Effect of aging on the non-linear elasticity and memory formation in materials

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Disordered solids under stress often evolve slowly as they age, leading to changes in their elastic response. We study two models for the microscopic evolution of properties of such a material; the first considers changes in the material strength while the second considers distortions in the microscopic geometry. In both cases, aging encodes a memory of the strain at which the system is prepared and dramatically affects the material’s non-linear elastic properties. To test the robustness of this effect, we conduct experiments on quasi-two-dimensional networks. Despite being vastly more complex than the simulation models, aging these laboratory samples under compression yields similar memories seen in the models. Our results demonstrate how aging produces directed elastic behavior that can be varied as a function of applied strain.

INTRODUCTION

The inevitable fate of a glass left on its own is to age and progressively lower its free energy in a rugged energy landscape [1–4]. Due to enormous relaxation times, the system does not reach equilibrium at any accessible time scale. As it ages, the elastic properties evolve as particles rearrange or bonds break and form. Because preparation into the initial metastable state can produce desired properties that are inaccessible in thermal equilibrium, aging is often considered to be detrimental since it allows the system to evolve away from this state.

It was recently proposed, however, that stress-induced aging can be exploited to manipulate an out-of-equilibrium solid to achieve desired elastic responses [5]. Imposing strain directs the manner in which a solid ages. This relies on the fact that straining a disordered system gives rise to a spatially varying stress pattern that depends sensitively on the applied deformation. Since each bond is subjected to a different amount of stress, aging affects each bond to a varying degree. The elastic response of a disordered system depends sensitively on the precise microscopic details of the bonds [6]. Different imposed macroscopic strains thus give rise to changes in the corresponding stiffnesses or elastic moduli.

In this paper, we focus on the nonlinear elastic response of a system that has been aged under an applied stress. We find that aging dramatically affects the nonlinear response. We are particularly interested in creating auxetic (i.e., negative Poisson’s ratio $\nu < 0$) materials. These are not ubiquitous in nature and are thought to have interesting applications [7–18]. Here we show that a system can become auxetic in the nonlinear elastic regime simply by aging under an applied strain. This result demonstrates the power of the directed-aging protocol.

Moreover, aging encodes in the response a memory of the strain at which the system is aged. By measuring certain features in the Poisson’s ratio (or the deformation energy) as a function of the imposed strain, we are able to read out the strain at which the system was aged.

We complement simulations, with experiments conducted on networks cut from sheets of foam that are aged under compression. Here, the non-linear Poisson’s ratio also shows a signature of the strain at which it was aged. That it appears in both experiments as well as simple models suggests that these memories are a robust feature of materials that are aged at large strains.

Using the single-bond stress correlation at different strains, we also characterize the inherent capacity of a network to be manipulated as a function of strain. For example, we show that the system is more tunable under compression than expansion and that networks are more tunable when they have a lower coordination number. Altogether, our results open the possibility of manipulating the elastic properties of a material far beyond the linear regime.

Models

We model a disordered solid as a random spring network, where each spring, indexed by $i$, has spring constant, $k_i$, and rest length, $\ell_{i,0}$. The total elastic energy is:

$$U = \frac{1}{2} \sum_i k_i (\ell_i - \ell_{i,0})^2 .$$

(1)

Here $\ell_i$ is the length of spring $i$.

To ensure that our networks are initially rigid, we derive the ensemble of central-force spring networks from packings of soft spheres at force balance under an external pressure [19–21]. The sphere centers define the locations of the nodes and overlapping spheres are connected by springs. The equilibrium spring length is chosen to be the distance between nodes, guaranteeing that in the absence of deformation, the system is unstressed and at zero energy. We characterize the connectivity of the network with the average coordination number $Z = 2N_b/N$, where $N_b$ is the number of bonds and $N$ is the number
of nodes. At the jamming transition, where the particles just touch, \( Z = Z_c = 2d \) is the smallest coordination number needed to maintain rigidity in \( d \) dimensions. We use networks that are above the jamming transition: \( \Delta Z \equiv Z - Z_c > 0 \).

A system that is under imposed stress ages in a manner that will reduce its internal energy \([1, 2]\). For a network of springs, such plastic deformation can change both the stiffnesses, \( k_i \), and the equilibrium lengths, \( \ell_{i,0} \). In a physical system both types of changes are important \([5]\). We disentangle their effects by using two models. 

**k-model:** This model, introduced earlier \([5]\), captures the weakening of the bonds. We assume that the rate at which a bond becomes weaker depends on its energy so that it weakens both when compressed or extended:

\[
d_\ell k_i = -\gamma k_i (\ell_i - \ell_{i,0})^2. \tag{2}\]

Here \( \gamma \equiv \frac{1}{\tau_0 (\ell_{i,0}^2)} \), where \( \tau_0 \) is a material-dependent relaxation time and \( \langle \ell_{i,0}^2 \rangle \) corresponds to the average of the square lengths of the bonds before aging. This model is similar in spirit to the design strategy in which bonds with large stresses are preferentially removed. \([6]\).

We conduct experiments where we age physical networks in such a way as to lessen geometrical changes in the networks as they age. These experiments are therefore designed to better approximate the \( k \)-model than the experiments conducted in Ref. \([5]\). We construct experimental systems by laser cutting 2D networks out of sheets of EVA (ethylene vinyl acetate) foam. We start with uncompressed networks and age them under compression. Such aging of a network makes it significantly smaller so that its geometry changes significantly. In order to avoid that as much as possible, we let the system relax after aging. This brings it back to almost, but not exactly, the original size and geometry. The dynamics are much faster at higher temperatures, so we perform all our experiments (compression as well as relaxation) in an oven at 50° C. This temperature is warm enough to increase the rate of aging significantly but not hot enough to damage the material. The compression and relaxation of a network constitutes one cycle of aging. We age our networks for multiple cycles until the dynamics slow down.

**\( \ell \)-model:** This model assumes that the stress in a bond is reduced by changing its *equilibrium length*. The rate of change of the length depends on a bond’s tension; they elongate under tension and shorten under compression:

\[
d_\ell \ell_{i,0} = \beta k_i (\ell_i - \ell_{i,0}). \tag{3}\]

Here, \( \beta \equiv \frac{1}{\tau_0 (\ell_{i,0}) (k_i)} \), \( \langle \ell_{i,0} \rangle \) is the average bond length and \( \langle k_i \rangle \) is the average spring constant. When the rate at which it undergoes creep is much slower than the time to reach force balance, this model reduces to the Maxwell model for viscoelasticity \([22]\). Each bond consists of a spring, which describes the rapid elastic behavior, in series with a dash-pot, which at long times accounts for the change in rest lengths of the spring. Similar dynamics have also been used to account for junction remodelling in epithelial cells \([23, 24]\).

We note that in Ref. \([5]\) we were able to test the effects of purely geometrical changes by taking an image of aged networks and laser-cutting a new network with the same geometry. By this protocol, the effects of aging on spring constants captured by the \( k \)-model are eliminated. Note also that geometrical changes were responsible for the bulk of the response in those experiments. Thus, those experiments, in which physical networks were simply aged at a fixed strain, should be closer to the \( \ell \)-model than to the \( k \)-model, although geometrical effects not taken into account in the \( \ell \)-model, such as bending of bonds in the network may also play a role.

We note that the two models can be expressed similarly and combined:

\[
\partial_t k_i = -\gamma k_i (\ell_i - \ell_{i,0})^2, \tag{4}
\]

\[
\partial_t \ell_{i,0} = -\beta \frac{\partial U}{\partial \ell_{i,0}}. \tag{5}\]

We assume aging is much slower than the time to reach force balance. The microscopic parameters evolve by steepest descent to minimize the energy at a rate proportional to the energy gradient.

Generally, there are multiple ways by which materials evolve under stress or strain. Work hardening is the strengthening of metals or polymers in response to strain \([25]\). Bone may undergo remodelling in response to loads, by increasing the bone mass \([26, 27]\). Frictional contacts increase their contact area over time, strengthening their interface \([28, 30]\).

**RESULTS**

*Aging under isotropic compression in simulations*

*Aging protocol and evolution of the energy landscape in the k-model:*

We begin with an unstressed network and compress it to the aging strain, \( \epsilon_{Age} \). The strain is \( \epsilon \equiv \frac{L - L_0}{L_0} \), where \( L \) is the length of the system while \( L_0 \) is the length in the unstrained system. (Compression corresponds to a negative strain.) To simulate quasistatic dynamics we compress in small steps, minimizing the energy with respect to the locations of the nodes at each step. Aging can be neglected during the measurement itself.

We measure the energy as a function of strain, \( U(\epsilon) \), for isotropic expansion and compression. In linear response, \( U(\epsilon) = \frac{1}{2} V B \epsilon^2 \), where \( V \) is the volume and \( B \)
is the bulk modulus. The initial network has no internal stresses, so that the global energy minimum is at zero strain where \( U(\epsilon = 0) = 0 \). Aging in the k-model only changes the spring constants so the global energy minimum remains at \( \epsilon = 0 \). After the system has been aged at \( \epsilon_{Age} \), we allow the system to re-expand to its original volume and measure the behavior with respect to \( \epsilon = 0 \) as origin.

As noted above, aging reduces the energy at the strain at which the system has been aged. As shown in Fig. 1(b), the energy at the strain \( \epsilon = \epsilon_{Age} \) is reduced after aging compared to the unaged material; the energy landscape has become very asymmetrical.

Since the state at \( \epsilon = 0 \) remains the global minimum with zero energy, there will be at least two states with low energy \( \epsilon = 0 \) and \( \epsilon = \epsilon_{Age} \). If \( \epsilon_{Age} \) is small, the low-energy states encompass the entire range \([0, \epsilon_{Age}]\); however, if \( \epsilon_{Age} \) is large, aging can result in two local energy minima separated by an energy barrier.

The low-energy state at \( \epsilon_{Age} \), is a memory in the landscape of the conditions under which the system was aged. By measuring the elastic properties, the aging strain can be read out.

### Poisson’s ratio aged under isotropic strain in k-model:

The Poisson’s ratio, \( \nu \), is the negative ratio of the transverse strain, \( \epsilon_r \), to an imposed uniaxial strain \( \epsilon \): \( \nu = -\frac{\epsilon_r}{\epsilon} \).

Within linear response for an isotropic material, \( \nu \) is a monotonic function of the ratio of the shear modulus, \( G \), to the bulk modulus, \( B \); \( \nu = \frac{d-2G/B}{(d-1)+2G/B} \), where \( d \) is the spatial dimension. At larger strains, \( \nu \) generically depends on the magnitude of \( \epsilon \). We measure \( \nu \) by compressing (or expanding) the system uniaxially, while allowing the system to relax in the transverse directions. If the system is isotropic then the transverse strain, \( \epsilon_r \), is the same in all transverse directions.

Fig. 1(b) shows that in the linear-response regime \( \nu \) decreases as it ages and ultimately may become negative. At long times, \( \nu \) monotonically decreases with \( |\epsilon_{Age}| \). This is consistent with the system aging in a directed manner [5]: that is, aging under compression lowers the bulk modulus more than it lowers the shear modulus.

The relaxation rate has a characteristic dependence on the imposed strain. The right-hand side of Eq. 2 scales as \( \epsilon_{Age}^{2} \), the energy of the system. To compare different aging strains, we rescale time in Fig. 1(b) as \( t_{Age}^{2} \). In Fig. 2 we show that \( \nu \) at \( \epsilon = \epsilon_{Age} \) approximately collapses as a function of \( t_{Age}^{2} \).

The nonlinearities in the energy landscape suggest that \( \nu \) may have interesting dependence on the strain. We show how \( \nu(\epsilon) \) evolves with time in Fig. 1(c). For the unaged systems, \( \nu \) depends very weakly on strain even up to 10% strain. As the system ages, \( \nu(\epsilon) \) is lowered, especially near \( \epsilon_{Age} \). It later develops a minimum near the aging strain, that deepens over time.

In Figure 2(d) we plot \( \nu(\epsilon) \) for different values of \( \epsilon_{Age} \), at the same scaled time \( t_{Age}^{2} \). In all the curves, a minimum appears in \( \nu(\epsilon) \) close to \( \epsilon = \epsilon_{Age} \) (vertical dashed lines). Thus, aging produces a memory of the strain at which it was prepared, allowing the aging strain to be read out from the minimum of the non-linear Poisson’s ratio.

Aging in the non-linear regime allows exotic behavior to be trained into a system. For example, a system can have a positive \( \nu \) within linear response but a large negative \( \nu \) for larger uniaxial compression (see the curve for the aging strain of \( \epsilon_{Age} = -0.05 \) in Fig. 2(d)). Therefore, compressing uniaxially leads to a transverse expansion, which is then reversed to transverse compression at larger values of strain.

### Aging protocol and evolution of the energy landscape in the \( \ell \)-model:

In the \( \ell \)-model, as the system ages under an imposed strain, it evolves by changing the rest lengths of the bonds, \( \ell_{i,0} \), to reduce the stresses and the elastic energy. We compress the system to the target strain, \( \epsilon_{Age} \), and then allow it to age. Since the rest lengths have evolved, the global energy minimum is no longer at zero strain. Since the energy minimum shifts, we measure the elastic properties with respect to the new (aged) global minimum. We denote \( \epsilon' \) to be the strain measured with respect to the unaged system and \( \epsilon \) measures the strain with respect to the aged system minimum.

At long times, all the stresses decay to zero and the global energy minimum shifts to \( \epsilon_{Age} \) as shown in Fig. 3(e). At intermediate time, before it reaches that point, the minimum lies between \( \epsilon' = 0 \) and \( \epsilon' = \epsilon_{Age} \). To find the position of the minimum, we minimize the energy with respect to the node locations, as well as the width and height of the box.

Note that the network shown for the k-model in Fig. 1a has a larger coordination number \( Z \) than the one used for the \( \ell \)-model in Fig. 1. While this choice is not essential, it allows a comparison of the different effects at the same range of strains between the two models. This is the cause of the difference between the two models for the unaged energy versus strain.

Besides the shift of the energy minimum, there are additional changes to the energy versus strain curves apparent in Fig. 3(e). First, we note that the curvature at the new global minimum, which determines the bulk modulus, \( B \), is greatly reduced. This lowers the Poisson’s ratio, as we will discuss in detail below. Another interesting feature is the kink in the nonlinear regime near \( \epsilon' = 0 \), corresponding to unstressed state of the unaged system (this corresponds to a strain of \( \epsilon = \frac{-\epsilon_{Age}}{1+\epsilon_{Age}} \), mea-
Figure 1. Left: the $k$-model at $ΔZ ≈ 1.51$. Right: $ℓ$-model at $ΔZ ≈ 0.53$. (a), (e) The compression energy versus strain for an unaged system and a system aged under 5% compression (dashed line). In the $k$-model the global minimum remains at zero strain, since bond lengths do not change while in the $ℓ$-model the global minimum shifts. (b) and (f) The Poisson’s ratio within linear response versus time. In the $ℓ$-model the curve is non-monotonic and has a minimum. (c) and (g) The evolution of the Poisson’s ratio as a function of strain. In the $k$-model, the local minimum occurs near the aging strain, $ε_{Age} = −0.05$ (vertical dashed line). In the $ℓ$-model the minimum corresponds to the strain needed to undo the volume change that occurred during aging (dashed lines). (d) and (h) The Poisson’s ratio versus strain. For the $k$-model these are measured at a constant $ε_{Age}$. The aging strain is denoted by vertical dashed lines. In the $ℓ$-model, the curves are at a constant $tε_{Age}$ and the dashed lines denote the strain corresponding to the aged system at asymptotic times, $−ε_{Age}/(1 + ε_{Age})$. Note that in (e) the strain is measured with respect to the unaged system while for (g) and (h) it is measured with respect to the new global minimum, which depends on the aging time and $ε_{Age}$.

Figure 2. The Poisson ratio at $ε = ε_{Age}$ as a function of scaled time approximately collapses.

Poisson’s ratio aged under isotropic strain in $ℓ$-model:

We first consider the evolution of the Poisson’s ratio within linear response, as shown in Fig. 1(f). Aging under constant compression $ε_{Age} < 0$, results in the Poisson’s ratio, $ν(ε → 0)$ decreasing with time. The decay is non-monotonic, with a local minimum at intermediate time scales. At large times it ceases to evolve and its asymptotic value depends on $ε_{Age}$. At small values of $|ε_{Age}|$, $ν$ remains positive. For systems aged at larger $|ε_{Age}|$, however, $ν$ decreases further and ultimately the system becomes auxetic for sufficiently large aging strain $|ε_{Age}|$. This is the behavior seen in experiments on aging networks [3].

By contrast to the behavior under compression, aging under a small constant expansion (blue curve) increases the linear-response Poisson’s ratio, $ν$ only slightly. We believe that this is a general feature of elastic systems and is due to instabilities that occur under compression but not under expansion. These instabilities reduce the stiffness to compression significantly, thus lowering $ν$. Instabilities have also been shown to give rise to materials with negative Poisson’s ratio in periodic structures [81].

The nonlinear Poisson’s ratio versus strain is shown for different times in Fig. 1(g). These curves have a minimum at $ε > 0$ (expansion), and the value of strain grows with time. The dashed lines in Fig. 1(g) showing the applied strains approximately match the location of the minima. This suggests that the minimum is associated with the strain needed to return to the same volume as the unaged system.

We next consider the nonlinear behavior of the $ℓ$-model.
at long aging times. Figure 3(h) shows the Poisson’s ratio versus strain for the unaged system and for systems aged at different values of $\epsilon_{\text{Age}}$. For the unaged system, $\nu(\epsilon)$ decreases when the system is compressed. At positive strains the aged systems develop a local minimum that is a memory of the strain at which they were aged. Unlike in the $k$-model, where the memory occurs at $\epsilon = \epsilon_{\text{Age}}$, the minimum in this case occurs near the strain which corresponds to that of the unaged system ($\epsilon' = 0$). Aging under compression reduces the volume and to return to the initial volume the system must be strained by $-\frac{\epsilon_{\text{Age}}}{1+\epsilon_{\text{Age}}}$. This is indicated by dashed lines in Fig. 3(h).

We believe, that the mechanical microscopic instabilities also play an important role in encoding the memories. These occur when the system is aged under compression. Expanding the system after the system has been aged, causes some of these instabilities to be “undone”. This results in the unusual nonlinear response near $\epsilon' = 0$.

### Aging under isotropic compression in experiments

In analyzing our experiments, the aging strain, $\epsilon'_{\text{Age}}$, is measured with respect to the unaged network. Our experimental protocol is to compress the network to the same size in every cycle. This means that it experiences slightly different compressive strains in each cycle. Once we stop aging the network, we bring it back to room temperature and measure $\epsilon_{\text{Age}}$, which is the strain at which the network was compressed with respect to the aged network. For aging under compression, $|\epsilon_{\text{Age}}| < |\epsilon'_{\text{Age}}|$.

The behavior of networks when aged at different $\epsilon'_{\text{Age}}$ is shown in Fig. 3. Poisson’s ratios are shown as a function of measurement strain, $\epsilon$, measured with respect to the aged networks. Blue, green and red data are from networks aged at $\epsilon'_{\text{Age}} = -0.1$, $-0.2$, and $-0.3$. The vertical dashed lines are at the corresponding $\epsilon_{\text{Age}}$. Both the red and green data show minima in the Poisson’s ratio at $\epsilon_{\text{Age}}$. This is similar to the memory seen in the $k$-model. It is likely that when $\epsilon_{\text{Age}}$ is very small, it is no longer very effective at aging; this is what is seen in the blue data.

The experimental data provides strong evidence that the $k$-model captures effects that can be realized in real materials. Our experimental results raise many interesting questions about the nature of aging in these systems. For example, the time dependence of aging and the effects of multiple aging cycles are not well understood. These results open up various avenues for further research that would allow us to understand the limits to which we can train a material.

### Aging under shear strain in simulations

To test the generality of the nonlinear effects of aging, we consider systems that are aged under a constant shear strain rather than under compression. We demonstrate that the ensuing change in elasticity is more subtle and does not necessarily follow the intuition gleaned from the case of aging under compression.

We shear the elastic networks by compressing along the x-axis by $\epsilon_{\text{Age}}$ and extended along the y-axis by $\frac{\epsilon_{\text{Age}}}{1+\epsilon_{\text{Age}}}$ to preserve volume. As before, we characterize the elastic behavior by measuring the Poisson’s ratio. However, aging under shear evolves the system to be anisotropic so that $\nu$ depends on the direction of applied strain. We focus on the Poisson’s ratio measured by straining either along the x-axis ($\nu_x$), or y-axis ($\nu_y$), while the system is allowed to relax in the transverse direction.

**Poisson’s ratio aged under shear strain in $k$-model:**

In Fig. 3(a) we show the stiffness to shear, measured from the elastic energy per unit strain squared, $G(\epsilon) = 2U_G/V\epsilon^2$. In the limit of $\epsilon \to 0$ this corresponds to the linear-response shear modulus. As expected, aging under shear lowers $G(\epsilon)$. The strain dependence of $G(\epsilon)$ shows additional features. In particular $G(\epsilon)$ has a minimum at a value of strain that depends on the aging strain. The minima occur at a strain that is slightly different than $\epsilon_{\text{Age}}$ but appears to be proportional to $\epsilon_{\text{Age}}$. 
The difference between $K_x$ and $K_y$ is the result of the asymmetry between compression and expansion in the non-linear regime in which the system is aged. We find that the stiffness to compression in the nonlinear regime is smaller than for expansion. Therefore, the stresses along the $y$-axis are larger, and as a result $K_y$ ages faster and becomes weaker than $K_x$. Thus, aging at shear strains in the nonlinear regime can affect different linear response moduli differently.

**Poisson’s ratio aged under shear strain in $\ell$-model:**

Fig. 4(d) shows the stiffness to shear as a function of the measuring strain, $G(\epsilon)$. For negative strains (i.e. in the direction which the system was aged) the stiffness grows, while for large positive strains the stiffness decreases. This is different from the case of aging under compression, where stiffness to compression decreases. Similar behavior has been reported in sheared gels. There the increase in stiffness was attributed to bonds aligning in a preferential direction defined by the shear deformation. Prior to shearing, the system was isotropic and each bond angle is equally probable. The shear deformation, in our case, tends to align the bonds along the $y$-axis. This increases the uniaxial stiffness in the $y$-direction, $K_y$, and reduces the stiffness in the $x$-direction, $K_x$ (see Fig. 4(f)). This is consistent with experiments and explains why in linear response $\nu_y > \nu_x$, as shown in Fig. 4(e).

The nonlinear behavior of $\nu_x$ and $\nu_y$ has a different dependence on the shear strain, as shown in Fig. 4(e). The slope of $\nu_x$ and $\nu_y$ have opposite signs. Interestingly, $\nu_y$ has a peak at negative values of strain and then falls steeply. We believe, that as in the case for compression, the sharp drop in $\nu_y$ is associated with instabilities. The threshold value of strain for these instabilities decreases with aging strain.

In summary, in both models aging under shear gives rise to a change in the stiffness to shear. The direction in which these evolve are not always obvious, and also result in nontrivial nonlinear behavior.

In real systems, local bond stiffnesses can be weakened ($k$-model) and local geometry can change ($\ell$-model). The differences between the behavior of the $k$-model and $\ell$-model could allow one to determine which of the two aging scenarios that they embody is dominant for a given system at a given strain. In the case of the $k$-model we find that $\nu_x > \nu_y$ while in the $\ell$-model $\nu_x < \nu_y$. In addition, the slope of $\nu_y$ at $\epsilon = 0$ has the opposite sign for the two models.
Aging under shear in experiments

In Ref. [5] we considered the effect of aging under a constant shear strain in foam networks, similar to those used here. There the system was not cycled to minimize the permanent deformation, and the initially square network became rectangular. We follow the same notation used here. There the system was not cycled to minimize the constant shear strain in foam networks, similar to those used in Ref. [5].

In the unaged system the Poisson’s ratio is virtually independent of direction, and \( \nu_{\text{unaged}} \approx \nu_{y} \). After aging the response becomes highly non-isotropic, with \( \nu_{y} > \nu_{\text{unaged}} > \nu_{x} \). This is consistent with the scenario of aging in the \( \ell \)-model, shown in Fig. 4(e). Though the comparison is only qualitative, it demonstrates that these models can capture complex behavior that arises in real materials. It opens the door to quantitative studies in the nonlinear regime and over a broader range of parameters.

Together with our experiments on aging under isotropic compression described earlier, these results show that physical networks can be manipulated by the aging protocol to more closely resemble the \( k \)-model or the \( \ell \)-model. Thus, both models are to some extent realizable in real networks.

Inherent nonlinear tunability of the Poisson’s ratio:

We have shown that aging at finite isotropic or shear strains, \( \epsilon_{\text{Age}} \), allows the Poisson’s ratio to be manipulated in the nonlinear elastic regime as well as in the linear regime. The ability to manipulate the non-linear behavior does not only depend on the aging dynamics but is an inherent property of how forces are transmitted in the network. In the linear regime, the pruning of bond \( i \) leads to a change in the bulk modulus, \( \Delta B_i \), that is uncorrelated with the change in the shear modulus, \( \Delta G_i \) [9] [32]. It is this property – the independence of bond-level response – that allows the linear-regime Poisson’s ratio to be tuned so successfully by pruning.

To quantify the inherent tunability of the Poisson’s ratio in the nonlinear regime, we consider the contribution of a single bond to the modulus [6], which we denote for compression \( B_i (\epsilon) = k_i \delta x_i B / \epsilon^2 \), where the extension \( \delta x_i B \) depends on the amplitude of the imposed strain. Essentially, this is the energy in a single bond per square unit strain; in the linear regime, it reduces to the contribution of bond \( i \) to the linear bulk modulus. If \( B_i \) is a constant independent of the amplitude of \( \epsilon \) for all \( i \), then the elastic behavior does not depend on \( \epsilon_{\text{Age}} \); the aging strain only changes the aging rate. The nonlinear correlations are characterized by comparing the correlations of \( B_i (\epsilon) \) with their corresponding values in linear response. We use the Pearson correlation function of two random variables, which is defined as:

\[
C (y, x) = \frac{\langle xy \rangle - \langle x \rangle \langle y \rangle}{\sigma_x \sigma_y}, \quad \text{where} \sigma \text{ is the standard deviation.}
\]

Fig. 5(a) shows \( C (B_i (\epsilon), B_i (0)) \) versus strain for both compression and expansion. Depending on the coordination number, \( \Delta Z \), correlations under compression decay differently from those under expansion: the correlations are similar below a threshold value of \( \Delta Z \), but then drop much more rapidly in the case of compression. This threshold, which appears to vanish in the limit of \( \Delta Z \rightarrow 0 \), signals the breakdown of linear response at the level of a single bond. Correlations under expansion remain significant even for small \( \Delta Z \).

The ability to tune the non-linear response to shear strain is characterized by \( C (G_i (\epsilon), G_i (0)) \), as shown in Fig. 5(b). Here, \( G_i (\epsilon) = k_i \delta x_i G / \epsilon^2 \) is measured at a shear strain of \( \epsilon \). The correlations decay faster when \( \Delta Z \rightarrow 0 \). This implies that the system is more tunable in the non-linear regime near \( \Delta Z \rightarrow 0 \).

We can also characterize the ability in the nonlinear regime to tune the bulk and shear moduli independently. Within linear response in two dimensions, the correlation, \( C (B_i (\epsilon), G_i (\epsilon)) \), was found to be small, \( \approx 0.17 \) [9]. At a finite compressive strain we find that this correlation becomes even smaller. Under expansion, the correlations increase but remain below 0.33.

We note that these correlations provide a measure of the inherent ability to tune a given modulus in the nonlinear regime. They do not depend on the protocol by which the system evolves under aging. They depend only on the nonlinear elasticity, and could be used to identify interactions or geometries that are particularly amenable to manipulation by aging.
Minimization of a cost function is a common design method for achieving certain target properties, such as auxetic behavior. While this is effective on a computer at sufficiently small system sizes, minimization is usually not possible to implement in the laboratory and is not scalable to arbitrarily large systems. Directed aging evolves a mechanical system so that the elastic properties of the system change depending on the imposed deformation and strain. In contrast to cost-function minimization by computer, directed aging can be performed on systems of any size both on the computer and in the laboratory [5]. Here we have shown that directed aging is highly effective at tuning the Poisson’s ratio even in the nonlinear elastic regime. This result is important to many applications, such as cushioning impact.

We find that directed aging profoundly affects the nonlinear behavior in a way that can be very different from how it changes the linear response. In some cases the Poisson’s ratio can be manipulated so that it changes sign as a function of strain. This could allow a designer to manipulate the energy landscape and create material with desired properties that vary as a function of the amplitude of deformation. More complex energy landscapes could clearly be achieved by varying the strain as the system ages.

Remarkably, the behavior in our minimal models is very similar to the aging experiments we conduct in foam samples. In both cases there is a memory of the strain they are aged, and it can be read out from the minimum of the Poisson’s ratio. We note however, that the models based on central-force spring networks neglect many effects such as the energy cost for varying the angle between adjacent bonds [34] and the bending and buckling of bonds at large strains. In addition, the dynamics during aging are far more complex, involving a broad range of time scales [5]. The striking similarity suggests that the governing principles by which a material ages is of broader generality and independent of the precise interactions and geometry.

Our results are also similar to the behavior seen in other more complex materials, such as, rubbers [35], foams [36] and actin networks [22]. In the Mullins effect, a variety of rubber undergo softening when they are strained [35][37][38]. Softening occurs up to the strain to which they were deformed, similar to behavior in the k-model. Sticky colloidal gels and glasses also exhibit shear softening [39][40]. The behavior we find for aging under compression in the ℓ-model is consistent with the scenario proposed by Lakes [36] to explain the transformation of solid foams, that yields a negative Poisson’s ratio. Furthermore, the stiffening under shear of fibrillar networks [32][41] is similar to our finding in the ℓ-model; such stiffening has been explained in terms of the change in the network geometry, consistent with the underpinnings of the ℓ-model. Thus, the effects we discuss here could apply more broadly in disordered soft matter systems, whose structure is sensitive to strain.

A key feature of these models, is that aging imprints a memory of strain at which the system was prepared. In the k-model, the minimum of the Poisson’s ratio marks the aging strain. In the ℓ-model the system remembers the strain that corresponds to the initial state. The difference between these behaviors, could provide an experimental test to distinguish the dominant effects. We note that in both models, memory is inherently a nonlinear effect, as it is measured from the strain dependence of response functions. This memory is another example of the broad range of memories that occur in out-of-equilibrium disordered systems [32][47]. Perhaps some of the insights gleaned from our models could be relevant there.

Finally, we have extended the theoretical understanding of tunability for linear response [6][33] to the nonlinear regime. The ability to train a response that depends on strain requires that stresses at different strains become uncorrelated. This is quantified by measuring correlation in $B_i(\epsilon)$ and $G_i(\epsilon)$ with their corresponding value in linear response. We find, that reducing the coordination number increases the ability to tune the system, and that it is much easier to tune the system under compressive strains than under expansion.

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