Effective medium theory of semiflexible filamentous networks

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(Dated: August 9, 2006)

We develop an effective medium approach to the mechanics of disordered, semiflexible polymer networks and study their response to both spatially uniform and nonuniform strain. We identify distinct elastic regimes in which the effective filament bending stiffness or stretch modulus vanishes. We also show that our effective medium theory predicts a crossover between affine and non-affine strain, consistent with both prior numerical studies and scaling theory.

PACS numbers: 87.16.Ka, 62.20.Dc, 82.35.Pq

Semiflexible polymer networks form a distinct class of gels whose mechanical properties remain at the frontier of both biophysical and materials research. These cross-linked polymer networks differ substantially from the flexible polymer gels and rubbers [1] due to the rigidity of the individual polymers [2, 3]. Because the thermal persistence length of the constituent filaments is much longer than the typical distance between cross-links, these materials can store elastic strain energy in both stretching and bending deformations of the filaments. The cytoskeleton of eukaryotic cells is a ubiquitous example of such a semiflexible network since it is composed of densely cross-linked, stiff protein aggregates [4]. This network dominates the mechanical properties of the cytosol and lies at the heart of the cellular force production and morphological control.

Theoretical studies of the elastic response of randomly cross-linked stiff filamentous networks have recently uncovered a surprising cross-over between distinct mechanical regimes of these semiflexible networks [5, 6]. For given filament elastic parameters there is a transition from strain energy storage in filament stretching modes at higher network densities to filament bending modes in more sparse networks. This transition is accompanied by a change in the geometry of the deformation field over mesoscopic lengths. At higher densities the network deforms affinely as expected from continuum elasticity theory while at lower densities, where the elastic energy is stored in bending modes, the network deformation field is nonaffine over large mesoscopic distances. Recent experiments [7] support the existence of this affine (A) to nonaffine (NA) cross-over. A fundamental understanding of the relation of the network architecture and individual filament mechanics to the collective elasticity of the network remains elusive. The previous theoretical work had numerically identified the cross-over in simulated networks and provided a scaling argument to account for the dependence of the critical network density upon the mechanics of the constituent filaments. This work, however, was unable to account for the dependence of the elastic moduli on filament properties and network density except in the affine regime.

In this letter we develop an analytical model of the mechanical response of two-dimensional disordered semiflexible networks. We introduce a mechanical mean-field or effective medium theory of the system that allows us to calculate the elastic response of the system to uniformly imposed as well as wave number dependent strain fields. From this mechanical response we identify an A/NA cross-over and obtain a phase diagram of the system showing the affine and non-affine regimes in addition to the mean-field rigidity percolation transition [8, 9]. Our study of the collective shear and bending moduli of the system demonstrates the presence of a natural length scale controlling the A/NA cross-over that corresponds to the analogous quantity determined from the earlier scaling theory and numerical data [6].

We study a model two-dimensional system constructed as follows. We arrange infinitely long filaments in the plane of a two-dimensional hexagonal lattice so that at each lattice point three filaments cross and that each lattice point is connected to its nearest neighbor by a single filament. A sketch of the network is shown in Fig. 1. The filaments are given an extensional spring constant α_m and a bending modulus κ_m. The cross-links at each lattice site do not constrain the angle between the crossing filaments. We introduce quenched disorder into this filament network by cutting bonds with a probability $1-p$, $0 < p < 1$ and study the zero frequency mechanical response of this disordered semiflexible network to uniform and inhomogeneous deformation fields in the linear response regime. Here, we do not explicitly consider thermal fluctuations, whose role in determining the longitudinal compliance of filaments has been discussed before [10]. These thermal effects can be incorporated in the present model through the parameter α_m [6].

The elastic energy of the strained network, which arises from the bending and stretching of the constituent filaments, can be written in terms of the displacement vector $\mathbf{u}_i$ at each lattice site i. To quadratic order in $\mathbf{u}$ the stretching (E_s) and bending (E_b) energies are

$$E_s = \frac{1}{2}\alpha_m \sum_{\langle ij\rangle} (\boldsymbol{u}_{ij}.\hat{\boldsymbol{r}}_{ij})^2 \tag{1}$$

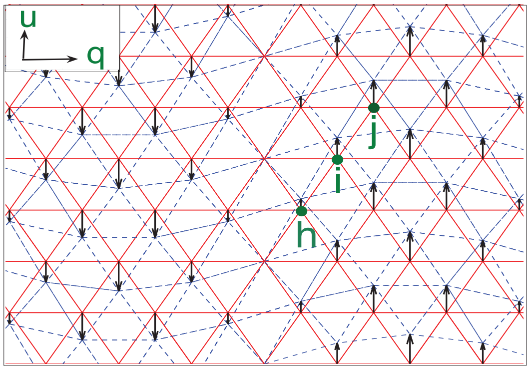

FIG. 1: (color online) Schematic figure of the filament network. The solid red lines represent the undeformed filament network, while the dashed blue lines show the deformation field having wavevector **q** and displacement amplitude **u** (shown in the upper left corner of the figure). The black arrows show the displacement field at each lattice point. This perfect lattice is disordered by making randomly placed cuts in the infinitely long filaments. These are not shown.

$$E_b = \frac{1}{2}\kappa_m R^{-2} \sum_{\langle \widehat{hij} \rangle} \left(\boldsymbol{u}_{ih} \times \hat{\boldsymbol{r}}_{ij} - \boldsymbol{u}_{ij} \times \hat{\boldsymbol{r}}_{ih}\right)^2, \quad (2)$$

where R is the equilibrium lattice constant, $\hat{\boldsymbol{r}}_{ij}$ is a unit vector directed from the i^{th} to the j^{th} equilibrium lattice site, and $\mathbf{u}_{ij}$ is the difference in the strain field between those lattice sites.

It is now simple to determine the collective elastic properties of the perfect lattice; doing so for the disordered lattice generated by randomly cutting the filaments is less trivial. We determine below the spring constant and bending modulus of a spatially uniform effective system [11] that reproduces the mechanics of our disordered system in an average sense, as we describe below.

We first apply a uniform dilation to our system so that all bonds are stretched by $\delta\ell$. There is no bending deformation. If we now replace a single filament segment connecting points (say) i and j (see Fig. 1) by one of spring constant α, the virtual force needed to the fix positions of i and j is $f = \delta\ell(\alpha_m - \alpha)$. If f were applied to the same segment in the unstrained network its change in length would be f/α_{eff} where $\alpha_{\text{eff}} = \alpha_m/a^* - \alpha_m + \alpha$ is the effective spring constant between lattice points i and j with a^* $(0 < a^* < 1)$ being a network material parameter that includes the contribution of the elasticity of the entire network. It may be written in term of the dynamical matrix of the lattice $\boldsymbol{D}(q)$ as

$$a^* = \frac{1}{3}\sum_q Tr\left[\boldsymbol{D}_s(q)\cdot\boldsymbol{D}^{-1}(q)\right] \quad (3)$$

where the sum is over the first Brillouin zone. $\boldsymbol{D}_{s,b}(q)$, $\boldsymbol{D}(q) = \boldsymbol{D}_s(q) + \boldsymbol{D}_b(q)$ define respectively the stretching and bending contributions to the full dynamical matrix and are given by

$$\boldsymbol{D}_s(q) = \alpha_m \sum_{\langle ij\rangle}\left[1 - e^{-i\boldsymbol{q}.\hat{\boldsymbol{r}}_{ij}}\right]\hat{\boldsymbol{r}}_{ij}\hat{\boldsymbol{r}}_{ij} \quad (4)$$

$$\boldsymbol{D}_b(q) = 4\kappa_m \sum_{\langle ij\rangle}\left[1 - \cos(\boldsymbol{q}.\hat{\boldsymbol{r}}_{ij})\right]\left(\boldsymbol{I} - \hat{\boldsymbol{r}}_{ij}\hat{\boldsymbol{r}}_{ij}\right) \quad (5)$$

with $\boldsymbol{I}$ the unit tensor and the sums are over nearest neighbors.

Due to linearity it follows that the additional displacement of this bond without the virtual force is $\delta u = \delta\ell(\alpha_m - \alpha)/\alpha_{\text{eff}}$. The extra displacement δu of the segment due to change in that filament segment's spring constant in the dilated network is the same as its extension in response to the force f being applied to it. Therefore we write the extra displacement on an arbitrary bond due to the substitution of a new elastic element as

$$\delta u = \frac{(\alpha_m - \alpha)\delta\ell}{\alpha_m/a^* - \alpha_m + \alpha}. \quad (6)$$

We now average this extra displacement over the ensemble of possible filament substitutions. In our disordered network we allow each segment to be present with probability p and absent representing a filament end, or cut with probability $1-p$ so that the statistical distribution of longitudinal spring constants is

$$P(\alpha') = p\delta(\alpha_m - \alpha') + (1-p)\delta(\alpha'). \quad (7)$$

To determine the elastic properties of the effective medium we adjust the medium spring constants α_m so that $\langle\delta u\rangle = 0$, *i.e.* the lattice displacement in our spatially homogeneous effective medium material is identical to the average displacement in the spatially heterogeneous disordered material.

Using this procedure we find that the disorder averaged displacement in a network with spring constant α is equal to the uniform displacement of a spatially homogeneous effective medium having spring constant α_m given by

$$\frac{\alpha_m}{\alpha} = \begin{cases} \frac{p-a^*}{1-a^*} & \text{if } p > a^*, \\ 0 & \text{if } p \le a^*. \end{cases} \quad (8)$$

The contribution of network bending to the effective medium spring constant arises only through the effect of the bending modulus in a^* in Eqs. (3,4,5). To determine how the shear modulus depends on the average filament length we note the mean filament length is $\langle L\rangle = pR(2-p)/(1-p)$. We plot in Fig. 2 using the filled symbols the effective medium spring constant as a function of mean filament length measured in units of R.

We now consider the response of the network to a $\boldsymbol{q}$-dependent strain as depicted in Fig. 1. In a manner analogous to the uniform dilation studied above we now apply the $\boldsymbol{q}$-dependent strain. We now modify both the bending modulus and spring constant of one filament spanning

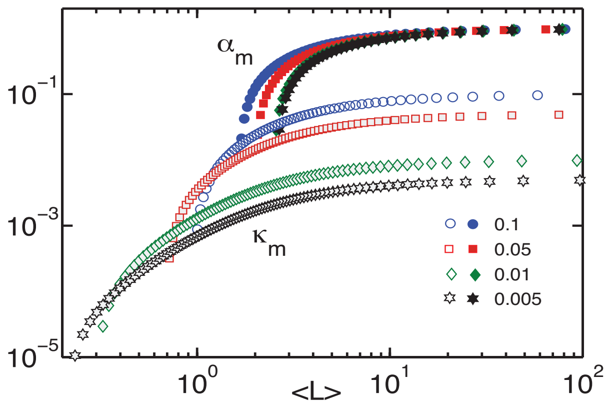

FIG. 2: (color online) The effective medium spring constant α_m (filled symbols) and bending constant κ_m (open symbols) with the average filament length $\langle L\rangle$ (legend shows different values of κ, with $\alpha = 1$).

lattice sites h, i, j so that: $\kappa_m \to \kappa, \alpha_m \to \alpha$ and compute the virtual force and torque needed to maintain the position of site i in the middle of the triad of lattice sites above. See again Fig. 1. From these forces using linearity we compute the components of the displacement of site i along that filament $\delta\ell_{||}$ perpendicular to it $\delta\ell_\perp$ in response to the elastic constant substitution made above. We find

$$\delta\ell_{||} = \frac{(\alpha_m - \alpha)(\boldsymbol{u}_{ij} + \boldsymbol{u}_{ih}).\hat{\boldsymbol{r}}_{ij}}{2(\alpha_m/a^* - \alpha_m + \alpha)},$$
$$\delta\ell_\perp = \frac{(\kappa_m - \kappa)(\boldsymbol{u}_{ij} + \boldsymbol{u}_{ih}).(\hat{z} \times \hat{\boldsymbol{r}}_{ij})}{\kappa_m/b^* - \kappa_m + \kappa} \qquad (9)$$

where a^* is defined in Eq. (3) and the analogous quantity b^* is defined by

$$b^* = \frac{1}{3N}\sum_q Tr\left[\boldsymbol{D}_b(q)\boldsymbol{D}^{-1}(q)\right] \qquad (10)$$

using the same sum over wavevectors as in Eq. (3). Here for semiflexible filaments on a triangular lattice interacting via cross-links that do not apply torques, the stretching and bending modes are orthogonal [12].

We now average these displacements over the disorder, and, to find the effective medium elastic constants we demand that the disorder-averaged displacements vanish so that

$$\langle\delta\ell_{||}\rangle = 0, \quad \langle\delta\ell_{||}\rangle = 0. \qquad (11)$$

The probability distribution for α is given by Eq. (7), but a nonzero value of the bending modulus at site i requires the presence of *both* filament segments on either side of that site so that

$$P(\kappa') = p^2\delta(\kappa - \kappa') + (1 - p^2)\delta(\kappa'). \qquad (12)$$

Since we consider uncorrelated distributions of the bending and elastic constants we find the effective

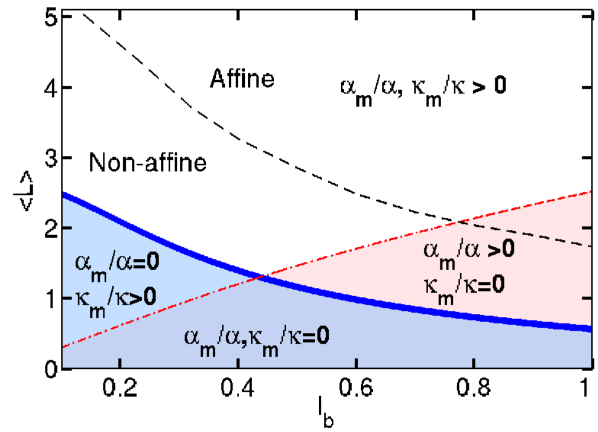

FIG. 3: (color online) The effective medium mechanical phase diagram spanned by $\langle L\rangle$ and l_b. The thick solid line marks the rigidity percolation transition where the material acquires a finite shear modulus. The dashed line shows the crossover from the non-affine to the affine regime.

medium elastic constants α_m and κ_m by solving Eqs. (9, 11) independently to arrive at

$$\frac{\alpha_m}{\alpha} = \begin{cases} \frac{p-a^*}{1-a^*} & \text{if } p > a^* \\ 0 & \text{if } p \le a^* \end{cases} \qquad (13)$$

$$\frac{\kappa_m}{\kappa} = \begin{cases} \frac{p^2-b^*}{1-b^*} & \text{if } p > \sqrt{b^*}, \\ 0 & \text{if } p \le \sqrt{b^*} \end{cases} . \qquad (14)$$

Figure 2 shows the effective medium values of α_m (filled symbols) and κ_m (open symbols) as a function of the filament mean length that is fixed by p for different values of single filament bending rigidity κ, at a fixed value of the single filament spring constant $\alpha = 1$. The unit of length is the lattice constant R (set to unity) and the energy units are arbitrary.

There are three length scales in the system: (i) the average length of filaments $\langle L\rangle$, (ii) a length $l_b = (\kappa/\alpha)^{1/2}$ associated with the relative ease of filament stretching to bending, and (iii) the mean distance between cross links, which, to a good approximation is $R = 1$[13]. We present a mechanical phase diagram of our system spanned by $\langle L\rangle$ and l_b in Fig.3 that shows the regimes corresponding to zero and finite values of κ_m and α_m. Generically for long enough filaments the system has a finite collective extension and bending modulus. As the mean filament length is reduced the collective shear modulus (proportional to α_m) vanishes at the rigidity percolation transition. There is also predicted a new rigid phase ($\alpha_m > 0$) that has a vanishing collective bending modulus. We further note that the lower range in l_b corresponds to nonthermal systems in which the distance between crosslinks is large compared with the molecular scale (i.e., at low volume fraction), but that with thermal effects, one effectively goes to higher l_b.

The collective elastic properties of the effective

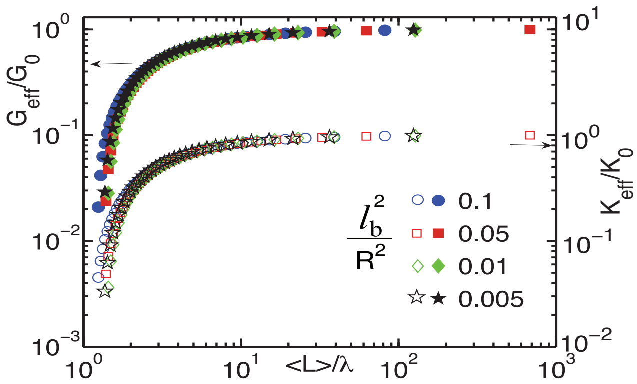

FIG. 4: (color online) The effective medium shear $G_{\rm eff}/G_0$ (filled symbols) and bending moduli $K_{\rm eff}/K_0$ (open symbols) normalized by their respective values for a perfect network are plotted as a function of the mean filament length $\langle L\rangle$ divided by the nonaffinity length $\lambda = R/(R/l_b)^z$ with $z = 0.25$. Data collapse is shown for four data differing in l_b. The legend shows different values of l_b^2.

medium can be calculated from the stored elastic energy density $\mathcal{E}$ under a given imposed network strain. For strain field of the form $\boldsymbol{u} = R\gamma\cos(q\cdot\mathbf{x})\hat{z}\times\hat{q}$ the shear $G_{\rm eff}$ and bending moduli $K_{\rm eff}$ can be extracted as the coefficients of the q^2 and q^4 terms of $\langle\mathcal{E}\rangle/\gamma^2$ where the angled brackets imply an angular average of the direction of $\mathbf{q}$ with respect to underlying lattice. $G_{\rm eff}$ is a function of α_m alone while $K_{\rm eff}$ is a function of κ_m and α_m.

In Fig. 4 we plot the effective medium shear and bending moduli as a function of $\langle L\rangle$. Motivated by earlier work[6] on the A/NA transition we have rescaled $\langle L\rangle$ by $\lambda = R(R/l_b)^z$ with $z = 1/4$. A comparison of $G_{\rm eff}$ in this figure with $\alpha_{\rm eff}$ from Fig. 2 demonstrates a remarkably accurate data collapse whose accuracy is enhanced as we move farther away from the rigidity percolation. Moreover, we find that the same rescaling factor generates an equally accurate collapse of the $K_{\rm eff}$ data.

The single parameter collapse of our calculated elastic moduli is highly reminiscent of the observed numerical data collapse semiflexible network simulations. Although the analytic result gives a scaling exponent $z = 1/4$ while the previous numerical results were consistent with $z = 1/3$, it is tempting to associate the new length scale introduced (λ) with the nonaffinity length that generated such a data collapse in the previous work. The effective medium approach does not allow us to explore the spatial heterogeneities of the strain field under uniformly imposed shear so it is not possible with this technique to explore the geometric interpretation of λ.

It should be noted, however, that this effective medium that fails to account for the correct spatial structure of the strain field in the disordered material does show an abrupt cross-over that appears mechanically identical to the A/NA cross-over and is controlled by a single emergent length scale, λ, which obeys a similar scaling relation to that found empirically from previous numerical results. From these $G_{\rm eff}$ plots (Fig. 4) we have extracted the A/NA cross-over from the location of the largest change in the slope of the curves. This A/NA boundary is plotted in Fig. 3.

In conclusion, we used an effective medium theory to explore the mechanical properties of disordered filament networks. We find that this mean-field approach to the mechanics of such networks captures the mechanical aspects of the A/NA cross-over including the identification of an emergent mesoscopic length scale λ controlling the mechanics of the system.

MD and AJL thank M. J. Thorpe for useful discussions and acknowledge support from nsf-dmr0354113. FCM acknowledges the hospitality of the UCLA chemistry department. This work was supported in part by the Foundation for Fundamental Research on Matter (FOM).

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[13] The mean distance between cross links is always slightly greater than R due to missing cross-linking filaments in sparse networks. Corrections to the mean distance between cross-links grow as $(1-p)^4$.