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LETTER TO THE EDITOR

Node coordination constraints and the elasticity of random central force networks

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Abstract. Random central force networks are constructed by relaxation of a bond-depleted lattice of Hooke's law springs under tension. The bond depletion procedure involves the random removal of the bonds connecting the nodes of a simple cubic lattice subject to constraints on the coordination of individual nodes. In one model, the random network, random bond depletion is only restricted by the requirement that no node have coordination greater than fourfold. In a second model, the continuous random network, random bond depletion is subject to the additional constraint that the individual node coordination remains at least twofold, thus precluding the formation of any dangling bonds. The elastic properties of the two types of network markedly differ only for small values of the mean node coordination, when the random network contains significantly higher fractions of threefold- and fourfold-coordinated nodes than the continuous random network.

The elastic properties of random networks of Hooke's law springs under a tension have been investigated by Tang and Thorpe [1, 2]. However, the networks generated by the random bond depletion of lattices may not represent appropriate structural models of some physical systems, since the process of random bond depletion does not place any constraint on the coordination of individual nodes in the network. Such constraints are found in a continuous random network model of a glass. He and Thorpe [3] generated such a network from the random bond depletion of a diamond lattice subject to the constraint that all nodes should be two, three or fourfold coordinated. Another system in which physical constraints might be expected to restrict the node coordination within a network model is a gel. Grimson [4] introduced a random central force network model of a gel in which an initial phase of random bond depletion proceeds as long as the bond designated for removal connects nodes at least one of which has coordination greater than fourfold. Any subsequent random bond removal is unrestricted. This model differs from that of He and Thorpe in that it allows for dangling bonds and disconnected nodes. In this letter the two models are compared as a function of the mean node coordination of the network.

An elastic network is constructed from a lattice where all the bonds between nearest-neighbour sites are Hooke's law springs. The sites of the initial lattice define the nodes of the elastic network. The Hooke's law spring connecting nodes i and j,

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located at $\boldsymbol{R}_i$ and $\boldsymbol{R}_j$ respectively, is characterized by a natural (unstretched) length L_0 and force constant K. The elastic potential energy for this model is given by [4]

$$E = \tfrac{1}{2}\sum_{i<j}{}' K_{ij}\left(|\boldsymbol{R}_i - \boldsymbol{R}_j| - L_0\right)^2 \tag{1}$$

where the prime denotes that the summation is only over nearest-neighbour bonds. $K_{ij} = K$ if the bond is present and $K_{ij} = 0$ if it is absent. The force $\boldsymbol{F}_i$ on the ith node of the network is given by

$$\boldsymbol{F}_i = -\partial E/\partial \boldsymbol{R}_i = \sum_j{}' K_{ij}\left(|\boldsymbol{R}_i - \boldsymbol{R}_j| - L_0\right)\mathcal{R}_{ij} \tag{2}$$

where

$$\mathcal{R}_{ij} = (\boldsymbol{R}_j - \boldsymbol{R}_i)/|\boldsymbol{R}_i - \boldsymbol{R}_j| \tag{3}$$

is the unit vector between nodes i and j. The equilibrium condition for the network is that the total force acting on each node of the network must vanish, i.e.

$$\boldsymbol{F}_i = -\partial E/\partial \boldsymbol{R}_i = 0 \tag{4}$$

for all i. For an undepleted network the sites of the lattice will correspond to equilibrium positions of the network nodes. However, following site or bond depletion of the lattice, if the natural length of the springs differs from the lattice spacing the force balance is destroyed and the network must deform so as to move the nodes of the network to their equilibrium positions. The equilibrium positions of the network nodes may be far away from the corresponding original lattice sites and a certain amount of static energy is stored in this relaxed network. The elastic properties of the network must be determined from the equilibrium structure. The networks are characterized by a mean node coordination r defined by

$$r = (1/N_n)\sum_i r_i = (1/N_n)\sum_{i,j}{}'(K_{ij}/K) \tag{5}$$

where N_n is the number of nodes in the simulation cell and r_i is the coordination of the ith node.

A bond-depleted lattice is first generated by randomly removing bonds from a simple cubic lattice subject to the constraint that the randomly selected bond is only removed if at least one of the nodes at either end of the bond has more than fourfold coordination. Bond depletion proceeds until $r_i \leqslant 4$ for all i. From [4] it is known that a value for the mean node coordination of $\langle r\rangle \simeq 3.5$ is obtained from this procedure. Further unrestricted random bond removal then occurs to achieve the required mean node coordination. This type of network is termed a random network and does not preclude the presence of dangling bonds or disconnected nodes. These features are not allowed in a continuous random network which is achieved by the same bond-depletion procedure described above with the additional constraint that the randomly selected bond is only removed if following removal the coordination of all nodes remains at least twofold.

The bond-depleted network is relaxed to equilibrium by solving purely dissipative equations of motion for the nodes given by [5]

$$\eta \, \mathrm{d}\boldsymbol{R}_i/\mathrm{d}t = -\partial E/\partial \boldsymbol{R}_i \tag{6}$$

for all i, where η is a friction coefficient. The coupled equations of motion for the nodes of equations (2) and (6) are solved numerically using the Euler method. Specifically, during the iteration procedure, the position of node i is determined from the previous configuration by

$$\boldsymbol{r}_i^{(n+1)} = \boldsymbol{r}_i^{(n)} + \alpha \boldsymbol{F}_i^{(n)} \tag{7}$$

where the parameter $\alpha = \Delta t/\eta$ (or the time step Δt) is adjusted to ensure the convergence of the iteration. In this work α was typically chosen to be in the range $0.1 < \alpha < 0.3$. The iteration process should stop when the force on every node of the network is zero. However, in practice, an appropriate small value for the force on any node, $|F_{\mathrm{m}}|$, is chosen that is good enough to give the final precision required. Thus the relaxation procedure is terminated when $|\boldsymbol{F}_i| < |F_{\mathrm{m}}|$ for all i. The elastic energy of the relaxed network is denoted by $E_0(\{K_{ij}\}, L_0)$.

Tang and Thorpe [1,2] have shown that for depleted networks of stretched springs, the elastic properties, within linear elasticity theory, are fully determined by the four independent quantities E_0, T, B and b. T is the tension, B the bulk modulus and b the shear modulus. The network tension may be determined by constructing an imaginary plane through the sample and calculating the force per unit area perpendicular to the plane produced by the bonds that cut the plane. This allows the tension to be calculated from the equilibrium structure as well as from the numerical derivative of the deformation energy used to calculate B and b [2,4].

Periodic boundary conditions were employed to maintain the network tension [1,2] and the nearest-neighbour lattice spacing defines the unit of length. The force constant of the springs sets the energy scale and a maximum force on any node of $|F_{\mathrm{m}}|/K = 10^{-3}$ was used to terminate the relaxation procedure. This typically corresponded to an accuracy in the elastic energy per unit volume E_0/K for the equilibrium network of order 10^{-8}. Such a choice leads to errors in the calculated elastic properties of networks that are less than the statistical uncertainty. The results presented here were obtained by bond depletion of $12 \times 12 \times 12$ simple cubic lattices with periodic boundary conditions averaged over ten independent runs with random initial configurations. This gives statistical errors of less than 1% in the mean elastic energy and tension of the networks. Note that as a result of starting from a simple cubic lattice of size $12 \times 12 \times 12$ it is difficult to achieve values of $\langle r \rangle < 2.15$.

All four independent elastic quantities show the same qualitative behaviour for both random and continuous random networks. For all values of the natural spring length, the elastic properties of the networks are monotonic decreasing functions of the mean node coordination. The mean values for the energy $\langle E_0 \rangle$ and tension $\langle T \rangle$ as a function of $\langle r \rangle$ in relaxed random and continuous random networks are shown in figures 1 and 2 for $L_0 = 0$ and $L_0 = 0.5$ respectively.

When $L_0 = 0$ it can be seen that with the continuous random network $\langle E_0 \rangle$ and $\langle T \rangle$ are linear functions of $\langle r \rangle$ over the range $2.15 \leqslant \langle r \rangle \leqslant 3.5$ which by extrapolation would vanish for $\langle r \rangle \simeq 2.05$. The corresponding results for the random network show little qualitative difference when $\langle r \rangle > 3$. But for $\langle r \rangle < 3$, $\langle E_0 \rangle$ and $\langle T \rangle$ for the

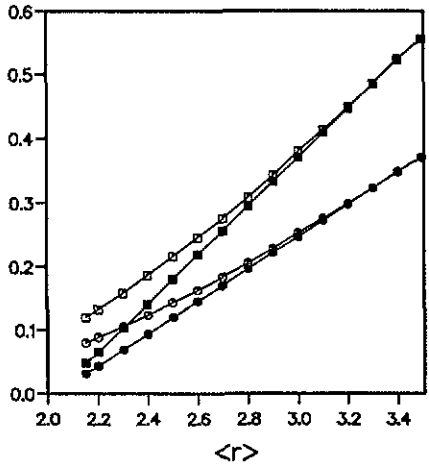

Figure 1. Elastic energy per unit volume $\langle E_0/K\rangle$ (■) and tension $\langle T/K\rangle$ (●) as a function of the mean node coordination $\langle r\rangle$ for continuous random networks ($2 \leqslant r_i \leqslant 4$ for all i) with $L_0 = 0$. Results for random networks ($r_i \leqslant 4$ for all i) are shown by the corresponding open symbols.

random network both show significant curvature as a function of $\langle r\rangle$. This results in significant quantitative differences between the random and continuous random networks for small values of the mean node coordination. An extrapolated value for the mean node coordination corresponding to vanishing elastic properties of the random network would be much less than two, a value considerably smaller than that for the continuous random network.

For $L_0 = 0.5$ figure 2 shows that for the continuous random network $\langle E_0\rangle$ and $\langle T\rangle$ are now only linear functions of $\langle r\rangle$ for $\langle r\rangle > 3$ and vanish for $\langle r\rangle \leqslant 2.2$. Once more, significant quantitative differences in the elastic properties between the random and continuous random networks only occur for $\langle r\rangle < 3$. The extrapolated value of $\langle r\rangle$ at which the elastic properties of the random network would vanish must clearly lie below two, but should be greater than when $L_0 = 0$. Thus while the natural spring length plays an important role in determining the absolute magnitude of the elastic properties, the differences between random and continuous random networks do not appear to depend significantly on L_0 and must result from the structural differences between the two network types.

Figure 3 shows the node coordination distributions as a function of the mean node coordination resulting from the two random bond-depletion procedures. For $\langle r\rangle \simeq 3.5$ the fraction of twofold-, threefold- and fourfold-coordinated nodes in the two types of network are almost the same, with small differences arising from the presence of a few singly coordinated and disconnected nodes (not shown) in the random network. As the mean node coordination is reduced, the two types of network show monotonic decreases in the fraction of fourfold-coordinated nodes, monotonic increases in the fraction of twofold-coordinated nodes and a maximum in the fraction of threefold-coordiated nodes for $\langle r\rangle \simeq 3$. The most visually striking difference between the two network types is in the fractions of twofold-coordinated nodes, but this is simply a result of precluding dangling bonds from the continuous random network. The most important difference in the node coordination distribution between the two types of

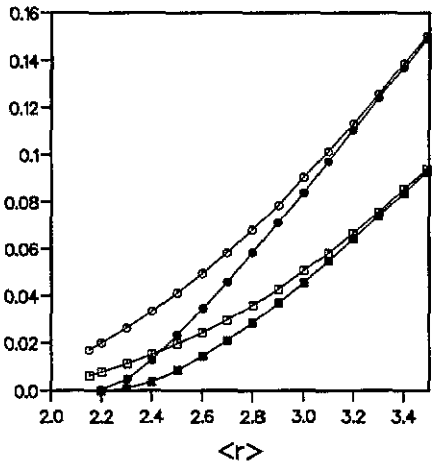

Figure 2. Elastic energy per unit volume $\langle E_0/K\rangle$ (■) and tension $\langle T/K\rangle$ (●) as a function of the mean node coordination $\langle r\rangle$ for continuous random networks ($2 \leqslant r_i \leqslant 4$ for all i) with $L_0 = 0.5$. Results for random networks ($r_i \leqslant 4$ for all i) are shown by the corresponding open symbols.

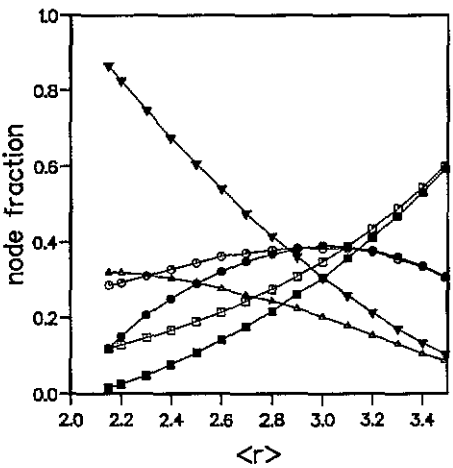

Figure 3. Node coordination distributions as a function of the mean node coordination $\langle r\rangle$. For continuous random networks ($2 \leqslant r_i \leqslant 4$ for all i) the node fractions are shown for coordinations of $r_i = 4$ (■), $r_i = 3$ (●) and $r_i = 2$ (▼), while for random networks ($r_i \leqslant 4$ for all i) the node fractions are shown by the corresponding open symbols.

network is the presence of significantly higher fractions of threefold- and fourfold-coordinated nodes in the random network for $\langle r\rangle < 2.8$.

Thus is can be concluded that the elastic properties of bond-depleted networks of springs at small values of the mean node coordination can show substantial differences as a result of controlling the node coordination distribution, in this case by allowing or not the formation of dangling bonds. Significant quantitative differences between the elastic properties of random and continuous random networks occur when the fraction of threefold-coordinated nodes differ markedly and it is the presence of significantly

higher fractions of threefold- and fourfold-coordinated nodes in the random network that give it a much stronger structure than the continuous random network at small values of the mean node coordination.

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