Soft Matter

Accepted Manuscript

ROYAL SOCIETY OF CHEMISTRY

www.rsc.org/softmatter

Thermalized connectivity networks of jammed packings

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(Dated: June 21, 2016)

Jammed packings of repulsive elastic spheres have emerged as a rich model system within which elastic properties of disordered glassy materials may be elucidated. Most of the work on these packings have focused on the case of vanishing temperature. Here, we explore the elastic properties of the associated connectivity network for finite temperatures, ignoring the breaking of bonds and the formation of new ones. Using extensive Monte Carlo simulations, we find that, as the temperature is increased, the resulting spring network shrinks and exhibits a rapidly softening bulk modulus via a cusp. Moreover, the shear modulus stiffens in a fixed volume ensemble but not in a fixed pressure ensemble. These counter-intuitive behaviors may be understood from the characteristic spectrum of soft modes near isostaticity, which resembles the spectrum of a rod near its buckling instability. Our results suggest a generic mechanism for negative thermal expansion coefficients in marginal solids. We discuss some consequences of bond breaking and an apparent analogy between thermalization and shear.

Jammed packings of repulsive elastic particles exhibit a number of remarkable features that makes them different from conventional solids and a rich model system for fragile matter [1, 2]. One of these features is an excess of soft, low-frequency vibrational modes [3], the so-called boson peak. Numerous consequences can be derived from this peculiar vibrational spectrum, with regards to, for instance, elastic or transport properties [4–6]. Some of these features are shared with lattices close to isostaticity [7–12], which may be exploited to develop meta-materials with novel mechanical properties [13, 14].

While most studies have focused on the zero-temperature consequences of the vibrational spectrum, we here study the impact of thermal fluctuations. Specifically, we consider the harmonic connectivity network obtained from a jammed packing of repulsive, frictionless spheres close to isostaticity, and study its mechanical properties as we heat up the system to a low but finite temperature.

Elastic properties of ordered and disordered networks of springs at finite temperatures have been studied previously [15–17]. Most recently, motivated by the attractive properties of highly responsive marginal solids for material science and biophysics, spring networks have been studied near the isostatic threshold [10–12, 18]. These studies revealed, amongst others, interesting anomalies in the entropic elasticity. While these studies have focused on networks that have soft bulk and shear moduli, as in rigidity percolation [19], it is a characteristic of jammed networks, studied in this work, to have a finite bulk modulus at isostaticity [8]. As we will see, this has major consequences for the impact of thermal fluctuations on the material properties of the network.

Simulation approach. To prepare the initial conditions for our simulations, we generate jammed packings of repulsive elastic spheres. Each packing is created through an energy minimization protocol with thermal equilibration, as described in Ref. [4]. The protocol results in a series of contact networks at different coordination numbers above a critical value $z_c = 4$.

The minimal contact number z_c per particle required for rigidity follows from Maxwell's counting argument [20]. Since there are d degrees of freedom for a point particle, one needs at least $z_c = 2d$ contacts to constrain the positions of all particles. Networks with z_c contacts are called isostatic.

Our hyperstatic contact networks ($z > z_c$) are then modeled as a network of Hookian springs. The effect of thermal fluctuations on our spring network is simulated as in Ref. [15] using the Metropolis Monte Carlo algorithm. Briefly, in each computational step, a vertex trial move is made. Energy decreasing steps are always accepted. Steps that increase the energy are accepted with a probability proportional to the corresponding Boltzmann factor. After each vertex trial move, a trial move to change the volume is performed.

Due to the small temperatures considered, crossing of bonds is very unlikely, although it is not penalized in our network simulations. Also, contacts do neither break nor form in the course of our network simulations. (Some consequences of bond breaking in particle packings are discussed below.)

We use these simulations to extract, first as a function of temperature, pressure and coordination, the elastic network properties. We mostly focus on the total network area $A(T, p, z)$ and the negative thermal expan-

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sion coefficient $NTE \equiv \partial_T \ln(A)$. The elastic response to uniform compression and shear are described by the bulk modulus $B^{-1} \equiv -\partial_p \ln(A)$ and the shear modulus $G \equiv \partial_\alpha \sigma_{\rm shear}|_{\alpha=0}$, respectively. (The shear stress $\sigma_{\rm shear}(T, p, z, \alpha)$ is a function of shear angle α.) These linear response coefficients are determined in our simulations from the mean squared displacement of area and shear-angle via fluctuation-dissipation relations [21]. Finally, we explore how the elastic properties change when adjust the pressure to keep the volume fixed (NVT ensemble).

I. SIMULATION RESULTS

At fixed tension (negative pressure), we observe that our spring networks contract linearly with increasing temperature, as was found previously in high-coordination number networks [15]. Importantly, we find that the contraction diverges in a characteristic way as the connectivity approaches isostaticity.

Specifically, we find that the negative thermal expansion coefficient, NTE, exhibits a singularity as the average contact number per particle approaches isostaticity. For vanishing tension τ, we find that the NTE scales inversely with the distance $\delta z = z - z_c$ from the critical contact number. For large tensions, on the other hand, the NTE$\sim \tau^{-1/2}$ is independent of contact number. Fig. 1 shows that the data for different T, δz and τ collapse onto a master curve when we plot NTE δz vs. $\tau \delta z^{-2}$. The deviations suggest that the collapse for larger tensions only works for small enough δz.

Like jammed packings, our networks are stiff to a uniform compression. A relative change $\delta A/A$ in area requires a similar relative change of the length of most springs. Consequently, the scale of the bulk modulus B_0 of zero-temperature jammed networks is set by the product of spring constant and co-ordination number.

By contrast, as we turn on temperature, we see that the material becomes much softer to compression, which is manifest by a cusp in the bulk modulus. Specifically, the difference $B^{-1} - B_0^{-1}$ scales as $T\delta z^3$ for vanishing tension, Fig. 2. As we increase the tension on the network, the behavior crosses over to being proportional to $T/\tau^{3/2}$.

The shear modulus is inconspicuous for small enough tension, as it follows the zero temperature result $G \sim \delta z$, which is small near the critical point due to soft modes. For fixed area, however, the shear modulus crosses over to $G \sim T^{1/3}$ once the product $T\delta z^3$ becomes of order unity, as shown in Fig. 3. This behavior is markedly different from the scaling $G \sim T^{1/2}$ observed in disordered spring networks near isostaticity [10].

In summary, we find that jammed networks contract upon heating and, respectively, become more tensed at fixed volume. As a consequence, the bulk modulus softens for fixed pressure and the shear modulus hardens for fixed volume.

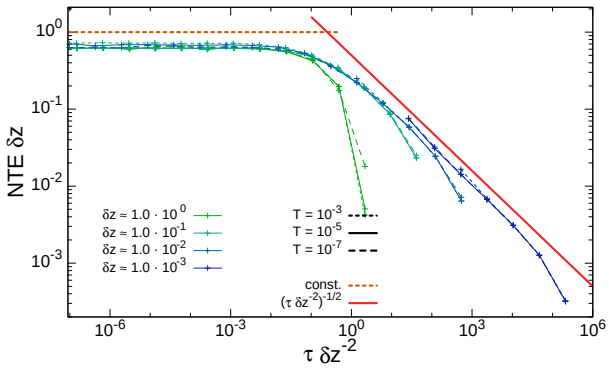

FIG. 1: Scaled Negative thermal expansion coefficient (NTE) as a function of rescaled tension $\tau\delta z^{-2}$ for various combinations of temperature and coordination number difference δz from criticality. The dashed and solid red lines represent our scaling predicitions for small and large tension. The network size is $N = 2000$.

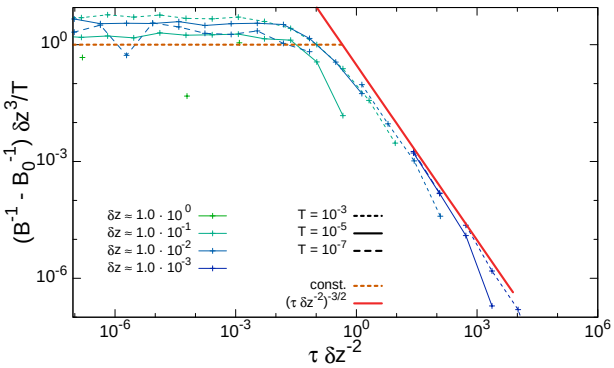

FIG. 2: The scaled difference of inverse bulk modulus at finite temperature and tension, B^{-1}, and at zero temperature and tension modulus, B_0^{-1}, on a double logarithmic scale as a function of scaled tension, $\tau\delta z^{-2}$. Different data sets correspond to different combinations of temperature and δz, as indicated in the legend. The dotted and solid orange line indicates our scaling predictions for small and large tension, respectively. The network size is $N = 2000$.

II. INTUITIVE PICTURE BASED ON A SQUARE LATTICE

We now show that these properties are a consequence of the peculiar vibrational spectrum of jammed networks.

The basic physics can be understood by considering the square lattice in Fig. 4. Despite being ordered, this isostatic lattice shares the vibrational properties of jammed networks at criticality. Soft modes are easily identified as zig-zag modes that can be excited at zero tension without energy cost. There are $O(N^{1/2})$ such zig-zag modes, one for each boundary node. The deformations induced by one of these soft modes are indicated by arrows in the figure.

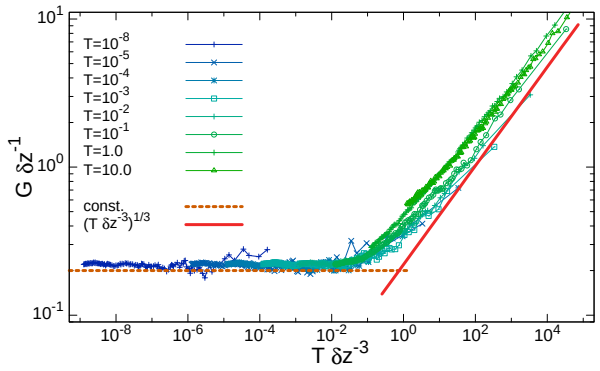

FIG. 3: Scaled shear modulus on a double logarithmic scale as a function of scaled temperature $T\delta z^{-3}$. Different data sets correspond to different combinations of temperature and δz, as indicated in the legend. The coordination δz is ranged from 10^{-2} to 1.99 in 100 log-spaced steps. The dashed and solid lines indicate our scaling predictions for small and large temperature, respectively. The network size is $N = 1600$.

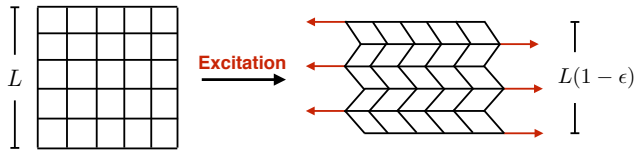

FIG. 4: **Effect of floppy mode fluctuations on a square lattice.** (Left) A square lattice of linear dimension L at vanishing temperature. (Right) The red arrows indicate one of the L^{d-1} soft modes of the lattice. Excitation of this particular soft mode, for instance by thermal fluctuations or shear stress, leads to the contraction of the vertical end-to-end distance. In this state, an external tension will pull out the zigzag undulations rather than stretch individual bonds. These features are generic to jammed networks, which likewise contract upon excitation as we argue based on the known spectrum of soft modes.

Given this cartoon picture of an isostatic network, an important observation can be made that may, at first, seem special to the square lattice: Exciting the said zigzag modes turns straight system-spanning lines into broken lines of shorter end-to-end distance. As a consequence, thermally exciting many such modes leads to a collapse of total network area (volume in 3D).

A finite negative thermal expansion coefficient is obtained when one rigidifies the lattice, which can be done in various ways. Here, we focus on adding δz extra contacts per node and on applying a tension τ. The erstwhile zero-frequency modes now stiffen in a characteristic way. As a consequence, the negative thermal expansion coefficient acquires a finite value that diverges as a powerlaw as δz and the tension vanish. The feature of straight lines reducing their end-to-end distance by thermal undulations is reminiscent of the physics of polymers [22, 23]. Indeed, we will see that the mode spectrum of near isostatic networks and semiflexible polymers has striking similarities.

Pulling the boundaries of the non-excited square lattice, i.e. one applies a tension, obviously stretches the springs in a simple affine way. This explains why the zero-temperature bulk modulus is given by the spring constant. At finite temperature, however, soft modes are excited and the lattice is characterized by jagged lines. An external stretching force now can pull out undulations of these jagged lines rather than affinely stretching bonds. The bulk modulus, thus, softens due to the presence of undulations that can be pulled out. This phenomenon is equivalent to the longitudinal response of semiflexible polymers being finite only at finite temperatures, where undulations exist that can be pulled out by external forces [22, 23].

At fixed area, exciting soft modes requires stretching of bonds. As a consequence, the tension in the networks increases. This increase in tension stabilizes the network by generating a finite shear modulus at isostaticity.

Next we turn these intuitive arguments into scaling arguments to show that, actually, the above behavior is not special to square lattices but characteristic for the harmonic response of jammed packings. Afterwards we will discuss the relation of our results to random networks generated in other ways, for instance by randomly cutting excess bonds.

III. SCALING ARGUMENTS

Our scaling analysis is based on an estimate of how the positional fluctuations of the nodes in the network depend on temperature and tension. These fluctuations can then be used to estimate the negative thermal expansion coefficient and the elastic moduli.

We assume that the spring constant k and rest length a of the springs (at vanishing temperature and pressure) is identical for all springs. Thus, it is convenient to measure lengths in units of a, energies in units of ka^2, the temperature T in units of ka^2/k_B and the tension in units of k. Equivalently, we set $a = 1$ and $k_B = 1$.

Since our goal is to explain the observed scaling laws, we will not keep track of numerical pre-factors of order unity.

A. Mean Square Displacement

We consider a large hyperstatic d-dimensional spring network with co-ordination number $z_c + \delta z$ above the minimal isostatic value $z_c = 2d$ needed for rigidity [20]. Close to the isostatic connectivity number, the vibrational mode spectrum exhibits an anomaly: The density $D(\omega)$ of modes with frequency ω approaches a plateau, called boson peak in the glass community, above a frequency of order $\omega_* \sim \delta z$. This behavior is in stark contrast to highly co-ordinated networks, which exhibit the

scaling $D(\omega) \sim \omega^2$ expected for a continuous elastic solid. The excess low-frequency modes result from extended soft modes that appear if the network is larger than a characteristic length scale $l_* \sim \delta z^{-1}$, the so-called point-to-set correlation length [24].

Importantly, under tension τ, the energy of an excited soft modes is shifted by an amount proportional to the rescaled tension τ: if a soft mode has frequency ω_0 under vanishing tension, its frequency ω_τ at finite tension is given by $\omega_\tau^2 = \omega_0^2 + c_1\tau$ with c_1 being a positive constant of order unity. The dependence of mode frequency on tension follows from the fact that a broken line reduces its end-to-end distance roughly by a fraction $(\delta R/a)^2/2 \sim \delta R^2$ when excited by a typical nodal displacement δR (see, e.g., the soft mode excitation in Fig. 4). For a careful derivation of the mode spectrum, see the argument in Ref. [5] and its revision in Ref. [25].

An excited single mode of frequency ω stores an energy of order $L^d \delta R_\omega^2 \omega^2$ in terms of the mean squared displacement δR_ω^2 (averaged over all nodes). At equilibrium, each mode should store an energy of $k_B T/2$, as dictated by the equipartition theorem. Thus, we expect that each floppy mode generates a mean square displacement of $\langle \delta R_\omega^2 \rangle \sim L^{-d} T \omega_\lambda^{-2}$, with the angled brackets indicating a thermodynamic average. The total mean square displacement $\langle \delta R^2 \rangle$ follows from the integral

$$\langle \delta R^2 \rangle \sim \int_{\omega_*}^{1} d\omega_0 \, \frac{T}{\omega_0^2 + c_1\tau} \tag{1}$$

$$\overset{\omega_* \ll 1}{\sim} \arctan\left[\sqrt{c_1\tau}/\omega_*\right] (c_1\tau)^{-1/2} \tag{2}$$

$$\sim \begin{cases} T\delta z^{-1} & \tau\delta z^{-2} \ll 1 \\ T\tau^{-1/2} & \tau\delta z^{-2} \gg 1 \end{cases} . \tag{3}$$

where we employed the above-mentioned plateau $D(\omega) = \text{const.} \sim O(L^d)$ in the density of states at zero-tension that appears above the frequency $\omega_* \sim \delta z \ll 1$. Note that the resulting scaling predictions are in good agreement with our data collapse in Fig. 5.

B. Contraction

Next we study how the (negative) expansion δA of the network area A depends on temperature and tension. Two contributions have to be considered. The first contribution is the affine expansion of network bonds in response to a finite tension. This zero-temperature contribution is positive but it turns out to be subdominant compared to the soft mode contribution at finite temperature and close to the critical point.

At zero temperature, the bulk modulus is given by $B_0 \sim z$, the average number of springs per node, because most springs are stretched by the same amount in response to a tension. Equivalently the relative extension ϵ_0 is given by $\epsilon_0 = \tau/B_0 \sim \tau/z$ in terms of the tension τ.

Heating induced contraction is caused by the excitation of floppy modes. As we have mentioned above, one excited floppy mode generates a contraction of order $\langle (\delta R_\omega/a)^2/2 \rangle \sim \langle \delta R_\omega^2 \rangle$. Thus, the heating-induced negative extension $\epsilon_T \sim -\langle \delta R^2 \rangle$ scales the same way as the mean square nodal displacements.

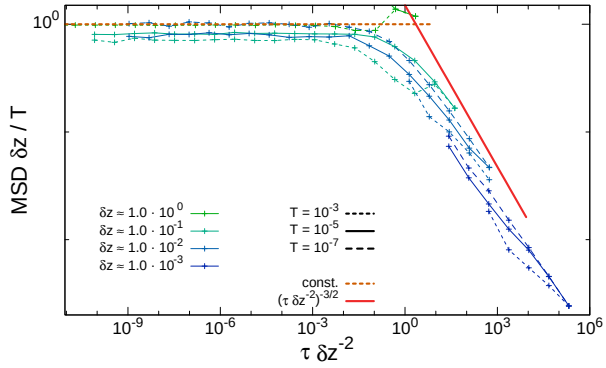

FIG. 5: Nodal fluctuations in spring networks derived from jammed particle packings. The scaled total mean square displacement $\langle \delta R^2 \delta z/T$ is shown as a function of $\tau\delta z^{-2}$ for combinations of two temperatures and three different δz. The asymptotics derived from our scaling arguments are plotted on top of the data.

We expect the total extension to be given by the sum of the both contributions,

$$\epsilon = \epsilon_0 + \epsilon_T \sim \frac{\tau}{z} - c_2 \langle \delta R^2 \rangle . \tag{4}$$

where we introduced another factor c_2 of order unity. The negative thermal expansion coefficient follows from differentiation with respect to temperature,

$$\text{NTE} = -\partial_T \epsilon \sim \begin{cases} \delta z^{-1} & \tau\delta z^2 \ll 1 \\ \tau^{-1/2} & \tau\delta z^2 \gg 1 \end{cases} . \tag{5}$$

These two asymptotic regimes are indeed consistent with our Monte Carlo simulation results in Fig. 1.

The bulk modulus follows from differentiation with respect to the tension, $B^{-1} = \partial_\tau \epsilon$. The temperature induced change is

$$B^{-1} - B_a^{-1} \sim -\partial_\tau \langle \delta R^2 \rangle \sim \begin{cases} T\delta z^{-3} & \tau\delta z^{-2} \ll 1 \\ T\tau^{-3/2} & \tau\delta z^{-2} \gg 1 \end{cases} . \tag{6}$$

We have thus obtained a scaling-level explanation for the observed cusp in the bulk modulus (Fig. 2).

C. Shear modulus and fixed volume ensemble

The zero-temperature shear modulus of jammed systems is soft close to isostaticity, as has been extensively studied in the literature [5, 9]. The characteristic shear modulus

$$G \sim \begin{cases} \delta z & \tau\delta z^{-2} \ll 1 \\ \tau^{1/2} & \tau\delta z^{-2} \gg 1 \end{cases} \tag{7}$$

is a direct consequence of the above spectrum and density of soft modes.

If the volume is kept fixed, temperature will generate a nonzero tension, which may stiffen the shear modulus. The tension generated can be estimated by setting $\epsilon = 0$ (or more generally to a given pre-described value) in (4),

$$\tau_{\epsilon=0} \sim \begin{cases} T\delta z^{-1} & T \ll \delta z^3 \\ T^{2/3} & T \gg \delta z^3 \end{cases} . \quad (8)$$

Inserting this scaling behavior for the tension into (7), we get

$$G \sim \begin{cases} \delta z & T \ll \delta z^3 \\ T^{1/3} & T \gg \delta z^3 \end{cases} . \quad (9)$$

We thus obtain the scaling $G \sim T^{1/3}$ in the critical regime, which is in agreement with simulations (Fig. 4).

IV. DISCUSSION

We have shown that the elastic properties of near-isostatic spring networks derived from jammed packings of elastic spheres exhibit critical behavior upon heating. At fixed finite tension, the negative thermal expansion coefficient (NTE) diverges as $\tau^{-1/2}$ at isostaticity ($\delta z = 0$). At vanishing tension, networks shrink with a NTE diverging as δz^{-1}. Concomitantly, the bulk modulus exhibits a cusp at criticality.

At fixed volume, the tendency to shrink leads to a rapid build-up of tension. As a consequence, the shear modulus at criticality stiffens with temperature as $G \propto T^{1/3}$. Our arguments predict that the region of stability is wider in heated networks, as illustrated in the phase diagram of Fig. 6: As the temperature increases tension builds up to stabilize the network down to $-\delta z \sim T^{1/3}$.

It is useful to compare our results with those of Dennison et al. [10], in which spring networks at fixed volume close to isostaticity were also studied. The authors found that the shear modulus at criticality increases with temperature as $G \sim T^{1/2}$. This intriguing result is in contrast with our setup, which as mentioned exhibits $G \sim T^{1/3}$.

This discrepancy indicates that the elastic properties of spring networks not only depend on the co-ordination number but also on the way that networks are prepared, as has been previously observed at zero temperature [8]. Dennison et al. started with highly co-ordinated networks and gradually removed springs until a given coordination number was achieved. These randomly diluted networks, which we call "pruned", are in contrast to our networks, which were generated at given co-ordination numbers directly from simulated particle packings. The comparison of the bulk moduli in both network types in Fig. 7 shows that pruned networks exhibit a soft bulk modulus for all temperatures in contrast to jammed networks, which have a bulk modulus of order the spring constant at vanishing temperature.

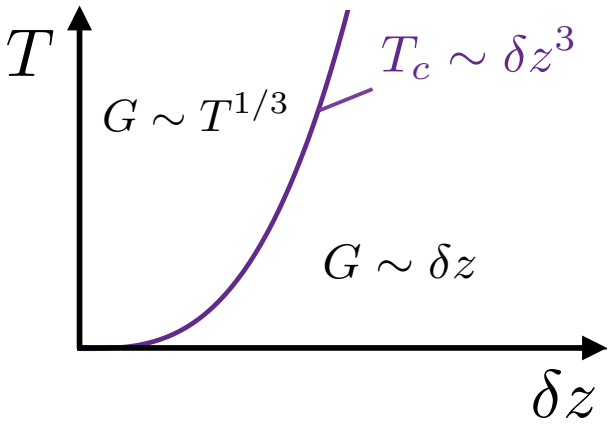

FIG. 6: **Effect of thermal fluctuations on the shear modulus at fixed volume.** In this schematic phase diagram of the shear modulus, we indicate the phase boundary $T_c \sim |\delta z|^3$ separating the zero-temperature behavior $G \sim \delta z$ from a thermally-stabilized regime $G \sim T^{1/3}$. The latter regime widens rapidly as temperatures are increased.

The structural differences of the different network types must also be accommodated in the analytical scaling picture. Our above arguments rely on the assumption that the pressure couples to the soft modes *only* via a change in frequency, which is well-established for jammed packings. However, for other network preparations, the pressure might also couple linearly to the soft modes (like a shear stress). This will lead to a softening of the bulk modulus, as can be accommodated by introducing an appropriate coupling term in our scaling arguments. A similar effect appears in polymer physics, where it may be easier to apprehend: The linear response of a semiflexible polymer to a longitudinal stretching force is quite stiff, unless the force exhibits a (possibly small) transverse component upon which the response is dominated by the soft bending modes.

Finally, we hypothesize that heating in close-to-isostatic networks is somewhat analogous to shearing the network by a shear angle $\alpha \sim T^{1/2}$: In other words, we are suggesting that all available floppy modes are excited by roughly the same amount of energy, proportional to α^2. This shear-equipartition assumption for soft modes is consistent with existing scaling analyses, in particular Ref. [7]. Moreover, the so far unexplained temperature scaling $G \sim T^{1/2}$ for pruned networks then becomes simply a consequence of the scaling $G \sim \alpha$ for isostatic pruned networks at zero tension [7]. Extended to the shearing of *jammed*, rather than pruned, isostatic networks, we moreover recover the scaling of the negative shear dilatancy observed for fixed pressure in Ref. [26]. At finite volume, on the other hand, we predict that shearing jammed networks will lead to a sharp

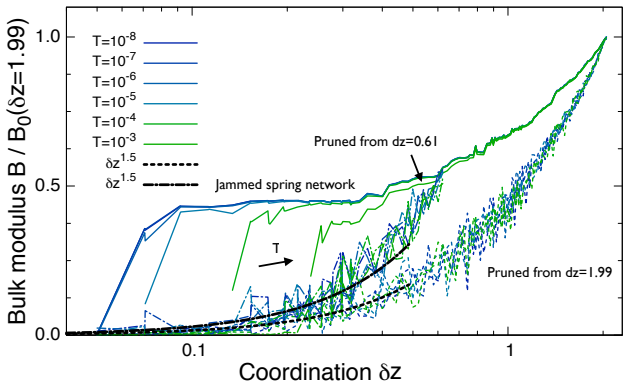

FIG. 7: **Effect of structure on bulk modulus. Bulk modulus for networks from packings(solid line) and their pruned counterparts where pruning started from $\delta z = 1.99$(dashed line) and $\delta z = 0.61$(solid and dotted line).** The bulk modulus softens with T for the networks from packings while it is temperature-independent for the pruned networks. The upper bound of the bulk modulus is given by the networks from jammed packings when considered as a variational problem. We see that the pruned networks do not couple to the tension like the networks from jammed packings do. The bulk modulus is compared to its value at zero-temperature far away from the isostatic point $\delta z = 1.99$. Note that the bulk modulus of pruned networks is well-described by a scaling $\propto \delta z^{1.5}$. The same scaling behavior was observed for shear moduli in the pruned networks of Ref. [7] close to isostaticity. In contrast, the networks for jammed packings maintain their bulk modulus approaching the isostatic point unless they get softened by temperature. In the limit of zero temperature the networks obtained from jammed packings have a large bulk modulus while the one of pruned networks vanishes, also see [8]. The wiggly data for pruned networks stems from every δz data point being pruned independently. System size is $N = 100$.

rise $G \sim \alpha^{2/3}$ of shear modulus with shear angle, contrasting with the linear rise in pruned networks [7].

Ultimately, the response of our thermalized networks to tension (pressures) results from the phonon spectrum being dominated by many soft modes with frequencies that sensitively depend on tension. Similar mode spectra arise in (semiflexible) biopolymer networks, which therefore also exhibit negative thermal expansion coefficients and shear dilatancy [27–29].

Our study focused on spring networks without the possibility of change in connectivity, in particular by bond breaking. A jammed random close pack of elastic spheres simply has no space to contract. Hence, heating of such packings without free space must immediately lead to bond breaking to ensure a mode spectrum consistent with the density constraint. Contraction as observed in our simulation may occur if a packing is over-coordinated (i.e. due to compression) or exhibits enough free volume and some degree of attractive interactions. We expect that the mean square displacement is the quantity most robustly observed in packings, as it does not rely on the precise coupling of external forces and local tension.

Acknowledgments

We would like to thank Matthieu Wyart for helpful discussions. Research reported in this publication was supported by the National Institute Of General Medical Sciences of the National Institutes of Health under Award Number R01GM115851, by a Simons Investigator award from the Simons Foundation (O.H.) and by the German Research Foundation (DFG) in the framework of the SFB 937/A15 (OH, CB), SFB 937/A16 (CH, CB) and of the Emmy Noether Program He 6322/1-1 (CH, CB). The content is solely the responsibility of the authors and does not necessarily represent the official views of the National Institutes of Health.

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