Stability and entropic elasticity of sub isostatic random-bond networks

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We study the elasticity of thermalized spring networks under an applied bulk strain. The networks considered are sub-isostatic random-bond networks that, in the athermal limit, are known to have vanishing bulk and linear shear moduli at zero bulk strain. Above a bulk strain threshold, however, these networks become rigid, although surprisingly the shear modulus remains zero until a second, higher, strain threshold. We find that thermal fluctuations stabilize all networks below the rigidity transition, resulting in systems with both finite bulk and shear moduli. Our results show a $T^{0.66}$ temperature dependence of the moduli in the region below the bulk strain threshold, resulting in networks with anomalously high rigidity as compared to ordinary entropic elasticity. Furthermore we find a second regime of anomalous temperature scaling for the shear modulus at its zero-temperature rigidity point, where it scales as $T^{0.5}$, behavior that is absent for the bulk modulus since its athermal rigidity transition is discontinuous.

I. INTRODUCTION

Materials such as plastics and rubbers as well as tissues and living cells contain polymer networks, which, among other roles, provide structural support to these materials. Tissues and cellular networks are especially sensitive to external stresses [1–7], and a number of theoretical and simulation studies have attempted to gain an understanding of what controls the response of such systems to deformations [8, 9]. In 1864, Maxwell showed that there is a connectivity threshold $z_c$, determined by the average coordination number of the network nodes, at which athermal networks of springs become rigid [10]. This threshold, referred to as the isostatic point, occurs when the number of degrees of freedom of the network nodes are just balanced by the number of constraints arising from the springs. This purely mechanical argument has been used to describe the stability systems ranging from emulsions and jammed particle packings [11, 12] to amorphous solids [13] and folded proteins [14]. Beyond this, theoretical work has shown that there are numerous ways of stabilizing a network, and therefore tuning its rigidity, below the isostatic point [15]. Examples include the addition of a bending stiffness to the model filaments [16, 17], by applying internal stresses via molecular motors [19, 20] or by placing the network under tension by applying a bulk strain to the system [21]. It has been shown that a network’s rigidity point can be shifted from the Maxwell point by adding these interactions and forces to the system. In the case of applying a bulk strain [21] the system can be stabilized by stretching the network until all the floppy modes have been pulled out, resulting in a critical strain at which the network is just rigid.

In addition to these athermal models, recent work has shown how temperature can stabilize a mechanically floppy network [22, 23]. It was found that at and below the isostatic point the network response to deformation, defined by the shear modulus, not only becomes finite when thermal fluctuations are present, but that it also shows an anomalous temperature scaling of $T^\alpha$, where $\alpha < 1$. This sub-linear temperature dependence indicates that a network would exhibit a larger resistance to deformation than would be expected from entropic elasticity, where one would expect a linear temperature dependence [24]. The origin of this anomalous temperature dependence remains unclear, and in addition there have been few studies into the effects of thermal fluctuations on sub-isostatic networks [22, 25–27]. Furthermore, in Ref. [23] a triangular lattice based network was used, and an open question is how general the anomalous regimes found are, since network architecture can have vast effects on a systems response to deformation [28, 29].

In this paper, we study the effects of thermal fluctuations on an under-constrained and mechanically floppy random-bond network. The architecture of a random-bond network is as different as possible from a triangular lattice network, as the nodes are arranged isotropically and their is a distribution of filament lengths. The random-bond model has been used previously to study the effects of applying a bulk strain on the rigidity of athermal networks [21]. We study the temperature dependence of the internal pressure, bulk modulus and shear modulus of a sub-isostatic random-bond network with an average coordination number of $z = 3 < z_c$, and furthermore, we examine how the bulk strain effects the temperature dependence of the network rigidity. We show that, as reported previously in Ref. [21], there exists a bulk strain threshold at which the system will begin to resist bulk deformations at zero temperature. However, the network does not begin to resist shear deformation until a second, higher, strain threshold is reached, and it is these two thresholds that control the network response to the applied deformations. We find anomalous scaling regimes for the shear modulus at and below its threshold, similar to the results of Ref. [23], where the bulk strain applied to the networks in this study takes
on a similar role to the connectivity in Ref. [23]. Interestingly, we find that, while the bulk modulus exhibits a similar anomalous scaling regime below its threshold, we find no temperature dependence at its strain threshold, at which there is a first order zero-temperature rigidity transition. The network behavior is summarized in the phase diagrams shown in Fig. 1.

![Phase Diagram](image)

**FIG. 1.** Schematic phase diagram showing behavior of the internal pressure \( P \), bulk modulus \( K_A \) and shear modulus \( G \) with bulk strain \( \epsilon \) and temperature \( T \) for sub-isostatic random-bond networks with connectivity \( z = 3 \).

### II. PHYSICAL PICTURE

Since Maxwell [10] it has been known that an athermal network of central-force springs will be floppy below a critical, isostatic connectivity threshold. This means that there is no energy cost for small bulk or shear deformations. When applying an increasingly large uniform bulk strain, such networks will begin to resist additional bulk deformations at a strain threshold corresponding to a rigidity transition [21], at which the network will be just rigid. Applying small deformations on a rigid network will cost energy, since the springs will be stretched, which results in a stable network exhibiting a non-zero bulk modulus at zero temperature.

A mechanically floppy network will also be stabilized by thermal fluctuations [22, 23]. The resulting network is rigid both above and below the rigidity point. A deformation of a mechanically floppy network results in a reduction of the number of microstates that the system can assume, even though the system energy remains unchanged. This results in a change in entropy as the system is deformed, which gives rise to a change in the free energy, resulting in non-zero elastic moduli at finite temperatures. Thus, below the rigidity point the network is stabilized by thermal fluctuations, as the entropic contribution to the moduli dominate over the mechanical contribution. When the network is sufficiently stretched, i.e., above the bulk strain rigidity threshold, all springs are under tension that causes the mechanical stretching energy (controlled by the spring constant) to dominate the thermal fluctuations in stabilizing the network, and the network rigidity then becomes independent of temperature. As thermal networks are always rigid, there is no bulk strain threshold at which the network becomes stable. However, if a network is taken to the rigidity point, we find that there can be an anomalous intermediate regime in which the network is stabilized by both temperature and the spring constant.

These three different regimes of network stability are defined by the bulk strain at the zero temperature rigidity transition. This strain depends on how constrained the system is, controlled, for example, by varying the connectivity of the network by changing the number of springs. Lowering the connectivity will lower the number of constraints in the network and it has been shown that sub-isostatic networks with increasingly lower connectivities need to be stretched increasingly more to become rigid [21].

### III. THE MODEL

In this paper we study the effects of thermal fluctuations and bulk strain on the stability of sub-isostatic random-bond networks. The random-bond network is constructed by placing \( N \) nodes randomly in a 2 dimensional box of area \( A \), which are then randomly connected by \( N_{sp} \) springs until the network reaches an average connectivity \( z = 2N_{sp}/N \). A schematic of a random-bond network is shown in Fig. 2. Periodic boundary conditions are used throughout, and the springs may cross the system boundaries. The springs have a rest length \( l_0 \), which will vary for each spring and, by construction, the average rest length will be half the system size. We use the average spring length \( \langle l_0 \rangle \) as the unit of length, and we note that for systems with the same density of nodes \( \langle l_0 \rangle \) grows as \( \sqrt{N} \), and as such there is no well defined thermodynamic limit. In this simple model the only two energy scales are the stretching energy and the thermal energy. The total energy of the network is given by the sum of the energy of all \( N_{sp} \) springs

\[
U = \frac{k_{sp}}{2} \sum_i N_{sp} \langle l_i - l_{0,i} \rangle^2.
\]

where \( k_{sp} \) is the spring constant and \( l_{0,i} \) the rest length of spring \( i \) which has length

\[
l_i = \sqrt{(x_2 - x_1)^2 + (y_2 - y_1)^2},
\]

where \( x_j, y_j \) are the coordinates of nodes \( j = 1, 2 \) that are connected by spring \( i \).

This network architecture is isotropic and differs qualitatively from a lattice-based network, for which the springs have either the same length or a narrow discontinuous distribution of lengths. In the random-bond network, there are springs with lengths of the order of the system size, which would prevent network collapse at finite temperature due to entropic forces [23, 30]. Thus, the random-bond model is stable to thermal fluctuations without an imposed tension at the boundaries.
We introduce the temperature $T$ and $y$ where $A$ is the rest area of a fully relaxed network at temperature $T = 0$, and $\epsilon$ is the applied strain. The $x$- and $y$-coordinate of each node are also scaled, defining new coordinates $x'$ and $y'$ for node $j$ as
\[ x'_j = x_j (1 + \epsilon). \]

We introduce the temperature $T$ using Monte Carlo simulations to study the equilibrium behavior of thermal systems.

### A. Elastic moduli and internal pressure

We determine the internal pressure and bulk modulus of the system under bulk strain. The internal pressure is defined as
\[ P = -\frac{\partial F}{\partial A}, \]
where $F$ is the Helmholtz free energy, and can be calculated as
\[ P = \frac{N}{A} k_B T + \frac{1}{2A} \sum_i \sum_j \langle f_{ij} \cdot l_{ij} \rangle \]
\[ = \frac{N}{A} k_B T - \frac{1}{2A} \sum_k \langle k_{sp}(l_k - l_0) \rangle, \]
where the first line contains a sum over all pairs of nodes and the second line contains a sum over all springs, since the force $f_{ij}$ between node $i$ and node $j$ is only non-zero if there is a spring connecting $i$ and $j$. The first term represents the ideal gas behavior and the second term corrects for spring interactions. By calculating the internal pressure at various areas we can then calculate the bulk modulus, which is defined by
\[ K_A = -A \frac{\partial P}{\partial A}. \]

In addition, we calculate the shear modulus $G$ of the networks at each bulk strain. $G$ is defined by
\[ G = \frac{1}{A} \frac{\partial^2 F}{\partial \gamma^2}, \]
where $\gamma$ is the shear strain. In order to shear the network we use Lees-Edwards boundary conditions [32], where the energy of the springs crossing the top boundary of the simulation box is modified to become
\[ E_{sp}(l) = \frac{k_{sp}}{2} \left( \sqrt{(x_{ij} + \gamma L_y)^2 + (y_{ij})^2} - l_0 \right)^2, \]
where $L_y$ is the height of the simulation box. We initially shear the networks at zero temperature, obtaining a configuration under shear, and then increase the temperature from zero. For these thermal systems, we calculate the shear stress $\sigma$ as in Refs. [23, 33]. The shear modulus can then be calculated by taking the derivative of the stress on the network with respect to $\gamma$ at $\gamma = 0$.

### IV. RESULTS

We calculate the pressure, bulk modulus and shear modulus for two-dimensional random-bond networks with a connectivity of $z = 3$ over a range of reduced temperatures $T^* = k_B T / k_{sp}(l_0)^2$ and bulk strains $\epsilon$. For these systems, the critical connectivity is $z_c \sim 4$. Thus, our networks are subisostatic and will be floppy at $T = 0$ and $\epsilon = 0$. Results for the pressure are presented in Fig. [3] for the bulk modulus in Fig. [4] and for the shear modulus in Fig. [5]. We first examine in detail the behavior of the properties related to bulk deformation, i.e., the pressure and bulk modulus, before examining the behavior of the shear modulus.

At zero temperature we find a strain $\epsilon_1$ (with corresponding area $A_1$) at which the network just becomes rigid, indicated by the solid black line in Figs. [3] and [4]. Here the network exhibits a finite pressure and bulk modulus above $\epsilon_1$, and zero pressure and bulk modulus below, and we hence define this strain threshold as the rigidity point. The pressure shows a linear dependence on area, increasing continuously as $P = c_1 (A - A_1)$ for $A \geq A_1$, where $c_1$ is a constant. Based on the definition of the bulk modulus given in Eq. (7) this means that $K_A = c_1 A$ for $A \geq A_1$ and $K_A = 0$ for $A < A_1$, i.e., a discontinuous increase in $K_A$, corresponding to a first order transition from a floppy to a rigid network at $\epsilon_1$. We note that the value of $\epsilon_1$ will differ for different
network configurations, as there is no well-defined thermodynamic limit for random-bond networks due to the average spring length growing with the system size. For the results presented in Figs. 3(a) and 3(a) a network with $\epsilon_3 = 0.0356$ was used. The first-order nature of the transition was present in all configurations studied.

When thermal fluctuations are present the network is rigid for all bulk strains, as can be seen in Figs. 3(a) and 3(a) where different temperatures are represented by the colored points. For small bulk strains ($\epsilon < \epsilon_1$) the network is stabilized by the thermal fluctuations and exhibits an increasingly large pressure and bulk modulus as the temperature is increased. As $\epsilon_1$ is approached we observe a regime where the pressure and bulk modulus for all temperatures start to join the zero temperature line, with the low temperature results starting to join the zero-temperature result sooner than the results for higher temperatures. For bulk strains greater than $\epsilon_1$ there is a mechanical regime, where tension is dominant over thermal fluctuations and the resistance to deformation depends only on the spring constant. However, we find that the pressure no longer increases linearly with area as $\epsilon_1$ is approached, even at low temperatures (see the inset of Fig. 3(a)), resulting in a continuous transition between the thermal-dominated regime and the mechanical regime.

In Figs. 3(b) and 3(b) we show the temperature dependence of the internal pressure and bulk modulus in the thermal, intermediate and mechanical regimes. Above $\epsilon_1$, we find that they are both independent of temperature; in this mechanical regime the network is completely stabilized by the spring constant and its response to deformation is invariant to temperature. At and below the rigidity point, the temperature dependence becomes more complex. Below $\epsilon_1$ the pressure in the network scales as $P \propto T^{\alpha}$. When the network is at zero strain $\epsilon = 0$ we find that $\alpha = 1$, as expected in analogy to entropic elasticity [24]. However, as the strain is increased we find $\alpha < 1$, with an exponent that decreases as the strain is increased, reaching $\alpha \sim 0.66$ as the critical strain is approached. We observe this dependence only at low temperatures $T^* < 10^{-5}$, with the pressure scaling linearly at higher temperatures. This varying temperature dependence of the pressure can be understood when we consider the behavior of pressure in the initial linear response regime. That is, at low bulk strains we find that the pressure scales linearly with area and at low temperatures can be expressed as $-P = m(T) + c(T)$, where $m(T)$ and $c(T)$ are constants for a given temperature $T$. It then follows that the bulk modulus will scale as $K_A = m(T)A$. By fitting this expression for the pressure to our simulation data we find that $m(T) \propto T^{0.66}$.
Hence, at low bulk strains \((A \sim A_0)\) \(c(T)\) dominates and we find a linear temperature dependence, while at higher bulk strains the system approaches a regime where \(n(T) \ast (A - A_0)\) dominates over \(c(T)\) and we hence observe a \(T^{0.66}\) dependence, with a mixed regime between the two. The bulk modulus then scales with \(T^{0.66}\) for all \(\epsilon < \epsilon_1\) at low \(T^*\) and linearly at higher temperatures. On dimensional grounds it follows that the pressure and bulk modulus must also have a dependence on the spring constant, and as \(P, K_A \propto T^{\alpha k_{sp}^{1-\alpha}}\).

For bulk strains close to \(\epsilon_1\), we find that \(P\) scales with the square root of temperature, \(P \propto T^{0.5}\), for \(T^* < 10^{-5}\) and linearly with temperature for \(T^* > 10^{-5}\), as shown in Fig. 3(b). In the \(T^{0.5}\) regime the network is again stabilized by both temperature and the spring constant, and we find that \(P\) scales as \(T^{0.5}k_{sp}^{0.5}\). We also observe that networks below the rigidity point can enter this regime as the temperature is increased. For these systems the pressure initially shows a \(T^{0.66}\) dependence before they then show a \(T^{0.5}\) dependence, indicating a regime that fans out from the zero-temperature rigidity point. The bulk modulus, however, exhibits a different behavior in this region, as for networks at \(\epsilon_1\) we find that \(K_A\) is independent of temperature. For networks just below this point we observe a rapid increase in the modulus with \(T\), before \(K_A\) reaches the zero-temperature value.

As the area is increased beyond \(\epsilon_1\), there is a clear inflection point in the zero-temperature (and low temperature) pressure, as can be seen in Fig. 3(a). At this point the pressure again increases linearly with area as \(-P = \epsilon_2(A - A_2)\), where \(\epsilon_2\) and \(A_2\) are larger than \(\epsilon_1\) and \(A_1\), respectively. This corresponds to a reorganization of the network, as the nodes change positions to minimize the system energy. This is illustrated in Fig. 3(c), where we plot the fraction of springs in the network that are activated (i.e., stretched or compressed such that \(l \neq l_0\)). At \(\epsilon_1\), we see the first springs become activated, followed by a significant jump at a higher value of \(\epsilon\). Furthermore, as the area is increased beyond this point, we find several more reorganizations, as can be seen by the kinks in Fig. 3(c) (there are also further kinks in the pressure in Fig. 3(a), although these are not visible on the log scale used). This is present in all configurations and system sizes studied, and in Fig. 3(c) we present data from additional configurations to illustrate this. The effect that this has on the bulk modulus can be seen in Fig. 3(a), where we see that there is a second distinct jump in \(K_A\), corresponding to a first order transition as the system rearranges, with further jumps present at higher areas, although again, these are not visible on the log scale used.

We now examine the behavior of the linear shear modulus \(G\), which we obtained by shearing the networks at each bulk strain. For athermal networks \(G\) is zero at low bulk strains, as one would expect for a floppy network before any of the springs become stretched. However, the shear modulus remains zero beyond \(\epsilon_1\), with the network not resisting shear deformation until it reaches a bulk strain \(\epsilon = \epsilon_2\) (see Fig. 3(a)). This strain corresponds to that at which we observed the second jump in the bulk modulus as shown in Fig. 3(a). Beyond this point the shear modulus increases linearly with the area and the network becomes rigid to shear deformation, indicating a continuous transition in \(G\). As for the pressure and bulk modulus, when thermal fluctuations are present we find a non-zero shear modulus throughout, with thermal, intermediate and mechanical regimes present, although here the intermediate regime is found at \(\epsilon_2\). The different regimes can be seen in Fig. 5, where we see \(G\) remaining constant with temperature above \(\epsilon_2\) and \(G\) scaling with \(T^0\) at and below \(\epsilon_2\), with \(\alpha \sim 0.66\) below and \(\alpha \sim 0.5\) in the intermediate regime.

![FIG. 5. Shear modulus \(G\) of random-bond spring network with \(N = 1000\) nodes against \(a\) bulk strain \(\epsilon\) and \(b\) reduced temperature \(T^* = k_B T / k_{sp} (l_0)^2\). Solid line in \((a)\) shows zero temperature behavior, while points are for thermal systems. Solid black line in \((b)\) shows \(T^{0.66}\) dependence while solid blue line shows \(T^{0.5}\) dependence.](image-url)
\[ P = |\epsilon - \epsilon_1|^k P(T|\epsilon - \epsilon_1|^{-l}), \]  

(11)

where \(a/b \) and \(k/l\) are the exponents in the intermediate regime for, respectively, the shear modulus and pressure. The best collapses of the data are shown in Figs. 6 and 7, where we use the critical exponents \(a, k = 1\) and \(b, l = 2\). The two collapses summarize the three regimes of network stability. The upper left branches show the mechanical regimes, the lower left branch shows the temperature dominated regime, where we find \(T^{0.66}\) dependence for the shear modulus and the varying \(T\) dependencies for the pressure, and the right branch shows the intermediate regime, where we find a temperature dependence of \(T^{0.5}\) for both \(G\) and \(P\).

The behavior of the sub-isostatic random-bond networks considered in this paper is similar to the behavior found in Ref. [23] for lattice based networks. The observed sublinear scaling of the shear modulus, \(G \propto T^\alpha\), for networks below the critical bulk strain was also found for lattice-based networks, albeit with different exponents, with \(\alpha \sim 0.66\) for the random-bond networks studied here and \(\alpha \sim 0.8\) for the triangular lattice networks studied in Ref. [23]. This indicates that, while sublinear scaling is not confined to lattice models, the exponent does depend on the topology of the network. In Ref. [23], it was proposed that the scaling may be due to the internal pressure \(P\), which at \(\epsilon = 0\) scales linearly with temperature, leading to \(G \propto k_{sp}^2 T^{0.8}\). This was in analogy to a study on athermal networks with an internal stress \(\sigma_m\) induced by molecular motors, where \(G \sim k_{sp}^{0.2} \sigma_m^{0.8}\) below the isostatic point [24]. However, as we find that the pressure begins to scale sublinearly with temperature as the bulk strain is increased from \(\epsilon = 0\) while the \(G \propto T^\alpha\) scaling remains, this proposed scaling would not be valid as one moves away from the rest area of the network at \(\epsilon \neq 0\). Indeed, the shear modulus shows the same temperature dependence as the bulk modulus, which scales as \(K_A = m(T)A\), where \(m(T) \sim T^{0.66}\) was obtained from the relation for the pressure \(-P = m(T) A (A - A_0) + c(T)\).

In addition to the similarities between the behavior found here for sub-isostatic, sub-critical random-bond networks and sub-isostatic lattice based networks, we also note the similarities between the behavior of networks at the bulk strain threshold corresponding to the rigidity point, and networks at the critical connectivity \(z_c\). In Ref. [23] it was found that the shear modulus behaved as \(G \propto T^{0.5}\) at \(z_c\) (at the critical connectivity the critical strain is zero, \(\epsilon_c = 0\) [21]), indicating that the stabilization of the network at the critical strain is similar to that at \(z_c\). We note that this is only true of the shear modulus, as we find a constant bulk modulus for low temperatures at \(\epsilon_1\). A possible reason for the differences in the observed temperature dependence between the two moduli would be the nature of the zero-temperature transition from zero to finite modulus, as the bulk modulus exhibits a first-order transition at \(\epsilon_1\) while the shear modulus exhibits a continuous transition at \(\epsilon_2\). We also note that the exponents found for the crossover scaling ansatz in Eq. (11), \(a = 1\) and \(b = 2\), are more mean field-like than those found for the critical connectivity case [18, 23].

Finally, the zero-temperature behavior of the random-bond networks considered here differs greatly from that of lattice based networks, exhibiting a non-continuous transition from a floppy to a rigid network as the bulk strain is increased [21] and exhibiting a regime where the system has a finite bulk strain but zero shear modulus. However, despite these differences in the athermal behavior, as previously mentioned the temperature dependence of the thermal stiffening of the network does not change qualitatively [23].
VI. CONCLUSION

In this paper we have studied the effects of thermal fluctuations on the elastic response of random-bond networks at various bulk strains. Our results show that, in agreement with previous studies, there is a bulk strain threshold at zero temperature for which the bulk modulus and pressure of a floppy network will become finite. We find that the transition for the pressure is continuous while it is discontinuous for the bulk modulus, jumping to a finite value at the rigidity point. We have also found that random-bond networks can exhibit further discontinuous transitions, as the networks rearrange to minimize their energy. Unusually, the random-bond networks studied here exhibit a regime where there is a finite bulk modulus but zero shear modulus at zero temperature. In these systems, the bulk strain threshold for a non-zero shear modulus is larger than that for a non-zero bulk modulus, and the shear modulus transitions continuously at its rigidity point.

When thermal fluctuations are present the network becomes stable for all strains, and the pressure and bulk modulus transitions continuously between a thermally dominated regime and a mechanical regime at the zero-temperature rigidity, while the shear modulus transitions continuously at its own bulk strain threshold. In between these two regimes, there exists a third, intermediate regime where the pressure and shear modulus depend on the square root of temperature (at their respective strain thresholds) while the bulk modulus remains constant, as the intermediate scaling occurs only at a continuous rigidity transition. Perhaps most interestingly, we find that the shear and bulk moduli exhibit an anomalous temperature scaling of $T^\alpha$ with $\alpha \sim 0.66$ below the critical strain, where we would expect to find normal entropic elasticity (linear temperature scaling $\approx T$). This behavior is similar to that reported in Ref. [24], where the shear modulus was found to scale as $T^{0.8}$, indicating that floppy networks of various topologies can exhibit anomalous temperature scaling.

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