The Principle of Independent Bond-Level Response: Tuning by Pruning to Exploit Disorder for Global Behavior

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Phys. Rev. Lett. **114**, 225501 — Published 4 June 2015

DOI: 10.1103/PhysRevLett.114.225501

The principle of independent bond-level response: tuning by pruning to exploit disorder for global behavior

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(Dated: May 5, 2015)

We introduce a principle unique to disordered solids wherein the contribution of any bond to one global perturbation is uncorrelated with its contribution to another. Coupled with sufficient variability in the contributions of different bonds, this "independent bond-level response" paves the way for the design of real materials with unusual and exquisitely tuned properties. To illustrate this, we choose two global perturbations, compression and shear. By applying a bond removal procedure that is both simple and experimentally relevant to remove a very small fraction of bonds, we can drive disordered spring networks to both the incompressible and completely auxetic limits of mechanical behavior.

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 global response of a solid 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 single bond from a perfectly 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], leaving their ratio 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: independent bond-level response. This independence refers not only to 1) the significant variation in the response at the individual bond level, but also, and more importantly, to 2) the dearth of strong correlations between the responses of any specific bond to different perturbations. To illustrate this principle, we consider the specific perturbations of compression and shear. We construct networks in which individual bonds are successively removed to drive the overall system into different regimes of behavior characterized by the ratio G/B of the shear modulus, G, to the bulk modulus, B. Starting from the same initial network, we can remove as few as 2% of the bonds to produce a network with a value of G/B that is either nearly zero (incompressible limit where the Poisson ratio is $\nu = 1/(d-1)$ in d dimensions) or nearly infinite (maximally auxetic with $\nu = -1$ [5]) merely by removing different sets of bonds. Moreover, by using different algorithms or starting with different networks, one can confine the region within which the bonds are removed to strips of controllable size, ranging from a few bond lengths to the size of the entire sample [6]. This has the practical consequence that one can achieve precise spatial control in tuning properties from region to region within the network – as is needed for creating origami [7, 8] or kirigami [9] materials.

We construct networks numerically by starting with a configuration of particles produced by a standard jamming algorithm [10, 11]. We place N soft repulsive particles at random in a box of linear size L and minimize the total energy until there is force balance on each particle. We work in either two or three dimensions and start with a packing fraction 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 [12]. We characterize the network by the excess coordination number $\Delta Z \equiv Z - Z_{\text{iso}}$, where Z is the average number of bonds at each node and $Z_{\text{iso}} \equiv 2d - 2d/N$ is the minimum for a system to maintain rigidity in d dimensions [13]. We note that networks produced this way have no long-range order [10], unlike networks constructed by randomly displacing cites on a lattice [14, 15].

For each network, we use linear response to calculate the contribution B_i of each bond i to the bulk modulus, $B = \sum_i B_i$. (B_i is proportional to the change in energy of bond i when the system is uniformly compressed, see the Supplementary Materials for details). The distribution of B_i in three dimensions is shown in blue in Fig. 1. In all plots, data is averaged over 500 networks, each with approximately 4000 nodes and an initial excess coordination number $\Delta Z_{\text{initial}} \approx 0.13$ (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 shear modulus, $G = \sum_i G_i$. (A finite system is not completely isotropic, so the shear modulus varies with direction [16]; we calculate the angle-averaged shear modulus, which approaches the isotropic shear modulus in the infinite system-size limit [17].) The resulting distribution for G_i is shown in purple in Fig. 1. Note that the distributions of the bond contributions to B and G are continuous, 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 forms a power law at low values of B_i or G_i, which is then terminated above $\langle B_i \rangle$ and $\langle G_i \rangle$ by approximately exponential cut-offs. Such significant variation in bond-level response is consistent with previous observations [18, 19] and is in stark contrast to a perfect crystal where the distributions would be composed of discrete delta functions.

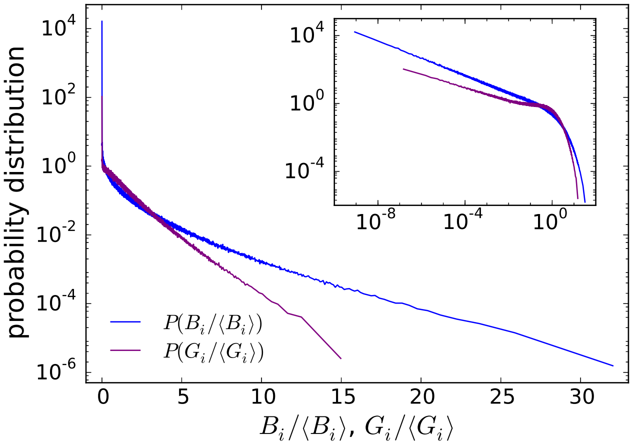

FIG. 1. Variation in bond-level response. Distribution on a log-linear scale (inset: log-log scale) of the contribution of each bond to the macroscopic bulk and shear moduli, B_i and G_i, for $3d$ networks with $\Delta Z_{\mathrm{initial}} \approx 0.13$. Here i indexes bonds. At low B_i or G_i, the distributions follow power-laws with exponents -0.51 and -0.38, respectively. At high values, the distributions decay over a range that is broad compared to their means, $\langle B_i \rangle$ and $\langle G_i \rangle$.

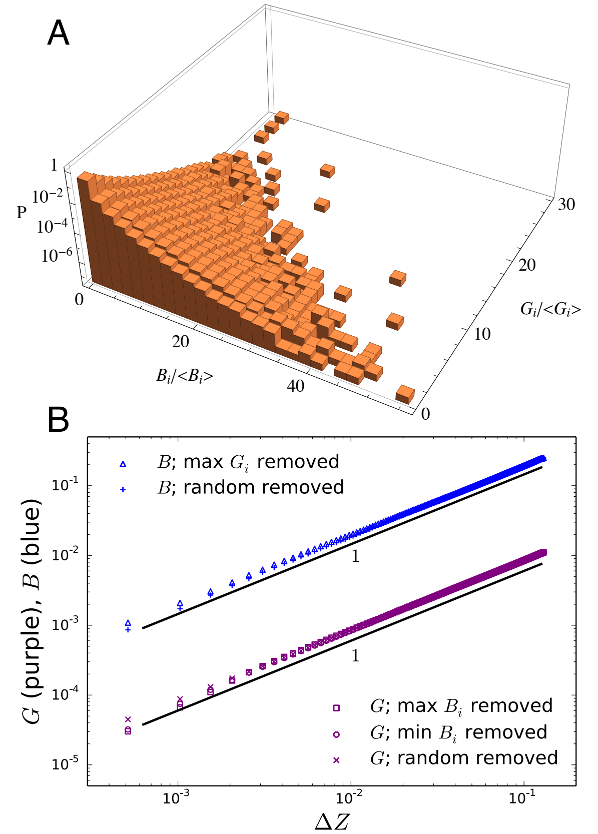

FIG. 2. Independence of bond-level response. (**A**) Joint probability distribution of B_i and G_i for $3d$ networks with $\Delta Z_{\mathrm{initial}} \approx 0.13$. There is little apparent correlation between the response to compression (B_i) and to shear (G_i) for a given bond i. (**B**) The value of G when bonds with the largest (purple squares) and smallest (purple circles) B_i are removed is nearly indistinguishable from G when bonds are removed at random (purple crosses). Similarly, B is very similar whether bonds with the largest G_i (blue triangles) are removed or bonds are removed at random (blue pluses).

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? Figure 2a shows the joint probability distribution $P(B_i, G_i)$. A strong positive correlation between B_i and G_i would produce a linear trend on this graph, which is clearly not observed. We conclude that the correlations are weak, although we note that they are also not vanishingly small (see Supplementary Material). This lack of strong correlation between B_i and G_i is again qualitatively different from what one would find for a simple crystal. Thus, Figs. 1 and 2a illustrate a previously-unrecognized property that is well obeyed by our disordered networks: independent 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, 20, 21], 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 of this procedure: at each step, a bond is removed only if each node it connects 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 [22]. As the excess coordination number decreases, the bulk and shear moduli vanish together, so that $G \sim B \sim \Delta Z$ [4, 20, 21][23] (see Fig. 2b). Therefore, as shown in Fig. 3, G/B is independent of ΔZ.

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 $B_i \to 0$, 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. Once the bond has been removed, the contributions B_i and G_i of the remaining bonds to the moduli must be recalculated because they depend on the connectivity of the entire network. This process of removing the bond with the smallest B_i and then recalculating the values of B_i for the remaining bonds is then repeated many times. Figure 2b shows that when bonds with the smallest B_i are successively removed, the *shear* modulus is linearly proportional to ΔZ. Furthermore, it is quantitatively indistinguishable from the case where bonds are removed at random. The ability to alter the behavior of B without affecting the behavior of G is a clear demonstration of the principle of independent bond-level response.

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 indeed 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 ΔZ is lowered by decompressing the system.

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, independent bond-level response predicts that the shear modulus will again decrease linearly with ΔZ, 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.

The algorithms mentioned above can be extended in a number of ways. As a further example, one can remove the bond with the largest contribution to the shear modulus to drive G/B towards zero. In this case, independent bond-level response implies that the bulk modulus will 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).

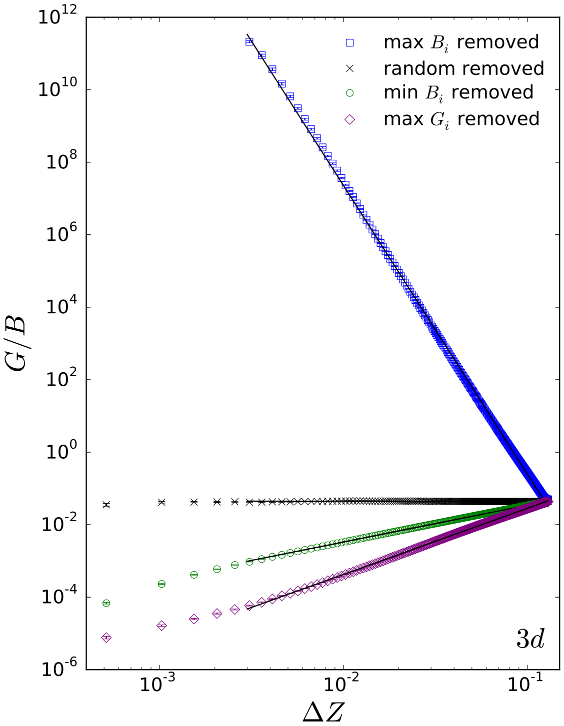

FIG. 3. Tuning global response in three dimensions. The ratio of shear to bulk modulus, G/B, for four pruning algorithms. Error bars (included) are smaller than the symbols. Lines are fits to the data over the indicated range and have slopes, from top to bottom, of -7.96, -0.01, 1.01, and 1.82. Starting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

Note that the presence of a non-trivial zero-frequency vibrational mode (which our bond-cutting procedure does not explicitly forbid) would herald an instability in the structure. We look for such modes by diagonalizing the dynamical matrix, but do not observe them until the system is at or extremely close to isostaticity [24] If we remove bonds with the *smallest* G_i, however, we find that zero modes appear when ΔZ is still quite large, preventing G/B from diverging as we would expect. A variant of our procedure could prevent this (*e.g.* by including a constraint that a removed bond not create any zero modes).

We can tune two-dimensional networks with equal 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. As shown in Fig. S2, the behavior of G/B is qualitatively similar to 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 ($G/B \sim \Delta Z^1$), 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 ΔZ, $G/B \sim 10^{10}$. 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/vanishes with slightly different power laws in two and three dimensions, the overall effect is no less dramatic.

Note that our procedures are remarkably efficient in tuning G/B. Figure 3 shows that by removing about 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, we are able to obtain differences in G/B that span over 17 orders of magnitude by pruning only $\sim 1\%$ of the bonds. This is even more surprising given the fact that B_i and G_i are somewhat correlated. The fact that we can still tune the ratio of the moduli so drastically demonstrates the robustness of the principle of independent bond-level response.

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 $2d$) or $+1/2$ (in $3d$). 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.

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 turn now to spatial correlations between cut bonds. Driscoll *et al.* [6] have conducted numerical simulations and experiments 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.

Since B_i (or G_i) in our simulations is proportional to the strain squared, our procedure is identical to that of Driscoll *et al.* [6] if we remove bonds with the *largest* contribution to the relevant elastic constant. So far, all the data we have presented are for systems with a sufficiently small $\Delta Z_{\text{initial}}$ so that the distribution of the cut bonds appears homogeneous. However, we find that G/B diverges/vanishes regardless of $\Delta Z_{\text{initial}}$ (see Fig. S3), demonstrating that the ability to drastically tune G/B does not depend on the spatial distribution of removed bonds or system size. When we remove the bond with the *smallest* contribution to B or G, the bonds are removed homogeneously throughout the system, independent of $\Delta Z_{\text{initial}}$. Our results, combined with the work of Driscoll *et al.*, means that elastic properties can be tuned not only globally but also on a local scale controlled by the initial connectivity – one region may be highly incompressible while a nearby region may be highly auxetic. This offers tremendous flexibility in the design of new and interesting materials.

We have presented a number of ways of tuning G/B in disordered networks by using the principle of independent bond-level response. However, these ideas may be extended to other global properties as well. For example, one can imagine controlling thermal expansion by tuning nonlinear terms. One can even consider different classes of systems, such as a disordered resistor network [25, 26] where one may be able to independently adjust the components of the conductivity tensor to design a highly anisotropic device. In general, to tune two properties relative to each other, one first must be able to quantify contributions at the single-bond level. The principle of independence holds if 1) there is a sufficient variation in the bond-level contributions (*i.e.* Fig. 1), and 2) the contribution of a bond to one property is not strongly correlated with its contribution to the other (*i.e.* Fig. 2a). One could then independently tune these properties by removing bonds that contribute disproportionally to one property or the other.

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 are not restricted to artificially constructed materials. Compressing a real network composed of springs that fail when stressed past a given threshold would lead to 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 [27]). Alternatively, there may be mechanisms to buckle or sever strongly stressed fibers (such as in actin networks [28]). 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.

We thank Bryan Chen, Michelle Driscoll, Heinrich Jaeger and Vincenzo Vitelli for important discussions.

This research was supported by the US Department of Energy, Office of Basic Energy Sciences, Division of Materials Sciences and Engineering under Awards DE-FG02-05ER46199 (A.J.L., C.P.G.) and DE-FG02-03ER46088 (S.R.N.). This work was partially supported by a grant from the Simons Foundation (#305547 to A.J.L.).

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