

Atomic-scale origin of viscoelastic response in model disordered solids from local inversion-symmetry breaking

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Viscoelasticity has been described since the time of Maxwell as an interpolation of purely viscous and purely elastic response, but its microscopic atomic-level mechanism in solids has remained elusive. We studied three model disordered solids: a random lattice, the bond-depleted fcc lattice, and the fcc lattice with vacancies. Within the harmonic approximation for central-force lattices, we applied sum-rules for viscoelastic response derived on the basis of non-affine atomic motions. The latter motions are a direct result of local structural disorder, and in particular, of the lack of inversion-symmetry in disordered lattices. By defining a suitable quantitative and general atomic-level measure of nonaffinity and inversion-symmetry, we show that the viscoelastic responses of all three systems collapse onto a master curve upon normalizing by the overall strength of inversion-symmetry breaking in each system. Close to the isostatic point for central-force lattices, power-law creep $G(t) \sim t^{-1/2}$ emerges as a consequence of the interplay between soft vibrational modes and non-affine dynamics, and various analytical scalings, supported by numerical calculations, are predicted by the theory.

I. INTRODUCTION

The viscoelasticity of solids has been the object of intense debate at least since the time of Maxwell. Continuum mechanics and relaxation models have flourished through all the last century, with many extensions proposed to capture different behaviours observed in metallurgy [1, 2]. For crystals with line defects, Andrade creep (whereby the relaxation shear modulus presents the power-law scaling $G(t) \sim t^{-1/3}$) has been convincingly explained by Nabarro, Mott and others in terms of dislocation dynamics [3, 4]. Internal friction, which represents the imaginary part of the viscoelastic response also known as the loss modulus G'' , has been interpreted in earlier models, in terms of diffusive motion of atoms associated with defect mobility.

In glasses the situation is more complicated, because dislocations are difficult to identify, and the origin of internal friction and complex relaxation behaviour observed typically (power-law or stretched-exponential) has remained unexplained. A recent work [5] has applied elegant field-theoretic methods within coherent-potential approximation, starting from the assumption of spatially heterogeneous static shear modulus, to successfully recover the α -wing asymmetry in the resonance peak of G'' in oscillatory rheology observed in experiments. However, the theory is on the continuum level, and does not clarify which microscopic (atomic-level) features ultimately control the viscoelastic response.

Recent simulation work [6] motivated by this problem in the context of metallic glasses, has shown that internal friction in glasses may have its origin in quasi-localized correlated motions that have an avalanche-like character. Furthermore, these excitations were found to be suppressed in regions of high icosahedral symmetry. Power-law creep $G \sim t^{-1/2}$ was recovered in previous

work using mean field theory [7] and average stress fluctuations [8]. The same result was found in a related field of athermal jammed solids, where simulations and scaling arguments [9, 10] based on Kelvin-Voigt viscoelasticity have been combined with asymptotics of the vibrational density of states (DOS) near the jamming transition (at which a jammed solid loses rigidity) with average contact number $Z = 6$, although the strongly non-affine motion of the particles, which is crucial for disordered and jammed solids [11, 12], was not explicitly taken into account in the scaling analysis [9]. We improve on these methods by taking into account the exact microstructure of the system as well as the non-affine motions of all particles, providing a direct link between the microscopic landscape and the frequency and time dependent shear modulus.

II. MODEL SYSTEMS

Here we re-examine this problem by considering three very different model systems of amorphous solids in 3d, of which 2d slices are given in Fig.1. We will work with a specific model of disordered harmonic spring networks formed from the low- T equilibration of dense Lennard-Jones fluids. This is a good model for atomic disordered solids (defective crystals, metallic glasses) but different from other types of disordered networks where the preparation protocol may change the critical exponents and the critical coordination numbers [14–17].

The first lattice is a random network of harmonic springs generated according to the protocol in Ref. [18]: a Lennard-Jones glass is formed and equilibrated in a metastable minimum, after which all nearest-neighbour interactions are replaced by harmonic springs, all with the same spring constant κ and with a relatively narrow

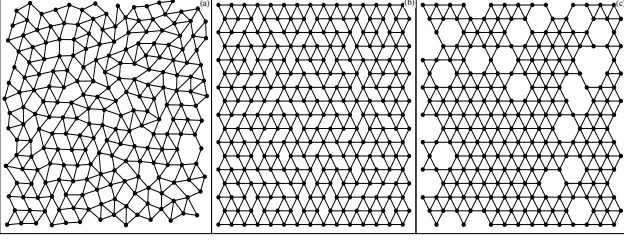


FIG. 1. Two dimensional schemata of our model systems. (a) the random network, (b) the fcc lattice with randomly cut bonds and (c) the fcc lattice with randomly removed atoms.

distribution of spring length R_0 . Upon randomly cutting the harmonic bonds in the sample, lattices with variable coordination number Z can be formed. In the present work this depletion process is performed in such a way that we get a very narrow distribution of coordination numbers to avoid effects stemming from fluctuating connectivity in the system.

The two fcc lattices (the bond-depleted, Fig.1b, and with vacancies, Fig.1c) are instead generated starting from a perfect fcc lattice with $Z = 12$ and same spring constant κ and lattice constant R_0 as the random lattice. The microstructure, and in particular the local symmetry, of the three lattices is, however, very different. For example, in Ref. [18] it was shown that the standard bond-orientational order parameter F_6 , which measures the spread in the orientations of bonds on the lattice [19], is practically equal to 1 for the bond-depleted fcc (for any Z value), whereas it is much lower ($\simeq 0.3$) for the random lattice.

For these models we develop an analytical theory of viscoelastic response based on the non-affine deformation formalism, which is a fully microscopic approach. Our analysis shows that, surprisingly, the oscillatory moduli of these systems fall onto a master curve after normalizing by an order parameter which describes the average degree of local inversion-symmetry on any atom. The same order parameter controls the non-affine particle rearrangements that have a cooperative quasi-localized character, which explains the findings of simulations [6]. Further, the power-law creep $G \sim t^{-1/2}$ found near the isostatic transition of all the three lattices is shown to be the consequence of both the excess of soft modes in the DOS, and crucially, also of the underlying non-affine dynamics.

III. FORMALISM

The starting point of our analysis is the microscopic equation of motion for a particle in a disordered lattice, which was derived for the case of a phenomenological damping motion with constant damping coefficient ν , in Ref. [13] and was shown, also in [13], to reduce to a simple

harmonic-oscillator type equation for the deviation variable \underline{x}_i which measures the particle displacement from the original position:

$$m\ddot{\underline{x}}_i + \nu\dot{\underline{x}}_i + \underline{H}_{ij}\underline{x}_j = \underline{\Xi}_{i,\kappa\chi}\eta_{\kappa\chi}. \quad (1)$$

We used the Hessian of the system $\underline{H}_{ij} = -\partial^2\mathcal{U}/\partial r_i\partial r_j = -\partial f_i/\partial r_j$ and the non-affine force $\underline{\Xi}_{i,\kappa\chi} = \partial f_i/\partial \eta_{\kappa\chi}$. Here, $\eta_{\kappa\chi}$ denotes the Cauchy strain tensor for a generic deformation field. For a shear deformation, $\kappa\chi \equiv xy$. The non-affine force $\underline{\Xi}_{i,\kappa\chi}$ represents the net force that acts on a particle that is en route towards its affine position. If the particle's original position in the undeformed lattice is \underline{R}_0 , the affine position is defined as $\underline{r}_{i,A} = \underline{\eta}\underline{R}_{i,0}$. In a perfectly centrosymmetric lattice, the particle en route towards this affine position receives forces from its nearest-neighbours which cancel each other out by symmetry, leaving the particle at equilibrium in the affine position. In a disordered lattice, due to local breaking of inversion-symmetry on the given particle, these forces do not cancel, and their vector sum is a net force that brings the particle to a final (non-affine) position which differs from $\underline{r}_{i,A}$. For a generic harmonic lattice with no pre-stress, the non-affine force vector is defined as $\underline{\Xi}_{i,\alpha\beta} = -R_0\kappa \sum_j \hat{n}_{ij}^\alpha \hat{n}_{ij}^\beta \underline{\hat{n}}_{ij}$, with R_0 and κ being the rest distance and force constant between the particles. The sum is performed over the nearest neighbours and includes the unit bond vector $\underline{\hat{n}}_{ij}$ pointing from atom i to j .

Normal-mode decomposition of the terms in Eq.(1) onto the eigenvectors \underline{v}_p (where $p = 1 \dots N$) of the Hessian, and taking the Fourier transform of the equation of motion as in Ref.[13], lead to the complex viscoelastic shear modulus for oscillatory shear deformation (with imposed frequency Ω):

$$G^*(\Omega) = G^A - 3\rho \int_0^{\omega_D} \frac{D(\omega)\Gamma(\omega)}{m\omega^2 - m\Omega^2 + i\nu\Omega} d\omega. \quad (2)$$

Here we introduced the frequency correlator of the non-affine forces, $\Gamma_{xyxy}(\omega) = \langle \hat{\Xi}_{p,xy} \hat{\Xi}_{p,xy} \rangle_{p \in \{\omega, \omega+\delta\}}$, where $\hat{\Xi}_{p,xy} = \underline{\Xi}_{xy} \cdot \underline{v}_p$. Also, $\rho = N/V$ is the atomic density, or number of atoms (or nodes) on the lattice per unit volume. G^A is the affine shear modulus (also known as the Born-Huang modulus), which is independent of the applied frequency Ω , and coincides with the elastic response in the limit $\Omega \rightarrow \infty$. Here, ω denotes the eigenmode frequency of internal vibrations of the lattice, and ω_D denotes the Debye frequency, i.e. the highest frequency of the vibrational spectrum. The latter spectrum, i.e. the normalized distribution of vibrational eigenmodes is represented by the DOS, denoted here as $D(\omega)$. The mass of the particles m is set to 1 for the reminder of the paper, since it's of no concern in the present work.

The above sum-rule allows the calculation of the complex shear modulus for any harmonic lattice for which both the DOS and the correlator function $\Gamma(\omega)$ can be easily

evaluated numerically. For the DOS we follow the same procedure as in Ref. [18], whereas for $\Gamma(\omega)$ we follow the procedure of Ref. [13]. This is a straightforward exercise for the three model lattices shown in Fig.1.

Next we consider the time-dependent shear modulus $G(t)$, which can be calculated by taking the inverse Fourier transform of $G^*(\Omega)$,

$$\begin{aligned} G(t) &= -3\rho \int_{-\infty}^{\infty} \int_0^{\infty} \frac{D(\omega)\Gamma(\omega) \exp(i\Omega t)}{\omega^2 - \Omega^2 + i\nu\Omega} d\omega d\Omega \\ &= 6\pi\rho t e^{-\frac{\nu}{2}t} \int_0^{\infty} D(\omega)\Gamma(\omega) \text{sinc}\left(\frac{1}{2}\sqrt{4\omega^2 - \nu^2}t\right) d\omega. \end{aligned} \quad (3)$$

Here $\text{sinc}(x) = \sin(x)/x$ denotes the cardinal sine function. Numerical evaluation of Eq.(6) for the three lattices at the two representative values of Z are reported in Fig.4. Again we took advantage of the simple cubic fitting Eq.(3) for the product $D(\omega)\Gamma(\omega)$, which allows one to avoid the problem of a numerical gap between zero frequency and the first eigenfrequency (this gap is not negligible for systems with $N < 10^5$ and our simulated lattices have $N = 5 \times 10^4$). For small times we observe a plateau that corresponds to the high-frequency affine response, after which a power-law decay is observed with an exponent comprised in the range between $-1/2$ and $-3/4$. This power-law can be understood mechanistically as follows.

IV. RESULTS

We have calculated $G^*(\Omega)$ for the three lattices with two different average coordination numbers $\langle Z \rangle$: $Z = 7.0$, where all lattices are mechanically well stable; $Z = 6$ (for the fcc with vacancies) and $Z = 6.1$ (for the two bond-depleted systems), i.e. very close to the point of marginal stability. First we calculated the vibrational density of states $D(\omega)$ and the correlator function $\Gamma(\omega)$, which are shown in Fig. 2. Since these quantities appear as the $D(\omega)\Gamma(\omega)$ product in Eq.(2), it is convenient to study this product as a single function of ω .

Remarkably, we notice from Fig. 2 that, although $D(\omega)$ and $\Gamma(\omega)$ behave differently for each of the three systems and have a rather complicated form, their product, when normalized by the quantity $\langle |\Xi|^2 \rangle / \rho$, shows a strikingly *universal* behaviour over the full frequency range, and can be fitted by a simple cubic function of ω , of the form

$$\frac{D(\omega)\Gamma(\omega)}{\langle |\Xi|^2 \rangle / \rho} \sim \omega^2(\omega_D - \omega). \quad (4)$$

Here, the quantity $\langle |\Xi|^2 \rangle$ is evaluated by taking the square of the absolute value of each vector $|\Xi_i|$, constructed for each atom i , and averaging over all atoms in the system. This same quantity has been used to form a suitably normalized order parameter in [18].

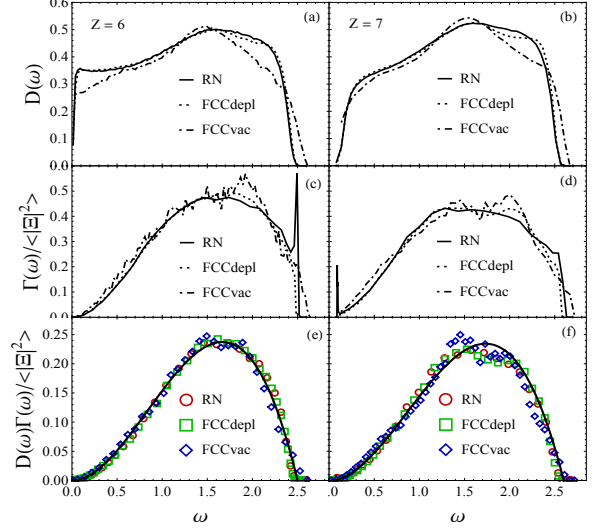


FIG. 2. Density of states $D(\omega)$ (a,b) and correlator function $\Gamma(\omega)$ (c,d) for the three different model systems and two different coordination numbers ($Z = 6.1$ and $Z = 7$, respectively). They are normalized by $\langle |\Xi|^2 \rangle / \rho$, proportional to the average absolute square of the non-affine force field to obtain a master curve. (e,f) show the product $D(\omega) \cdot \Gamma(\omega)$ which appears in the formula for $G^*(\Omega)$. Remarkably, the product of the two functions can be conveniently fitted by a simple cubic function $\omega^2(\omega_D - \omega)$, represented as a solid line.

Since $D(\omega)$ approaches a low- ω plateau in the limit of marginal stability ($Z \rightarrow 6$), as is known from many studies in the past [20, 22]), the low frequency behaviour of $D(\omega) \cdot \Gamma(\omega)$ is dominated by the correlator function $\Gamma(\omega) \sim \omega^2$, a result that was derived in Ref. [23]. It is interesting to note Dirac-delta spikes in $\Gamma(\omega)$, which happen at frequencies that correspond to strongly-localized modes: at $Z = 6$ a spike is visible near the top of the spectrum, where modes tend to be Anderson-localized. At $Z = 7$, instead, a spike is visible at a frequency close to the Ioffe-Regel crossover [21] (and to the boson peak frequency) where modes are also strongly localized [18].

Let us now consider the viscoelastic response of the three models systems. We use the convention of splitting the complex shear modulus into its real and imaginary part $G(\Omega) = G'(\Omega) + iG''(\Omega)$. Both moduli can be calculated according to

$$G'(\Omega) = G^A - 3\rho \int_0^{\omega_D} \frac{D(\omega)\Gamma(\omega)(\omega^2 - \Omega^2)}{(\omega^2 - \Omega^2)^2 + \nu^2\Omega^2} d\omega \quad (5)$$

$$G''(\Omega) = 3\rho \int_0^{\omega_D} \frac{D(\omega)\Gamma(\omega)\nu\Omega}{(\omega^2 - \Omega^2)^2 + \nu^2\Omega^2} d\omega, \quad (6)$$

and are plotted in Fig.3. In the numerical calculation we implemented the convenient cubic form of the product $D(\omega)\Gamma(\omega)$ that was shown above to be an excellent fitting to the numerical evaluation of these functions. Various scalings have been reported in the plots, which can be ex-

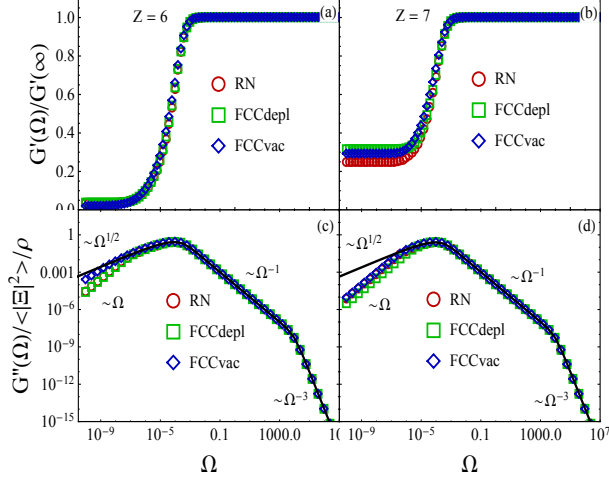


FIG. 3. G' (a,b) and G'' (c,d) of our three model systems for $Z = 6$ (left) and $Z = 7$ (right), respectively. In (c,d) in order to collapse the loss modulus of the three systems onto a single master curve, we have normalized by the factor $\langle |\Xi|^2 \rangle / \rho$.

tracted from the asymptotic analysis of Eq.(4)-(5). Most notable of which are the low frequency scalings of $G''(\Omega)$, which agree very well with EMT results from Ref. [7] ($G'' \sim \Omega^{1/2}$ for $Z \approx 6$ and $G'' \sim \Omega$ for $Z = 7$). Deviations from their numerical results ($G'' \sim \Omega^{0.41}$) might be caused by finite temperature effects in their simulations (whereas our calculation is carried out at $T = 0$). In our work we have used different values of the damping coefficient ν to study its influence on the results. We found that it has no influence on the qualitative behaviour of G' and G'' , besides when it approaches very small values, where we get divergent results. Aside from that ν only shifts the values to smaller Ω and expands the range of the $\sim \Omega^{-1}$ scaling in G'' . We therefore chose a quite large value $\nu = 10'000$ to demonstrate this behaviour clearly and to focus on the physically important case of overdamped dynamics typical of amorphous solids (metallic glasses, organic glasses, foams etc).

We focus on the limit overdamped systems, which is both important and turns to be amenable to analytic simplifications. For large ν and large times we can simplify the expression in Eq.(6). First we take $\sqrt{\nu^2 - 4\omega^2} \approx \nu - 2\frac{\omega^2}{\nu}$, where we use $\omega \ll \nu$. We insert this into Eq.(6) and use the definition of $\sinh(x)$ to

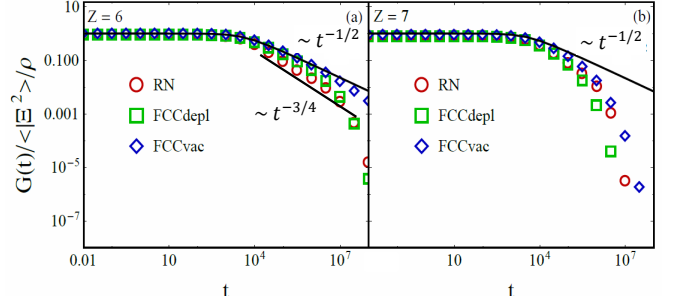


FIG. 4. $G(t)$ of our three model systems for $Z = 6$ (left) and $Z = 7$ (right). We have indicated the different scaling ranges. One can see that the power-law scaling, which is present on 3 – 4 orders of magnitude in time for $Z = 6$, breaks down for $Z = 7$. This is due to the expanding Debye $\sim \omega^2$ regime in the DOS, see text.

get

$$\begin{aligned} G(t) &\approx 12\pi\rho e^{-\frac{\nu}{2}t} \int_0^\infty \frac{D(\omega)\Gamma(\omega) \sinh(\frac{\nu}{2}t - \frac{\omega^2}{\nu}t)}{\omega^2(\nu - 2\frac{\omega^2}{\nu})} d\omega \\ &= 6\pi\rho \int_0^\infty \frac{D(\omega)\Gamma(\omega)(e^{-\frac{\omega^2}{\nu}t} - e^{-\nu t + \frac{\omega^2}{\nu}t})}{\omega^2(\nu - 2\frac{\omega^2}{\nu})} d\omega \\ &\approx 6\pi\rho \frac{1}{\nu} \int_0^\infty \frac{D(\omega)\Gamma(\omega)}{\omega^2} e^{-\frac{\omega^2}{\nu}t} d\omega. \end{aligned} \quad (7)$$

In the last step we have used $\nu \gg 2\omega^2/\nu$ and $\nu t - \omega^2 t/\nu \gg 1$. This corresponds to a system of Maxwell elements with relaxation times $\tau = \nu/\omega^2$. We now recall the standard relationship between the DOS and the eigenvalue spectrum $\rho(\lambda)$ of the Hessian matrix, $D(\omega)d\omega = \rho(\lambda)d\lambda$, with $\omega^2 = \lambda$. At the isostatic point of disordered solids, $Z = 6$ the DOS develops a plateau of soft modes, which is visible in our Fig.2(a). This limit corresponds to the scaling $\rho(\lambda) \sim \lambda^{-1/2}$ in the eigenvalue distribution, which arises from the dominance of random-matrix behaviour in the spectrum, and this scaling can be derived e.g. from the famous Marcenko-Pastur distribution of random-matrix theory, as discussed recently in [24]. In our DOS, a scaling $\rho(\lambda) \sim a + \lambda^{-1/2}$, where a is a constant, is more appropriate since we are in fact slightly above $Z = 6$, and this will explain the power-law exponents in $G(t)$ larger than 1/2 in our calculations. However, we will stick to the simple $\rho(\lambda) \sim \lambda^{-1/2}$ for the asymptotic analysis. Recall now that $\Gamma(\omega) \sim \omega^2$, from the analytical theory of non-affine deformations [23], which implies $\tilde{\Gamma}(\lambda) \sim \lambda$. Inserting these results in the last line of Eq.(7) we obtain the following Laplace transform which can be easily evaluated to:

$$\begin{aligned} G(t) &\sim \int_0^\infty \frac{\rho(\lambda)\tilde{\Gamma}(\lambda)}{\lambda} e^{-\lambda t} d\lambda \sim \int_0^\infty \frac{\lambda^{-1/2}\lambda}{\lambda} e^{-\lambda t} d\lambda \\ &\sim t^{-1/2}. \end{aligned} \quad (8)$$

This scaling for the power-law creep modulus was

shown in simulations of creep in athermal jammed systems in Ref. [9], using a system of Kelvin-Voigt elements (whereas we use a standard-linear-solid or Zener material). The theoretical argument that was proposed to explain the scaling $t^{-1/2}$ was not fully microscopic, because the correlator between eigenmodes and shear field was taken to be independent of the eigenfrequency, hence constant on average for a given frequency interval. This is not a physically justified approximation, because the correlator $\Gamma(\omega)$ in our data (and also in Ref.[13]) displays a strong (and non-random) dependence on the eigenfrequency as one can see in Fig. 2 (c,d). Our model improves substantially on this aspect, by including the eigenfrequency dependence of the non-affine correlator into the theoretical analysis of the scaling. In this way, our framework provides a direct link between the microscopic nonaffine dynamics and the viscoelastic moduli.

V. CONCLUSION

Using the non-affine response formalism, we studied three model harmonic lattices with disorder, which have very different microstructure (as reflected in e.g. different values of bond-orientational order parameter as shown in previous work [18]). Yet, the three different lattices have qualitatively the same (universal) viscoelastic response,

i.e. G' and G'' collapse onto master curves as a function of frequency, once the moduli are normalized by a factor $\langle |\Xi|^2 \rangle / \rho$, where ρ is the atomic density. Here $\langle |\Xi|^2 \rangle$ is crucially related to the symmetry that controls this universality: the local degree of inversion-symmetry. This is evident from the definition of the non-affine force vector for harmonic lattices: $\Xi_{i,xy} = -R_0 \kappa \sum_j \hat{n}_{ij}^x \hat{n}_{ij}^y \hat{n}_{ij}$. The norm of this vector is clearly identically zero for all atoms in a perfectly centrosymmetric lattice with no defects, whereas its value is larger for lattices where the inversion-symmetry is lowered. Hence, the parameter $\langle |\Xi|^2 \rangle$ crucially is proportional to the overall (spatially-averaged) degree to which local inversion symmetry is broken in a disordered lattice.

These results thus identify the atomic-scale origin of internal friction and viscoelastic response in amorphous solids (e.g. glasses) with the local inversion-symmetry breaking, which is the same effect that causes a softer elastic response [18] and is associated with quasi-localized avalanche-like non-affine motions [25]. Our framework provides a clear theoretical explanation to recent simulations results [6] where internal friction was shown to correlate with cooperative non-affine motions and regions of lower local symmetry. This framework will play an important role for the rational design of new materials with tailored viscoelastic response and energy absorption properties in many materials science and engineering applications.

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