more of its affine response as $z$ is made to approach the Maxwell isostatic limit. In regime III, the stiffness grows independently of $\tilde{\kappa}$ as stretching modes become dominant. This is evident in the collapse of the stiffening curves from finite $\kappa$ networks as they emulate the behavior of the $\kappa = 0$ network at a particular connectivity.

Remarkably, these characteristic features of stiffening are insensitive to the local geometry, i.e., Mikado vs lattice-based and even dimensionality when comparing networks with the same average connectivity. The insensitivity with respect to dimensionality only holds, however, below the respective isostatic thresholds, which are different for 2D and 3D. We show in Figure 2.5b that the different local geometries of 2D Mikado and 2D lattice-based networks of the same $z$ show quantitative agreement, once the difference in fiber density $\rho$ has been taken into account by the appropriate normalization procedure of Equation 2.9. A similar insensitivity with respect to dimensionality is shown in Figure 2.5c. Moreover, for the same connectivity, even the strain thresholds $\gamma_0$ and $\gamma_c$ coincide for 2D Mikado and 2D lattice-based networks, as well as for 2D and 3D lattice-based networks [88, 103]. Apart from a simple geometric prefactor to the stress or modulus, it is therefore equally suitable to study lattice-based networks in 2D or 3D.
2.4 Results

Figure 2.5: (a) Stiffness $K$ of a 2D lattice-based network as a function of simple shear strain $\gamma$ on a network with $L/l_c = 3$, $z = 3.2$ (black symbols) and with $L/l_c = 9$, $z = 3.8$ (gray symbols). In either network, the stiffness takes a constant value $K = G_0$ in the limit of $\gamma \to 0$. From the threshold strain $\gamma_0$ at the onset of nonlinear stiffening (enlarged green symbols), $K$ increases dramatically until $\gamma = \gamma_c$, set by the $\tilde{\kappa} = 0$ limit (dashed curves). For $\gamma > \gamma_c$, all curves collapse as $K$ becomes independent of $\tilde{\kappa}$ and dominated by fiber stretching. Both $\gamma_0$ and $\gamma_c$ shift to lower strains with increasing $z$.

Inset: The linear modulus $G_0$ as a function of fiber rigidity $\tilde{\kappa}$ also shows two elastic regimes: $G_0 \sim \tilde{\kappa}$ (dashed line of unit slope) and $G_0 \sim \tilde{\kappa}_0 \sim \mu$ (solid horizontal line). Symbol colors represent the same $z$ values indicated in the main plot. (b) The stiffening curves of 2D Mikado (open symbols, $L/l_c = 11$, $z = 3.6$) and 2D lattice-based (filled symbols, $L/l_0 = 6$, $z = 3.6$) networks, normalized by their respective $\mu \rho$, show the same stiffening features with coinciding $\gamma_0$ and $\gamma_c$. (c) Likewise, normalized stiffening curves from 3D lattice-based (red symbols) and 2D lattice-based (black symbols) networks, both with $z = 3.2$ show the same behavior.
Figure 2.6: For networks with high connectivity and cross-linking density such as in the 2D lattice with $z = 3.8$ and $L/l_c = 9$ (filled symbols are the gray data set from Figure 2.5a, shown here in a larger vertical scale), an apparent “softening” of the network is observed as $K$ dips slightly relative to $G_0$. This artifact is not present for lower $z$ and $L/l_c$ (e.g. $L/l_c = 3$ in Figure 2.5a) or when undulations are introduced to the fibers (open symbols) by applying a small uniform macroscopic compressive strain ($\varepsilon < 1\%$) normal to the network boundaries.

2.4.2 Fiber buckling

Previous computational studies on both lattice and off-lattice based networks have reported that the transition from linear to nonlinear regime under strain is marked by an initial softening of the modulus [71, 72]. Softening appears as a dip in $K$ relative to $G_0$ which occurs due to buckling of the filaments under compression. However, to our knowledge, experimental demonstration of softening has remained elusive. We suggest that the buckling-induced softening is an artifact of simulations using networks of initially long and straight fibers. This is observed in Figure 2.5b as well as in Figure 2.6. When we introduce fiber undulations by initially compressing the network, no such softening is observed. In this pre-compressed state, the undulating fibers undergo increased bending but do not buckle. We expect that in biopolymer networks, the filaments naturally exhibit undulations either from defects or prestress, and hence would not demonstrate buckling-induced softening under strain. We also find in our simulations that softening only occurs in networks with high $L/l_c$, as the whole length of the fiber will not be able to release the affine displacements due to stretching or compression by exploring the non-affine bending modes but instead it buckles locally. This is in contrast to networks with a much lower $L/l_c$, where softening is not observed Figure 2.5a (black symbols) and Figure 2.5c.
2.4 Results

2.4.3 Linear regime

The linear regime (I) is characterized by a constant modulus \( G_0 \) for \( \gamma \ll \gamma_0 \). As mentioned above and shown in the inset of Fig. 2.5a, the linear elasticity exhibits two distinct regimes: a bend-dominated one in which \( G_0 \sim \tilde{\kappa} \) and one in which \( G_0 \sim \mu \) is independent of \( \tilde{\kappa} \) and is stretch-dominated. The dependence of the crossover on \( L/l_c \) arises due to the fact that the displacements of contiguous segments on a semiflexible fiber are not independent of each other [100]. This means that segments near the endpoints deform less affinely since they are relatively less constrained than the ones near the middle. The non-affinity length scale \( \lambda_{NA} \) associated with the range of non-affine regions near the endpoints is determined by \( l_c, l_b \) as follows [70, 100, 115]:

\[
\lambda_{NA} = l_c \left( \frac{l_c}{l_b} \right)^\zeta .
\] (2.10)

The ratio \( L/\lambda_{NA} \) then determines the crossover as

\[
\frac{G_0}{G_{AFF}} \sim \left( \frac{L}{\lambda_{NA}} \right)^{2/\zeta} \sim \tilde{\kappa} \left( \frac{L}{l_c} \right)^{2/\zeta} ,
\] (2.11)

where \( G_{AFF} = A\mu\rho \) is the linear modulus in the affine limit and the prefactor \( A \) depends on the local network geometry. The above scaling also reflects the coupling of \( \tilde{\kappa} \) to \( L/l_c \) in the modulus. For \( L \lesssim \lambda_{NA} \), the modulus is governed by the softer bending modes in the network. On the other hand, for \( L > \lambda_{NA} \), the modulus is governed by stretching modes. The exponent \( \zeta \) depends on the local network geometry. In the off-lattice 2D Mikado, \( \zeta \simeq 2/5 \) [70, 100], whereas in 3D FCC lattice-based networks \( \zeta = 1 \) [115]. Here, we show that 2D triangular lattice-based networks also have \( \zeta = 1 \), as in the 3D FCC.

In Figure 2.7a, the data obtained for a broad range of \( L/l_c \) collapse on the master curve (Equation 2.11) with slope \( 2/\zeta = 2 \). A significant systematic deviation is seen for data corresponding to relatively small values of \( L/l_c \). This has been observed in a previous study on 3D FCC lattice-based networks where the deviation is attributed to a possible different scaling in the vicinity of the rigidity percolation threshold [115]. However, on replacing \( L \) by \( L - L_r \), where \( L_r \) is the minimum fiber length at this threshold, we obtain an excellent collapse for all values of \( L/l_c \) with the same \( \zeta = 1 \) (Figure 2.7b). It follows that in the linear regime \( G_0/G_{AFF} \sim \tilde{\kappa} \left( \frac{L - L_r}{l_c} \right)^2 \), which we show in the inset of Figure 2.7b. Indeed, the scaling \( G_0/G_{AFF} \sim \kappa L^2 \) is known for 3D FCC lattice-based networks for \( L \gg L_r \) [115]. Interestingly, such scaling behavior with a similar correction to \( L \) has been observed in experiments on hydrogels [133].
Figure 2.7: Collapse of linear modulus with non-affinity length scale (a) without and (b) with $L_r$ correction. In this 2D lattice-based simulation, we find $L_r \approx 2.94$. Red symbols represent networks in the vicinity of the rigidity percolation regime. The inset of (b) shows the collapse of the linear modulus according to Equation 2.11.
2.4 Results

2.4.4 Bending correlation length

The exponent $\zeta$ characterizing the non-affinity length scale can be calculated using mean-field arguments to be $\frac{2}{5}$ [70]. With this exponent, the modulus of off-lattice Mikado networks can be quantitatively captured by Eq. (2.11). The mean-field implicitly assumes that the non-affinity length scale is larger than the bending correlation length

$$\lambda_b = l_c \left( \frac{l_b}{l_c} \right)^\zeta.$$  \hspace{1cm} (2.12)

Moreover, both $\lambda_{NA}$ and $\lambda_b$ are assumed to be larger than $l_c$. It has been previously pointed out that in the limit of very flexible rods or for low concentrations, Eq. (2.12) would predict $\lambda_b < l_c$, which is an unphysical result [70]. Thus when $l_b/l_c$ becomes very small, by fixing $\lambda_b = l_c$ one obtains $\zeta = 1$ and $\lambda_{NA} = l_c^2/l_b$. Since the non-affinity length scale obtained under the assumption that $\lambda_b = l_c$ is the same as found in 2D and 3D lattice-based networks, it seems that indeed the bending correlation length is very close to $l_c$. One does not expect this to hold for $L$ approaching $L_r$ where highly non-affine deformations would include bending that occurs on length scales much larger than $l_c$. However, as we show above by making the empirical correction $L - L_r$, the scaling of Equation 2.11 is extended all the way up to the threshold length $L_r$ of rigidity percolation.

2.4.5 Prefactors to the modulus

As discussed above, the primary difference between lattice-based and off-lattice network structures is in their bending correlation length. With the appropriate exponent $\zeta$, the linear modulus from both off-lattice and lattice-based networks can be quantitatively captured by Equation 2.11, providing a unified description of the linear mechanics of fibrous networks independent of the detailed microstructure. In general, we can write the linear modulus as

$$G_0 (\tilde{\kappa}, L/l_c) = A \mu \rho \tilde{\kappa} \left( \frac{L - L_r}{l_c} \right)^{2/\zeta}$$

$$= A \mu \rho \tilde{\kappa} \left( \frac{L}{l_c} \right)^{2/\zeta}, \quad L \gg L_r.$$  \hspace{1cm} (2.13)

A summary of the parameters $\zeta$ and $A$ are shown in Table 2.1 for the three network geometries used in our simulations.

The prefactors in Table 2.1 can be used to write an expression for the modulus as measured from experiments and use it to quantitatively compare with 3D lattice-based
Table 2.1: Exponent $\zeta$ and prefactor $A$ for Equation 2.13 extracted from the collapse using Equation 2.11.

<table>
<thead>
<tr>
<th>Network</th>
<th>$\zeta$</th>
<th>$A$</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>2D triangular lattice-based</td>
<td>1.0</td>
<td>$4.2 \pm 0.2$</td>
<td>[107]</td>
</tr>
<tr>
<td>3D FCC lattice-based</td>
<td>1.0</td>
<td>$1.5 \pm 0.3$</td>
<td>[115]</td>
</tr>
<tr>
<td>2D Mikado</td>
<td>$\frac{2}{5}$</td>
<td>$(5 \pm 1) \times 10^{-4}$</td>
<td>[70]</td>
</tr>
</tbody>
</table>

networks. If we substitute the corresponding expressions for $\rho_{3D}$ and $\tilde{\kappa} = \frac{\varphi}{4\pi\rho}$ (see Appendix), we obtain

$$G_0 = \frac{A}{z} \left( \frac{L}{L_c} \right)^2 E\varphi^2,$$

where $A = \frac{4\sqrt{2}}{12\pi}$. Indeed, it has been shown that the fiber rigidity $\tilde{\kappa}$ is proportional to the protein concentration in reconstituted collagen networks and that $G_0$ scales as the square of the protein concentration [88, 103].

### 2.4.6 Nonlinear stiffening

The shear and negative normal stress from 2D networks are plotted in Figure 2.8 as a function of strain and fiber rigidity. For comparison, results from a Mikado network with a higher local connectivity are also shown. In the linear regime, $\sigma \sim \gamma$ while $-\tau \sim \gamma^2$, as expected from symmetry arguments [71, 78, 104, 105]. The negative sign in the normal stress is characteristic of biopolymer gels and has been observed in experiments [78, 106], where it may be attributed either to the asymmetric force-extension curve of the constituent fibers [49] or to non-affine deformations in athermal networks leading to an effective network-level asymmetry in the response [71, 105]. As $\gamma$ increases, the shear and normal stress become increasingly comparable in magnitude.

We can now unambiguously define $\gamma_0$ as the strain at which $|\sigma| = |\tau|$ [78]. From this onset of stiffening, both stresses begin to increase nonlinearly with strain as $\gamma \rightarrow \gamma_c$. For $\gamma > \gamma_c$, both stress curves converge to their respective $\kappa = 0$ limits, in a similar manner observed for the $K$ vs $\gamma$ curves in Figure 2.5. In the very large strain limit when most of the fibers have aligned in the strain direction, the shear response is again linear in strain, while the normal response saturates to a constant.

Interesting features in each nonlinear regime are further revealed in the $K$ vs $\sigma$ curves (Figure 2.9). Nonlinear regime II is initiated by the points $(\sigma_0, K_0)$ at the onset strain $\gamma_0$, and proceeds as a $K \sim \sigma^\alpha$ stiffening, with $\alpha$ increasing as $\tilde{\kappa}$ is decreased (lower inset of Figure 2.9). In the stretch-dominated regime III all curves converge to $K \sim \sigma^{1/2}$ independently of $\tilde{\kappa}$ [71, 114]. These results are consistent with prior theoretical work showing an evolution of exponents from $\alpha \approx 1/2$ through $\alpha \approx 1$ and higher values with decreasing $\tilde{\kappa}$ [114]. That such evolution of $\alpha$ with concentration is also
Figure 2.8: (a) Shear stress $\sigma$ and (b) negative normal stress $-\tau$ as a function of $\gamma$ and $\tilde{\kappa}$ in a 2D lattice-based network with $L/l_c = 3$, $z = 3.2$. Red data is from the Mikado simulation with $L/l_c = 11$, $z = 3.6$. For low strains, the linear regime shows $\sigma \sim \gamma$ and $-\tau \sim \gamma^2$. The stresses at the onset strain $\gamma_0$ of stiffening are indicated by green symbols interpolated by the thick dashed schematic curve. Beyond the onset of stiffening, both stresses are still dependent on $\tilde{\kappa}$ and increase nonlinearly with strain until reaching the critical strain $\gamma_c$ marked by the arrow. Beyond $\gamma_c$, all curves collapse onto the $\tilde{\kappa} = 0$ limit as they become independent of $\tilde{\kappa}$. For very large strains, $\sigma \sim \gamma$ while $-\tau \sim \text{const}$. Inset: At the onset of nonlinear stiffening, $-\tau_0 \approx \sigma_0$, and both scale linearly with $\tilde{\kappa}$ as in the linear regime.
Figure 2.9: Stiffness $K$ as a function of shear stress $\sigma$ for different $\tilde{\kappa}$ in a 2D lattice-based network with $L/l_c = 3$, $z = 3.2$. The lines connecting the data points only serve as visual guides. The green points correspond to $(\sigma_0, K_0)$ at the threshold strain $\gamma_0$ for the onset of stiffening and are replotted in the inset (upper panel) for the full range of $\tilde{\kappa}$. The red dashed line of unit slope interpolates through the green points showing $\sigma_0/K_0 \sim \text{const}$ for $\tilde{\kappa} \lesssim 10^{-2}$. In regime II, the networks undergo $K \sim \sigma^\alpha$ stiffening (black dashed lines), after which the curves collapse onto the $\tilde{\kappa} = 0$ limit in regime III showing $K \sim \sigma^{1/2}$. The lower panel of the inset shows the evolution of the stiffening exponent $\alpha$ with fiber rigidity in regime II.

consistent with recent experiments on collagen networks [88]. In contrast to what has been proposed in [134], our results show there is no unique exponent $\alpha = 3/2$ in the initial stiffening regime. The threshold strain $\gamma_0$ at the onset of stiffening is characterized by the points of stiffness $K_0$ scaling linearly with shear stress $\sigma_0$ (Figure 2.9, red dashed line). This feature can be understood as follows. For low stresses, the network energy density is dominated by soft bending modes since $G_0 \sim \tilde{\kappa}$ (Figure 2.5a, inset) [135, 136]. Moreover, these networks stiffen at the onset stress $\sigma_0 \propto \tilde{\kappa}$ (Figure 2.8, inset). From these observations, together with the fact that $K$ and $\sigma$ have the same units, it follows that $\sigma_0$ and $G_0 \approx K_0$ should depend in the same way on network parameters. Thus, the points $(\sigma_0, K_0)$ should exhibit a linear relationship, as seen in networks for $\tilde{\kappa} \lesssim 10^{-2}$, which means that the onset strain $\gamma_0 \propto \sigma_0/K_0$ must be independent of $\tilde{\kappa}$ (upper inset of Figure 2.9).
2.4 Results

2.4.7 Stretch-dominated elasticity

Stiffening in regime III is governed predominantly by fiber stretching. Since bending interactions are no longer important, it is expected that the network mechanics closely resembles that of a floppy “rope” network for which $\kappa \equiv 0$. Such rope networks are unstable to small deformations, i.e., the linear elastic modulus vanishes. However, these networks become rigid for sufficiently large deformations [102]. The critical strain $\gamma_c$ can be identified as the strain marking the onset of rigidity in a rope network [103] and its nonlinear mechanics can be used to understand the $K \sim \sigma^{1/2}$ scaling. One way this has been explained in the past is that in a marginally stable isostatic network resists shear deformations nonlinearly such that $\sigma \sim \gamma^2$. The modulus near the critical strain therefore scales as $K \sim |\gamma| \sim \sigma^{1/2}$ [114, 126].

An intuitive picture is based on the simple idea of a gradual recruitment of stretched fiber strands. As soon as the deformation reaches $\gamma_c$, stretched strands are engaged at a rate $\frac{dn_s}{d\gamma} \propto r(1 - n_s)$, where $n_s$ is the number density of stretched fiber strands. In this empirical relation, $(1 - n_s)$ ensures that the number density of stretched fiber strands has a finite upper bound. As a first approximation, the constant $r$ assumes that fiber strands are recruited independently and uniformly into stretching. Using the boundary condition $n_s(\gamma \leq \gamma_c) = 0$, we obtain an exponential saturation

$$n_s(\gamma - \gamma_c) \sim 1 - e^{-r(\gamma - \gamma_c)} , \quad \gamma > \gamma_c.$$

From this we can write the modulus as $K \sim \mu \rho n_s(\gamma - \gamma_c)$. Similarly, the stress can be written as $\sigma \sim \mu \rho \int_{\gamma_c}^{\gamma} n_s d\gamma \sim (\gamma - \gamma_c) - \frac{1}{r} n_s(\gamma - \gamma_c)$. For $|\Delta \gamma| = |\gamma - \gamma_c| > 0$, a linear expansion of Equation 2.15 in the neighborhood of $\gamma_c$ leads to $K \sim |\Delta \gamma|$ and $\sigma \sim |\Delta \gamma|^2$. It immediately follows that $K \sim \sigma^{1/2}$. Indeed, an apparent scaling $K \sim |\Delta \gamma|^{f/2}$ and $\sigma \sim |\Delta \gamma|^{(1+f)/2}$ can be seen from simulations of networks approaching the rope limit (Figure 2.10).

We note that although the $K \sim \sigma^{1/2}$ has received much attention in previous studies [114, 126], it is not a universal feature. In fact, for the broad range of connectivities of sub-isostatic networks, including disordered honeycomb networks with $z \gtrsim 2$, the exponent $f$ has been shown to depend on network connectivity, giving rise to a whole range of exponents in the scaling $K \sim |\Delta \gamma|^{f}$ [103] leading to $K \sim \sigma^{f/(1+f)}$. The empirical relation described above using the naïve picture of stretched fiber recruitment has therefore limited validity. It does however provide an intuitive picture of the stretch-dominated stiffening regime provided that one is aware of this caveat.
**Chapter 2 Modeling athermal sub-isostatic fiber networks**

Figure 2.10: Stiffening curves of a 2D lattice-based network with \( z = 3.2 \) and different fiber rigidities, plotted vs \( |\gamma - \gamma_c| \) for \( \gamma > \gamma_c \) approach the rope network limit \( \tilde{\kappa} = 0 \), showing an apparent scaling \( K \sim |\gamma - \gamma_c|^{f \ll 1} \). The red dashed curve is the fit \( K = \mu \rho n_s (\gamma - \gamma_c) \) using Equation 2.15. The black dashed line of unit slope serves as visual guide. The inset is a plot of stress vs \( |\gamma - \gamma_c| \), with the fit \( \sigma = (\gamma - \gamma_c) - \frac{1}{2} n_s (\gamma - \gamma_c) \) shown by the red dashed curve. The black dashed line of slope 2 is a visual guide.

### 2.5 Discussion and Implications

In this chapter, we have presented the modeling framework as well as the general features one observes in the elastic behavior of sub-isostatic athermal fiber networks. Athermal fiber networks can be used to model the mechanics of biologically-relevant networks such as collagen. It is a priori unclear whether one needs to take into account the detailed microstructure of a biopolymer network into a computational model. Most models are based on lattice-based or off-lattice network architectures. The primary advantage of a lattice-based approach is computational efficiency. By contrast, off-lattice networks, though computationally intensive, would appear to be more realistic, in the sense that the network structure has a built-in spatial disorder which is a key feature of biopolymer networks. Here we show that despite the structural differences, these two approaches can be unified and are equally suited to describe most aspects of the mechanical response of athermal fiber networks. The remarkable insensitivity of the generic features in the stiffening curves with respect to network architecture as well as dimensionality for the same connectivity implies the existence of a single master curve that depends only on \( (\gamma, \tilde{\kappa}, z \leftrightarrow L/l_c) \), which captures the full range of elastic regimes in sub-isostatic athermal fiber networks. Thus, for a given connectivity, one can effectively capture the general
features of elasticity in athermal fiber networks using either 2D or 3D lattice-based models.

The linear elasticity of these network is described by the modulus characterized in terms of a non-affinity length scale $\lambda_{NA}$, which depends on the underlying network structure. Our approach is based on networks of cross-linked fibers allowing for an unambiguous and intuitive definition of the non-affinity length scale $\lambda_{NA}$. The concept of non-affinity length scale can also be extended to branched networks by considering $t_c$ as the average branching distance. The scaling relation in Equation 2.11 with the geometry-dependent exponent $\zeta$ captures the crossover behavior in the linear regime. The non-affinity length scale can be derived for a given network using mean-field arguments. However, we show that with an empirical fiber length correction $L - L_r$, the scaling relation can even capture the linear mechanics of networks close to the rigidity percolation where mean-field behavior is no longer expected.

The onset of the nonlinearity is marked by a threshold strain at which the magnitude of the normal stress becomes comparable to the shear stress. Experimental determination of $\gamma_0$ is typically based on an arbitrary criterion such as the strain at which the differential modulus is twice or three times the linear modulus [88]. The normal stress becoming comparable in magnitude to the shear stress could provide an unambiguous definition of the onset of nonlinear stiffening. However, the advantage of defining $\gamma_0$ based on stress could be nullified in experiments due to ambiguity in the measurement of normal stresses. Any form of prestress in the network would offset the self-generated normal stresses induced by shear strain. From the threshold strain $\gamma_0$ until a critical strain $\gamma_c$, the stresses and modulus increase nonlinearly. This nonlinear stiffening in regime II shows $K \sim \sigma^\alpha$ scaling and is consistent with prior studies where the exponent evolves with the fiber rigidity. The modulus scales linearly with the shear stress at the onset of nonlinearity, which from our physical picture is an indication that this regime is still dominated by fiber bending. This also suggests that $\gamma_0$ is independent the material properties of the network. In the final nonlinear regime, stretching interactions become the dominant contributor to the network mechanics. Indeed, our simulations show that in regime III, the stresses and moduli collapse onto the curve associated with the $\tilde{\kappa} = 0$ case independent of the fiber bending rigidity. This suggests that above the critical strain, the network mechanics should be completely described by a pure $\tilde{\kappa} = 0$ “rope” network.
2.6 Appendix: Line Density Calculation of Lattice-Based Networks

On any lattice with uniform bond lengths \( l_c \), the line density can be calculated as the total length of bonds per unit volume, i.e., \( \rho = n_b l_c / v_0 \) where \( n_b \) is the number of bonds in a unit cell of volume \( v_0 \). In a two-dimensional diluted triangular lattice, there are three lines intersecting a vertex. Enclosing a vertex by a circle of radius \( l_c / 2 \), the total length of the enclosed bonds is \( 3p l_c \). Dividing by the area of the circle and multiplying by the circle packing fraction of a triangular lattice which is \( \pi / (2\sqrt{3}) \), we have:

\[
\rho_{2D} = \frac{3p l_c}{\pi \left( \frac{l_c}{2} \right)^2} \left( \frac{\pi}{2\sqrt{3}} \right) = \frac{\tilde{\rho}_{2D}}{l_c} ; \quad \tilde{\rho}_{2D} = \frac{6p}{\sqrt{3}}.
\]

In the case of a 3D diluted FCC lattice, we can imagine six lines intersect each vertex. Enclosing a vertex by a sphere of radius \( l_c / 2 \), the total length of the enclosed bonds is \( 6p l_c \). Dividing by the volume of the sphere and multiplying by the packing fraction of the FCC lattice which is \( \pi / (\sqrt{18}) \), we have:

\[
\rho_{3D} = \frac{6p l_c}{\frac{4}{3} \pi \left( \frac{l_c}{2} \right)^3} \left( \frac{\pi}{\sqrt{18}} \right) = \frac{\tilde{\rho}_{3D}}{l_c^2} ; \quad \tilde{\rho}_{3D} = \frac{12p}{\sqrt{2}}.
\]

The bending rigidity can be mapped to concentration via the volume fraction. In an arbitrary 3D network structure of \( N \) homogeneous cylindrical rods with length \( L \), cross-section \( \pi a^2 \), and Young’s modulus \( E \), the volume fraction is proportional to the line density:

\[
\varphi = \frac{\pi a^2 N L}{V} = \pi a^2 \rho.
\]

Combining this with the definition of the reduced fiber rigidity \( \tilde{\kappa} = \frac{\pi a^4 E}{\rho l_c^2} = \frac{1}{4} \frac{a^2}{l_c^2} \tilde{\kappa} \), we obtain for example in the case of the 3D FCC \( \varphi = 4\pi \tilde{\rho} \tilde{\kappa} \) or:

\[
\varphi = \pi a^2 \rho_{3D} = \frac{12p a^2}{\sqrt{2} l_c^2} = \frac{48p}{\sqrt{2}} \tilde{\kappa}.
\]