Tuning by pruning: exploiting disorder for global response and the principle of bond-level independence

Carl P. Goodrich* and Andrea J. Liu
Department of Physics, University of Pennsylvania, Philadelphia, Pennsylvania 19104, USA

Sidney R. Nagel
James Franck Institute, The University of Chicago, Chicago, Illinois 60637, USA

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We exploit the intrinsic difference between disordered and crystalline solids to create systems with unusual and exquisitely tuned mechanical properties. To demonstrate the power of this approach, we design materials that are either virtually incompressible or completely auxetic. Disordered networks can be efficiently driven to these extreme limits by removing a very small fraction of bonds via a selected-bond removal procedure that is both simple and experimentally relevant. The procedure relies on the nearly complete absence of any correlation between the contributions of an individual bond to different elastic moduli. A new principle unique to disordered solids underlies this lack of correlation: independence of bond-level response.

The properties of amorphous solids are essentially and qualitatively different from those of simple crystals [1]. In a crystal, identical unit cells are interminably and symmetrically repeated, ensuring that all cells make identical contributions to the solid’s global response to an external perturbation [2, 3]. Unless a crystal’s unit cell is very complicated, all particles or inter-particle bonds contribute nearly equally to any global quantity, so that each bond plays a similar role in determining the physical properties of the solid. For example, removing a bond in an ordered array or network decreases the overall elastic strength of the system, but in such a way that the resistance to shear and the resistance to compression drop in tandem [4] so that their ratio is nearly unaffected. Disordered materials are not similarly constrained. We will show that as a consequence, one can exploit disorder to achieve a unique, varied, textured and tunable global response.

A tunable global response is a corollary to a new principle that emerges for disordered matter: independence of bond-level response. This independence refers not only to the dearth of strong correlations between the response of different bonds, but also, and more importantly, to the response of any specific bond to different external perturbations. We will demonstrate this by constructing selected-bond-removal networks, where individual bonds, or springs, are successively removed to drive the overall system into different regimes of behavior, characterized by ratios of different mechanical responses. Starting from the same initial network, we can remove as few as 2% of any specific bond to different external perturbations. We work in either two or three dimensions and start with a packing fraction, \( \phi \), that is above the jamming density. After minimizing the energy of a configuration, we create a network by replacing each pair of interacting particles with an unstretched spring of unit stiffness between nodes at the particle centers [11]. We characterize the network by the excess coordination number \( \Delta Z = Z - Z_{\text{iso}} \), where \( Z \) is the average number of bonds at each node and \( Z_{\text{iso}} = 2d - 2d/N \) is the minimum for a system to maintain rigidity in \( d \) dimensions [12].

For each network, we use linear response to calculate the contribution \( B_i \) of each bond \( i \) to the bulk modulus, \( B = \sum_i B_i \) (see Appendix for details). The distribution of \( B_i \) in three dimensions is shown in blue in Fig. 1, where data are averaged over 500 configurations, each with approximately 4000 nodes and an initial excess coordination number \( \Delta Z_{\text{initial}} \approx 0.127 \) (corresponding to a total number of bonds that is about 2% above the minimum needed for rigidity).

Similarly, we can start with the same initial network and calculate \( G_i \), the contribution of each bond to the angle-averaged shear modulus, \( G = \sum_i G_i \). (A finite system is not completely isotropic, so the shear modulus varies with direction [13]; we calculate the angle-averaged shear modulus, which approaches the isotropic shear modulus in the infinite system size limit [14].) The resulting distribution for \( G_i \) is shown in purple in Fig. 1.

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* cpgoodri@sas.upenn.edu
Note that the distributions of the bond contributions to $B$ and $G$ are continuous, very broad, and non-zero in the limit $B_i, G_i \to 0$. That is, some bonds have nearly zero contribution to the bulk or shear modulus while others contribute disproportionately. For both $B$ and $G$, the distribution decays as a power law at low values of $B_i$ or $G_i$. These power laws are terminated above $\langle B_i \rangle$ and $\langle G_i \rangle$ by approximately exponential cut-offs. In comparison, the distributions for a perfect crystal would be composed of discrete delta functions.

We next ask if there is a correlation between how an individual bond responds to shear and how it responds to compression. Do bonds with a large contribution to the bulk modulus also have a proportionately large contribution to the shear modulus? Fig. 2a shows the joint probability distribution $P(B_i, G_i)$. There is a nearly vanishing (but not identically zero) correlation between how individual bonds respond to shear and how they respond to compression. This is qualitatively different from what one would find for a simple crystal. Thus, Fig. 2a illustrates a previously-unrecognized property that is very well obeyed by disordered networks: independence of bond-level response.

This new property suggests that one can tailor the behavior of the network by selectively removing (pruning) those bonds that contribute more or less than the average to one of the moduli. By so doing, one can decrease one modulus with respect to the other.

First, we consider the known case of rigidity percolation [4, 15, 16], where a bond is picked at random and removed. This pruning is repeated until the system becomes unstable at $\Delta Z = 0$. We have implemented a slight variation to this procedure: at each step, a bond is removed only if each node connected to this bond has at least $d + 1$ remaining bonds in $d$ dimensions. This is the condition for local stability of a particle in the original jammed packing [17]. As the excess coordination number decreases, the bulk and shear moduli vanish together, so that $G \sim B \sim \Delta Z$ [4, 15, 16] (see Fig. 2b). Therefore, as shown in Fig. 3, the ratio $G/B$ is independent of $\Delta Z$.

We now implement the idea of selected-bond removal in a variety of ways. First we remove the bond with the smallest $B_i$, namely the weakest contribution to the bulk modulus (provided, as above, that each node connected to this bond has at least $d + 1$ remaining bonds). Since the distribution $P(B_i)$ is continuous and nonzero as $B_i \to 0$, the bond removal has almost no effect on the bulk modulus. However, since there is little correlation between the contribution of each bond to the bulk and shear moduli, there is a much larger effect on the shear modulus. The contributions $B_i$ and $G_i$ of the re-
remaining bonds to the moduli are then recalculated and the procedure is repeated to remove the bond with the smallest $B_i$. Figure 2b shows that when bonds with the smallest $B_i$ are successively removed, the shear modulus linearly proportional to $\Delta Z$. Furthermore, it is quantitatively identical, within numerical precision, to when bonds are removed at random. The ability to alter the scaling of the bulk modulus without affecting the scaling of the shear modulus is a clear demonstration that the principle of independence of bond-level response allows for very precise tuning of global properties.

Since removing bonds with the smallest $B_i$ has little effect on the bulk modulus, we would expect $G/B \to 0$ as $\Delta Z \to 0$. As shown in Fig. 3, we find that $G/B \sim \Delta Z^{\mu_{B_+}}$, with $\mu_{B_+} = 1.01 \pm 0.01$. This behavior is identical to the scaling found in the original jammed sphere packings, where $\Delta Z$ is lowered by decompressing the system. In decompressing a jammed packing, this suggests that the contacts most likely to disappear are those which contribute minimally to the bulk modulus, providing theoretical insight into why jamming has anomalous $G/B$ behavior.

We can drive the same initial network to the opposite limit, $G/B \to \infty$, by successively removing bonds with the largest contribution to $B$. As before, independence of bond-level response predicts that the shear modulus will again decrease linearly with $\Delta Z$, as we indeed find (see Fig. 2b). However, the bulk modulus will decrease more quickly, as prescribed by the high $B_i$ tail of the distribution, suggesting that the ratio $G/B$ should increase. The result of this successive bond-removal algorithm is shown by the blue squares in Fig. 3. We find that $G/B \sim \Delta Z^{\mu_{B_+}}$, where $\mu_{B_+} = -7.96 \pm 0.01$. Thus, the increase in $G/B$ occurs with a much steeper power law than the decrease of $G/B$ when the bond with the smallest contribution to $B$ is removed. This power law implies that the distribution $P(B_i/\langle B_i \rangle)$ evolves as bond pruning proceeds.

The algorithms mentioned above can be extended in a number of ways. For example, one can remove the bond with the largest contribution to the shear modulus to drive $G/B$ towards zero. In this case, independence of bond-level response implies that the bulk modulus would respond as if bonds were removed randomly, so that $B \sim \Delta Z$ (see Fig. 2b). However, the shear modulus decreases more rapidly; we find $G/B \sim \Delta Z^{\mu_{G+}}$, where $\mu_{G_+} = 1.82 \pm 0.01$ (purple diamonds in Fig. 3).

We can also tune two-dimensional networks with equal

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**FIG. 3.** Tuning global response in three dimensions. The ratio of shear to bulk modulus, $G/B$, for four pruning algorithms. Error bars (included) are smaller than the symbols. Lines are fits to the data over the indicated range and have slopes, from top to bottom, of -7.96, -0.01, 1.01, and 1.82. Starting with the same initial conditions, we can tune global response by 16 orders of magnitude by pruning of order 2% of the bonds.

**FIG. 4.** Tuning global response in two dimensions. The ratio of shear to bulk modulus, $G/B$, for four pruning algorithms. Error bars (included) are smaller than the symbols. Lines are fits to the data over the indicated range and have slopes, from top to bottom, of -5.36, -0.26, 1.27, and 3.05. Starting with the same initial conditions, we can tune global response by 17 orders of magnitude by pruning of order 1% of the bonds.
ease. We construct spring networks in two dimensions with approximately 8000 nodes and an initial coordination number of $\Delta Z_{\text{initial}} \approx 0.047$, which is about 1% above the minimum needed for rigidity. Figure 4 shows $G/B$ as bonds are pruned towards $\Delta Z \to 0$ for the same four selected-bond removal algorithms as in Fig. 3. When bonds with the smallest $B_i$ are removed, we find that $G/B \sim \Delta Z^{\mu_{B-}}$ with $\mu_{B-} = 1.27 \pm 0.01$. This is close to the behavior known for jammed packings ($G/B \sim \Delta Z^2$), though it is certainly not as clean as in three dimensions. When we prune bonds that resist compression the most (largest $B_i$), we find that $G/B \sim \Delta Z^{\mu_{B+}}$, where $\mu_{B+} = -5.36 \pm 0.01$. At the smallest $\Delta Z$, $G/B \sim 10^{10}$. Finally, when bonds with the largest $G_i$ are removed we find that $G/B \sim \Delta Z^{\mu_{G+}}$, with $\mu_{G+} = 3.05 \pm 0.01$. Although $G/B$ diverges/varies with slightly different power laws in two dimensions, the overall effect is no less drastic.

Note that our procedures are remarkably efficient in tuning $G/B$. Figures 3 and 4 show that by removing less than 2% of the bonds in three-dimensional networks we can obtain a difference of more than 16 orders of magnitude in the tuned value of $G/B$, depending on which bonds we prune. In two dimensions, pruning is similarly efficient; starting with the same initial configuration we are able to obtain differences in $G/B$ that span over 17 orders of magnitude by pruning only $\sim 1\%$ of the bonds. We also note that our bond-cutting procedures do not create any zero-frequency vibrational modes in the system, which would herald an instability in the structure.

The limit $G/B \to 0$ corresponds to the incompressible limit of a solid where the Poisson ratio, $\nu = (d - 2G/B)/[d(d - 1) + 2G/B]$ in $d$ dimensions, reaches its maximum value of $\nu = +1$ (in 2$d$) or +$1/2$ (in 3$d$). The limit $G/B \to \infty$ corresponds to the auxetic limit where the Poisson ratio reaches its minimum value of $\nu = -1$. By using these different pruning algorithms, we can tailor networks to have any Poisson ratio between these two limits. This ability provides great flexibility in the design of network materials.

We turn now to spatial correlations between cut bonds. Driscoll et al. [18] have conducted numerical simulations in which they removed bonds with the largest strain under uniaxial or isotropic compression or shear. They showed that the cut bonds form a damage zone whose width increases and diverges as the initial excess coordination number, $\Delta Z_{\text{initial}} \to 0$; for sufficiently small $\Delta Z_{\text{initial}}$, the pruned bonds are homogeneously distributed throughout the entire system. Outside this zone, they found that the network is essentially unaffected.

When pruning bonds with the largest contribution to $B$ or $G$, all the data presented thus far are for systems with a sufficiently small $\Delta Z_{\text{initial}}$ so that the distribution of the cut bonds appears homogeneous. In our simulations with large $\Delta Z_{\text{initial}}$, where the damage zone is smaller than the size of our system, we find that $G/B$ still diverges/varies, but does so when $\Delta Z > 0$. When we remove the bond with the smallest contribution to $B$ or $G$, the bonds are initially removed homogeneously throughout the system, independent of $\Delta Z_{\text{initial}}$. The existence of tunable strong spatial correlations in the cut bonds, as found by Driscoll et al. [18], allows one to create textured materials spatially varying mechanical properties. One region may be highly incompressible while a nearby region may be highly auxetic. This offers a great variety in the mechanical response of these networks.

For many materials [5] the Poisson ratio decreases with increased connectivity of the constituent particles and increases with packing density. We note that neither of these correlations hold for the algorithms we have introduced for tuning the Poisson ratio (or ratio of shear and bulk moduli). We can reach $G/B \to \infty$ (minimum Poisson ratio) or $G/B \to 0$ (maximum Poisson ratio) by removing the same number of bonds from the same starting configuration. Neither the overall connectivity nor the overall density is different in the two final states. Thus, our procedures for producing tunable Poisson ratio materials are fundamentally different from correlations considered in the literature.

We have presented a number of ways of tuning $G/B$. Our results suggest that these ideas may be extended to other global properties (e.g., thermal expansion or electrical response [19, 20]) where the response can be written in terms of sums over bond contributions. As long as there is independence of bond-level response, one should be able to tune the ratio of global properties by using the same protocol of removing bonds that are especially susceptible (or especially unsusceptible) to a given global perturbation.

Our results demonstrate that disordered networks provide particularly elegant opportunities for constructing mechanical metamaterials with tunable, flexible and spatially textured response. However, the algorithms we have presented may not be restricted to artificially constructed materials. For example, compressing a network composed of springs that fail when stressed past a given threshold would result in the same network as removing springs with the largest $B_i$, provided that the threshold is sufficiently small. It is also not beyond imagination that one could selectively break bonds at the nanoscale level in response to global perturbations in complex solids. Indeed, biology appears to be able to target structures in networks that are under particularly high stress and to enhance their strength (such as in trabecular bone [21]). Alternatively, there may be mechanisms to buckle or sever strongly stressed fibers (such as in actin networks [22]). It is interesting to ask if such selective repair or destruction of biological structures changes ratios of different mechanical responses such as the Poisson ratio.
Appendix A: Calculation of bond-level elastic response

We consider networks of nodes connected by unstretched central-force springs with stiffness $k = 1$. Let $\vec{\delta r}_i$ be the total strain on bond $i$ when the system is deformed according to some strain tensor $\epsilon_{\alpha\beta}$. The change in energy of the network is then given to lowest order by

$$\Delta E = \sum_i k \delta r_{i,\parallel}^2,$$  \hspace{1cm} (A1)

where $\delta r_{i,\parallel}$ is the component of $\delta r_i$ that is parallel to the bond direction. Thus, the bond that contributes the most (least) to the response to a given boundary deformation is the one with the largest (smallest) $\delta r_{i,\parallel}^2$. To remove the bond that contributes the most to the bulk modulus, for example, one would remove the bond with the largest $\delta r_{i,\parallel}^2$ under compression. This procedure can be implemented in either a simulation or an experiment.

In practice, for our computations, we use linear algebra to calculate the response of each bond more efficiently, as follows. The bulk elasticity of a system is described to linear order by the elastic modulus tensor $c_{\alpha\beta\gamma\delta}$, so that if the system is distorted by the symmetric strain tensor $\epsilon_{\alpha\beta}$, the change in energy is given to leading order by

$$\Delta E/V = \frac{1}{2} \epsilon_{\alpha\beta} c_{\alpha\beta\gamma\delta} \epsilon_{\gamma\delta},$$  \hspace{1cm} (A2)

where $V$ is the volume of the system. In general, there are 6 (21) independent components of the elastic modulus tensor in two (three) dimensions, but in the isotropic limit this reduces to just the bulk modulus $B$ and the shear modulus $G$.

The components of $c_{\alpha\beta\gamma\delta}$ are calculated from the change in energy of the system under various boundary deformations using Eq. (A1). The strain $\delta r_i$ can be decomposed into two distinct parts. First there is an affine strain set directly by the strain tensor. However, this results in a nonzero net force, $\vec{f}_m$, on each node $m$, leading to a secondary non-affine response. This non-affine response is calculated by solving the following system of equations

$$\mathcal{M}_{mn} \vec{u}_{m,NA} = \vec{f}_n,$$  \hspace{1cm} (A3)

where $\mathcal{M}_{mn}$ is the Hessian matrix and $\vec{u}_{m,NA}$ is the non-affine displacement of each node. The total strain $\delta r_i$ of bond $i$ is calculated from the sum of the affine and non-affine displacements of the two nodes that the bond connects. Since $\Delta E$ can be written as a sum over bonds, so too can the elastic modulus tensor:

$$c_{\alpha\beta\gamma\delta} = \sum_i c_{i,\alpha\beta\gamma\delta}.$$  \hspace{1cm} (A4)

Under the deformation $\epsilon_{\alpha\beta}$, the change in energy of bond $i$ is

$$\Delta E_i = \frac{1}{2} \epsilon_{\alpha\beta} c_{i,\alpha\beta\gamma\delta} \epsilon_{\gamma\delta},$$  \hspace{1cm} (A5)

$c_{i,\alpha\beta\gamma\delta}$ thus completely describes the bond-level elastic response for bond $i$, and can be used to calculate the quantities $B_i$ and $G_i$ considered in the main text.

The global bulk and shear moduli are linear combinations of the components of the elastic modulus tensor. In two dimensions, they are

$$B = \frac{1}{2} (c_{xxxx} + c_{yyyy} + 2c_{xxyy})$$
$$G = \frac{1}{2} (4c_{xyz} + c_{xxxx} + c_{yyyy} - 2c_{xxyy}),$$

while in three dimensions they are

$$B = \frac{1}{2} (c_{xxxx} + c_{yyyy} + c_{zzzz} + 2c_{gyyz} + 2c_{xzzz} + 2c_{xzyy})$$
$$G = \frac{1}{15} (3c_{gyyz} + 3c_{xzzz} + 3c_{xzyy} + c_{xxxx} + c_{yyyy} + c_{zzzz} - c_{gyzz} - c_{xzzz} - c_{xzyy}).$$

Finite disordered systems are never perfectly isotropic, so the shear modulus always has some dependence on the angle of shear. The above expressions for $G$ represent the angle-averaged shear modulus, which reduces to the shear modulus in the isotropic limit of infinite system size. We calculate the contribution of bond $i$ to the bulk and shear moduli in exactly the same way:

$$B_i = \frac{1}{2} (c_{i,xxxx} + c_{i,yyyy} + 2c_{i,xxyy})$$
$$G_i = \frac{1}{2} (4c_{i,xxyy} + c_{i,xxxx} + c_{i,yyyy} - 2c_{i,xxyy}),$$

in two dimensions, and

$$B_i = \frac{1}{2} (c_{i,xxxx} + c_{i,yyyy} + c_{i,zzzz} + 2c_{i,gyyz} + 2c_{i,xzzz} + 2c_{i,xzyy})$$
$$G_i = \frac{1}{15} (3c_{i,gyyz} + 3c_{i,xzzz} + 3c_{i,xzyy} + c_{i,xxxx} + c_{i,yyyy} + c_{i,zzzz} - c_{i,gyzz} - c_{i,xzzz} - c_{i,xzyy}).$$

in three dimensions.

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