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Anisotropic structural predictor in glassy materials

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There is growing evidence that relaxation in glassy materials, both spontaneous and externally driven, is mediated by localized soft spots. Recent progress made it possible to identify the soft spots inside glassy structures and to quantify their degree of softness. These softness measures, however, are typically scalars, not taking into account the tensorial, anisotropic nature of soft spots, which implies orientation-dependent coupling to external deformation. Here, we derive from first principles the linear response coupling between the local heat capacity of glasses, previously shown to provide a measure of glassy softness, and external deformation in different directions. We first show that this linear response quantity follows an anomalous, fat-tailed distribution related to the universal $\omega^4$ density of states of quasilocalized, nonphononic excitations in glasses. We then construct a structural predictor as the product of the local heat capacity and its linear response to external deformation, and show that it offers an enhanced predictability of plastic rearrangements under deformation in different directions, compared to the purely scalar predictor.

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The soft spots are characterized by a degree of softness determined by the typical magnitude of $c_a$, $|c_a|$, in its vicinity.

$$dc_a dy \approx -\mathcal{U}'' \cdot \mathcal{M}^{-1} \cdot (\mathcal{U}'' \cdot \mathcal{M}^{-1} \cdot \mathcal{U}'' \cdot \mathcal{M}^{-1}) \cdot (f_a \cdot \mathcal{M}^{-1})$$

$$- (\mathcal{U}'' \cdot \mathcal{M}^{-1} \cdot \mathcal{U}'' \cdot \mathcal{M}^{-1}) : (\mathcal{M}^{-1} \cdot \mathcal{U}'' \cdot \mathcal{M}^{-1} \cdot f_a).$$

(2)

Equation (2), valid for the largest values of $dc_a/dy$, shows that these emerge from a fourth power of $\mathcal{M}^{-1} \sim \omega^{-2}$ (scalingwise), coupled to the energy anharmonicity tensor $\mathcal{U}''$, to the internal force vector $f_a$ and to the mismatch force vector $\mathcal{U}''$. Note that similarly to $c_a$ (see the expression above), the existence of frustration-induced internal forces $f_a$—an intrinsic signature of glassy disorder—is essential for the emergence of abnormally large values of $dc_a/dy$. While the expression for $dc_a/dy$ [in Eq. (2) or its exact counterpart in Ref. [27]] is universal, the specific information regarding
the applied deformation $\mathcal{H}(\gamma)$ for which the linear response is calculated is encapsulated in the partial derivative $\partial/\partial \gamma$ [27], here through the mismatch force $\mathcal{U}'$. The validity of the analytic expression for $dc_{a}/d\gamma$ has been directly verified using numerical simulations [27].

Universal anomalous statistics. To further establish the linear responses $dc_{a}/d\gamma$ as a fundamental physical quantity that is intrinsically related to quasilocalized soft glassy modes, we consider next the large tail of its statistical distribution. The latter can be predicted based on Eq. (2) and the universal DOS of soft glassy modes, $D_{\Omega}(\omega)$ $\sim\omega^4$. Considering the eigenrepresentation of $dc_{a}/d\gamma$ and invoking the same considerations as in Ref. [6], one can show that objects such as those appearing on the right-hand side of Eq. (2) are far more sensitive to quasilocalized glassy modes than to extended phonons as $\omega \to 0$ and that the $\omega$ dependence emerges only from $\mathcal{M}^{-1} \sim \omega^{-2}$. Consequently, we have $dc_{a}/d\gamma \sim \omega^{-3}$ and $p(dc_{a}/d\gamma)$ is predicted to satisfy $p(dc_{a}/d\gamma) = D_{\Omega}(\omega)d\omega/d(dc_{a}/d\gamma) \sim (dc_{a}/d\gamma)^{-3/8}$ in the large $dc_{a}/d\gamma$ limit.

To test this prediction, we performed extensive numerical simulations of a conventional computer glass former for both simple and pure shear [27] and extracted the statistics of $dc_{a}/d\gamma$. The results are presented in Fig. 2(a) and are in great quantitative agreement with the theoretical prediction. We thus conclude that $dc_{a}/d\gamma$ attains anomalously large values described by universal fat-tailed statistics related to the universal DOS of soft quasilocalized glassy modes, $D_{\Omega}(\omega) \sim \omega^4$. The relation between $dc_{a}/d\gamma$ and quasilocalized modes suggests that the spatial distribution of the former features localized structures, which will be used next to construct a generalized structural predictor in glasses.

A structural predictor. We have at hand two quantities that appear to capture the essential physical properties of soft spots in glassy materials. First, the LHC $c_{a}$ is a signed scalar whose magnitude $|c_{a}|$ quantifies the degree of softness of soft spots, i.e., it provides a measure for how small the activation barrier for irreversible rearrangements is in some unknown direction. Second, the linear response coupling of the LHC to deformation in a certain direction $dc_{a}/d\gamma$ is a signed quantity that provides a measure for the degree by which externally applied forces affect the activation barrier in the direction in which they are applied. How do the two quantities combine to form a generalized anisotropic structural predictor in glasses? As both $c_{a}$ and $dc_{a}/d\gamma$ are signed quantities and as both are predicted to attain anomalously large values at the loci of soft quasilocalized modes, we expect large positive values of the product $c_{a} dc_{a}/d\gamma$ to single out a subpopulation of the soft spots (previously defined by $|c_{a}|$) that is most relevant for the imposed deformation in a certain direction. Consequently, we propose it as a generalized anisotropic structural predictor in glasses.

As a first test of this idea, we invoke it to predict the large tail statistics of $c_{a} dc_{a}/d\gamma$. As we have $c_{a} \sim \omega^{-4}$ and $dc_{a}/d\gamma \sim \omega^{-8}$ in the small $\omega$ limit, the spatial overlap prediction implies $c_{a} dc_{a}/d\gamma \sim \omega^{-12}$, which leads to $p(c_{a} dc_{a}/d\gamma) \sim (c_{a} dc_{a}/d\gamma)^{-17/12}$ in the large $dc_{a}/d\gamma$ limit [using $D_{\Omega}(\omega) \sim \omega^4$]. This prediction is quantitatively verified in Fig. 2(b) for both simple and pure shear, lending strong support to the idea that the product $c_{a} dc_{a}/d\gamma$ indeed characterizes well-defined soft spots.

We next turn to the spatial properties of $c_{a} dc_{a}/d\gamma$, and first consider the glass realization shown in Fig. 1, which is shown again in Fig. 3(a). The product $c_{a} dc_{a}/d\gamma$ under both simple and pure shear in the positive direction is shown in Figs. 3(b) and 3(c). Here, black and red correspond respectively to positive and negative values of $c_{a} dc_{a}/d\gamma$ (the thickness of the lines quantifies their magnitude). Two major observations can be made: (i) Soft spots that are revealed by $c_{a} dc_{a}/d\gamma$ indeed overlap those revealed by $c_{a}$ alone, and in fact they are more pronounced. (ii) There exist two subspecies of soft spots, one that is positively coupled to deformation in a given direction (black) and one that is negatively coupled to it (red), and these subspecies depend on the direction of the deformation [cf. Figs. 3(b) and 3(c)]. Consequently, the product $c_{a} dc_{a}/d\gamma$ reveals orientation-dependent soft spots that offer enhanced predictive power compared to scalar indicators, which will be tested next.

Quantifying the predictive power of the structural predictor. We first demonstrate the predictive power of $c_{a} dc_{a}/d\gamma$ using the example in Fig. 3; we expect plastic events to occur at one of the softest black (red) spots in Fig. 3(b) when the glass undergoes simple shear deformation in the positive (negative) directions, and similarly for Fig. 3(c) in relation to pure shear in the positive and negative directions. This expectation is fully supported by the results of AQS deformation simulations [27] in the four different directions, as shown by the triangles in Figs. 3(b) and 3(c).

To systematically quantify the predictive power of the proposed structural predictor, we performed extensive computer simulations of a large ensemble of glass realizations deformed in the four different directions and tracked the location of the first plastic event in each one of them. To quantify the degree of predictability, we used the following metric: The system is divided into bins of linear size $\xi = 5$ particle diameters, comparable to the localization length of soft quasilocalized modes [21,23], and assigned a value obtained from the average of the structural indicator inside the bin and all of its neighboring bins (implying that the actual coarse-graining length is in fact larger than $\xi$). A plastic event is assigned a rank $\lambda$ that corresponds to the fraction of the bins with a higher value than that of the bin in which it
In Fig. 4(a) we consider simple shear in the positive direction, C, and plot \( c_{\alpha} \frac{dc_{\alpha}}{d\gamma} \) for positive values of \( \lambda \) (negative ones are set to zero) of it (squares), exhibiting essentially no predictive power, i.e., the curve is quite close to \( C(\lambda) = \lambda \). Negative values of \( c_{\alpha} \frac{dc_{\alpha}}{d\gamma} \) provide excellent predictions for plastic events once the deformation direction is reversed (that is, simple shear in the negative direction is applied), as shown in Fig. 4(b). In fact, when the deformation direction is reversed, the black and red soft spots simply reverse their roles (while \( |c_{\alpha}| \) remains the same, as it is independent of the direction of the driving force), as shown in Fig. 4(b). Essentially the same results are obtained for pure shear [27], as expected from symmetry, further demonstrating the superior predictive power of \( c_{\alpha} \frac{dc_{\alpha}}{d\gamma} \).

**Concluding remarks.** The results presented above show that \( c_{\alpha} \frac{dc_{\alpha}}{d\gamma} \) is a promising structural predictor in glasses. It is a first-principles, model- and system-independent physical quantity that reveals and highlights the orientation dependence of soft spots inside disordered glass states. The transparent analytic structure of \( c_{\alpha} \frac{dc_{\alpha}}{d\gamma} \), and its relation to quasilocalized soft excitations [6], allows us to gain physical insight into the origin of localized soft spots in glasses and their universal statistical properties. Our structural predictor involves only snapshots of nondeformed glasses and the interparticle interactions. The emerging properties of soft spots strongly echo the original Falk-Langer concept of shear transformation zones (STZs) [32] and should help advancing the development of predictive elastoplastic models. Finally, we believe that our results offer a tool to probe the basic physics of glasses including structural relaxation, aging, memory effects, and nonlinear yielding transitions.

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[25] Note that in Ref. [6] the following definition has been used, \( E_\alpha = \lim_{T \to 0} \left( \frac{1}{2} k_B T \right)^{-1} \left( \langle \phi_\alpha \rangle_T - \phi_\alpha(0) \right) \), where \( E_\alpha \) has been termed the normalized local thermal energy (LTE). While this definition identifies with the definition of the zero-temperature local heat capacity in Eq. (1), i.e., \( E_\alpha = c_\alpha \), we use here the latter because it may appear more physically intuitive.