Thermalized connectivity networks of jammed packings

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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 the consequences of bond breaking and an apparent analogy between thermalization and shear.

Packings of repulsive elastic particles have emerged as a rich model granular system with potential relevance to amorphous solids [1, 2]. A number of remarkable features emerge, in particular, when the packing is close to the onset of rigidity. One of the key characteristics of jammed packings is an excess of low-frequency vibrations [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 materials 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 [5]. 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 Hookean springs. The effect of thermal fluctuations is studied using Metropolis Monte Carlo simulations. Due to the small temperatures considered, crossing of bonds is very unlikely, although it is not penalized in our simulations. Also, contacts do neither break nor form in the course of the simulation. Consequences of bond breaking are discussed below.

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 expan-
sion coefficient, NTE, exhibits a singularity as the average contact number per particle approaches isostaticity. For vanishing tension \( \tau \), 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 \( T, \delta z \) and \( \tau \) collapse onto a master curve when we plot NTE \( \delta z \) vs. \( \tau \delta z^{-2} \). The deviations suggest that the collapse for larger tensions only works for small enough \( \delta z \).

Like jammed packings, our spring networks are affinely deformed when compressed at zero temperature. In other words, compressive deformations merely change contact lengths but not contact angles. Consequently, the bulk modulus \( B_0 \) of zero-temperature jammed networks is set simply by the spring constant and the 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.

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
FIG. 4: Effect of floppy mode fluctuations on a square lattice. (Left) A square lattice of linear dimension $L$ at vanishing temperature. The red arrows indicate one of the $L^{d-1}$ soft modes of the lattice. (Right) 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 zig-zag 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 mode spectrum.

Next we turn these intuitive arguments into scaling arguments to show that, actually, the above behavior is not special for 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]. Within such a network, there are of order $\lambda^{d-1}$ springs at the boundary of a region of linear dimension $\lambda$. Cutting those boundary springs removes $\lambda^{d-1}$ constraints. If this number of relieved constraints is larger than the excess number $\delta z \lambda^d$ of constraints (i.e. springs) we can expect floppy modes in the considered region. Thus, floppy modes extend up to a characteristic length scale $\lambda_{\text{max}} \sim \delta z^{-1}$ [5].

In order to excite a floppy mode of extension $\lambda$ such that transverse displacements of order $u$ are generated one has to supply energy of roughly $e_u \sim \lambda^d \omega^2 u^2$ in terms of the mode frequency

$$\omega^2 = \lambda^{-2} + c_1 \tau$$

of a floppy mode of extension $\lambda$ [5] ($c_1$ is a positive constant of order unity). For vanishing tension, $\tau = 0$, the above energy arises because $\lambda^d$ springs are each deformed by a strain of order $u/\lambda$. The dependence on the tension follows from the fact that a broken line reduces its end-to-end distance roughly by a fraction $(u/a)^2/2 \sim u^2$ when excited by a transverse displacement of characteristic wave length $a = 1$, as is typical for soft modes (see, e.g., the soft mode excitation in Fig. [4]). For a careful derivation of the mode spectrum, see Ref. [5].

At equilibrium, each mode should store an energy of $k_B T/2$ by the equipartition theorem. Thus, we expect
that each floppy mode acquires a mean square amplitude of $\langle u^2 \rangle \sim T\lambda^{-4}\omega_\lambda^{-2}$. The total mean square displacement $\langle \delta R^2 \rangle$ of a node in the network can be expected to be dominated by the transverse (non-affine) displacements caused by the excitation of soft modes. To estimate the resulting fluctuations, we need to sum up the mean square displacements generated by all orthogonal floppy modes that have overlap with a given focal node.

Since there are $\lambda^{d-1} \sim \int_{1}^{\lambda_{max}} D(\lambda) d\lambda$ floppy modes of extension $\lambda$ overlapping with a given node, we have a density $D(\lambda) \sim \lambda^{d-2}$ of floppy modes per unit “wave length”. The mean square displacement of the focal node is thus given by the integral

$$\langle \delta R^2 \rangle \sim \int_{1}^{\lambda_{max}} d\lambda D(\lambda) \frac{T}{\lambda^{d}\omega_\lambda^2 (\int_{1}^{\lambda_{max}} D(\lambda) d\lambda)^{-1}} \sim T \int_{1}^{\lambda_{max}} d\lambda (1 + c_1 \tau \lambda^2)^{-1} \sim \arctan \sqrt{c_1 \tau \lambda_{max}} (c_1 \tau)^{-1/2} \sim \begin{cases} T\delta z^{-1} & \tau\delta z^{-2} \ll 1 \\ T\tau^{-1/2} & \tau\delta z^{-2} \gg 1 \end{cases}$$

where we used $\lambda_{max} \sim \delta z^{-1}$ in going to the last line. Note that these predictions are in agreement with our data collapse in Fig. 5.

**B. Contraction**

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

Since in an affine deformation all springs are stretched equally, we can estimate the affine (“stiff”) contribution to the bulk modulus simply by $B_a \sim z$, the average number of springs per node. Equivalently the affine relative extension $\epsilon_a$ is given by $\epsilon_a = \tau/B_a \sim \tau/z$ in terms of the tension $\tau$.

The non-affine contraction is caused by the excitation of floppy modes. As we have mentioned above, one excited floppy mode generates a contraction of order $\langle (u/a)^2 \rangle \sim \langle u^2 \rangle$. Thus, the non-affine contraction $-\epsilon_{na} \sim \langle \delta R^2 \rangle$ scales the same way as the mean square nodal displacements.

We expect the total extension to be given by the sum of the affine and non-affine contributions,

$$\epsilon = \epsilon_a + \epsilon_{na} \sim \frac{\tau}{z} - c_2 \langle \delta R^2 \rangle .$$

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

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

The NTE is shown in Fig. 1 and exhibits the two asymptotic scalings predicted by the above expression.

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

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

The predicted cusp in the bulk modulus is reproduced in simulations, as shown in 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

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

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 (6),

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

Inserting this scaling behavior for the tension into (9), we get

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

FIG. 5: Scaled total mean square displacements as a function of $\delta z^{-2}$ for combinations of two temperatures and three different $\delta z$. The asymptotics derived from our scaling arguments are plotted on top of the data.
We thus obtain the scaling $G \sim T^{1/3}$ in the critical regime, as was shown in 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 $\delta z^{-1}$. 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. 4. 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 co-ordination 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.

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 assuming that all available floppy modes are excited by roughly the same amount of energy, propor-
tional to $a^2$. 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. [22]. At finite volume, on the other hand, we predict that shearing jammed networks will lead to a sharp 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 [23–26].

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. Compression 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 suspect 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.

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