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Zohar Schwartzman-Nowik,[1] Edan Lerner,[2] and Eran Bouchbinder[1]

[1]*Chemical and Biological Physics Department, Weizmann Institute of Science, Rehovot 7610001, Israel*
[2]*Institute for Theoretical Physics, University of Amsterdam, Science Park 904, 1098 XH Amsterdam, The Netherlands*

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 ω^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.

Introduction. At the heart of resolving the glass mystery resides the need to quantify the disordered structures inherently associated with glasses and to relate them to glass properties and dynamics, most notably spontaneous and driven structural relaxation [1,2]. Numerous attempts to address and meet this grand challenge have been made [3–20], aiming at defining structural indicators with predictive powers. Achieving this goal would constitute major progress in understanding glassiness and would provide invaluable insight for developing macroscopic theories of deformation and flow of glasses.

Recently accumulated evidence suggests that spatially localized soft spots are the loci of glassy relaxation, and hence are highly relevant for glass dynamics. These localized soft spots have been related to quasilocalized, nonphononic excitations in glasses [4–6], whose universal ω^4 density of states (ω is the vibrational frequency) has been also established recently [21–24]. Among the structural predictors proposed, most relevant here is the normalized local thermal energy [6], which quantifies the interparticle interaction contribution to the zero-temperature heat capacity, termed hereafter the local heat capacity (LHC) c_α (α is the interaction index).

The LHC c_α is a general (system- and model-independent), first-principles statistical mechanical quantity that reveals soft spots in glassy materials [6]. Yet, the LHC is a scalar that quantifies the resistance to motion *in some unknown direction*. That is, as previously proposed structural predictors in glasses (with the exception of Refs. [15–18]), the LHC misses important tensorial, anisotropic information about the coupling to deformation in a certain direction. For example, an extremely soft spot can be completely decoupled from external forces applied in a certain direction and hence irrelevant for the glass response in this direction.

In this Rapid Communication, we develop and quantitatively test a theory that allows us to identify particularly soft glassy structures, explicitly revealing their anisotropic nature and their intrinsic coupling to the direction of externally applied forces. The theory is developed in two steps; First, the linear response coupling of c_α to external deformation tensors $\mathcal{H}(\gamma)$, parametrized by a strain amplitude γ, is derived. The resulting quantity $dc_\alpha/d\gamma$ is shown to follow an anomalous, fat-tailed distribution related to the universal ω^4 density of states of quasilocalized, nonphononic excitations in glasses. Second, a structural predictor in the product form $c_\alpha \, dc_\alpha/d\gamma$ is physically motivated and shown to filter out soft spots that are not coupled to the external deformation of interest. Finally, a metric for quantifying the predictive power of structural predictors is proposed and extensive computer simulations are used to show that $c_\alpha \, dc_\alpha/d\gamma$ offers an enhanced predictability of plastic rearrangements under deformation in different directions, compared to the LHC c_α alone.

Linear response coupling of the LHC to external deformation. The starting point for our development is the zero-temperature local heat capacity [6,25]

$$c_\alpha \equiv \frac{1}{\frac{1}{2}k_B} \frac{\partial \langle \varphi_\alpha \rangle_T}{\partial T}\bigg|_{T=0}, \qquad (1)$$

where $\langle \varphi_\alpha \rangle_T = \int \varphi_\alpha(\boldsymbol{x}) \exp\left(-\frac{\mathcal{U}(\boldsymbol{x})}{k_B T}\right) d\boldsymbol{x} / \int \exp\left(-\frac{\mathcal{U}(\boldsymbol{x})}{k_B T}\right) d\boldsymbol{x}$, $\boldsymbol{x}$ is a vector of the positions of all particles, φ_α is the potential energy of any pair of interacting particles, $\mathcal{U}(\boldsymbol{x}) = \sum_\alpha \varphi_\alpha$, and k_B is Boltzmann's constant. The sum over the LHC, $\frac{1}{2}k_B \sum_\alpha c_\alpha = \partial \langle \mathcal{U} \rangle_T / \partial T|_{T=0}$, is the thermodynamic, zero-temperature heat capacity C_V.

An analytic low-temperature expansion of $\langle \varphi_\alpha \rangle_T$ allows us to explicitly calculate c_α [6], which takes the form $c_\alpha = \boldsymbol{\varphi}''_\alpha : \boldsymbol{\mathcal{M}}^{-1} - \boldsymbol{f}_\alpha \cdot \boldsymbol{\mathcal{M}}^{-1} \cdot \boldsymbol{\mathcal{U}}''' : \boldsymbol{\mathcal{M}}^{-1}$, where $\cdot$ denotes a contraction over a single index of the relevant tensors and $:$ over two indices. A prime, here and hereafter, denotes a partial derivative with respect to $\boldsymbol{x}$, $\boldsymbol{f}_\alpha = \boldsymbol{\varphi}'_\alpha$ are frustration-induced internal forces, and $\boldsymbol{\mathcal{M}} \equiv \partial^2 \mathcal{U}/\partial \boldsymbol{x} \partial \boldsymbol{x}$ is the Hessian matrix whose
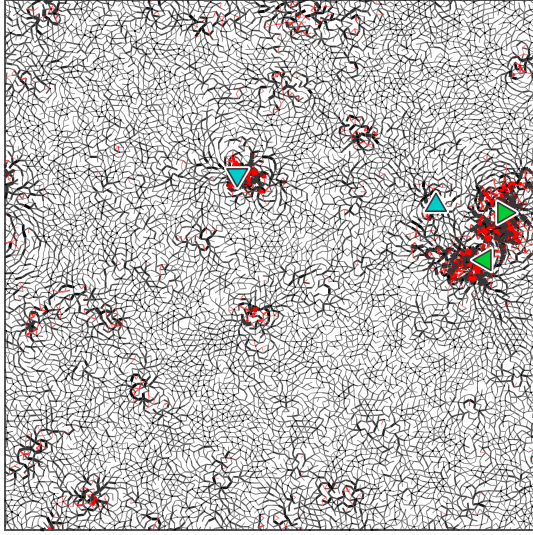
FIG. 1. The local heat capacity (LHC) c_α [cf. Eq. (1)] for a glass composed of $N = 10\,000$ particles [27]. The magnitude of c_α is represented by the thickness of the lines connecting particles and black (red) correspond to positive (negative) values (see Ref. [27] for details about the thresholding procedure employed). Regions with anomalously large $|c_\alpha|$, i.e., soft spots, are clearly observed. The right (left) triangles correspond to the first plastic events under positive (negative) simple shear AQS deformation and the up (down) ones to the first plastic events under positive (negative) pure shear AQS deformation.

eigenvalues are ω^2, where ω is a vibrational (normal mode) frequency. The low-frequency vibrational spectra of glasses feature, in addition to extended phononic excitations (long-wavelength plane waves), also quasilocalized nonphononic excitations, sometimes termed soft glassy modes [21–23]. The latter follow a universal density of states (DOS) $D_{\rm G}(\omega) \sim \omega^4$ that is different from Debye's theory [26].

The LHC c_α is far more sensitive to soft quasilocalized modes than to extended phonons, i.e., it filters out the effect of phonons, and is dominated by its second contribution, which is proportional to the frustration-induced internal forces $\boldsymbol{f}_\alpha$ and to $(\mathcal{M}^{-1})^2$ (scalingwise) [6]. The spatial distribution of c_α reveals soft spots (see Fig. 1) which are highly correlated with the loci of plastic rearrangements under external driving (marked by the superimposed triangles, to be further discussed below).

The soft spots are characterized by a degree of softness determined by the typical magnitude of c_α, $|c_\alpha|$, in its vicinity

(note that the local stiffness ω^2 of the potential energy landscape scales as $c_\alpha^{-1/2}$), which quantifies the collective potential energy barrier that should be overcome in order to induce a structural rearrangement (the barrier is proportional to ω^6 in the cubic approximation [19,28]). The LHC c_α, however, contains no information whatsoever about the *direction*, neither in the potential energy landscape of the glass nor in real space, in which the barrier is lowest. Consequently, an external driving force applied in a certain direction or a spontaneous thermal fluctuation that generates a local force in a certain direction, may or may not push a soft spot towards its activation barrier. In short, soft spots are expected to be tensorial, anisotropic objects that cannot be comprehensively described by scalar measures.

To demonstrate the tensorial, anisotropic nature of soft spots, let us focus again on Fig. 1, where four plastic rearrangement events are presented (triangles). Each of these correspond to the first plastic event of the same glass under external deformation applied in four *different directions*. In particular, we applied athermal quasistatic (AQS) [27,29,30] simple and pure shear

$$\mathcal{H}_{\rm simple} = \begin{pmatrix} 1 & \pm\gamma \\ 0 & 1 \end{pmatrix} \quad \text{and} \quad \mathcal{H}_{\rm pure} = \begin{pmatrix} 1 \pm \gamma/2 & 0 \\ 0 & 1 \mp \gamma/2 \end{pmatrix},$$

respectively, in both the positive and negative ($+$ and $-$) directions, where γ quantifies the amplitude of deformation. All four plastic events occurred at soft spots, i.e., regions of abnormally large LHC, but at four *different* ones. This clearly demonstrates that soft spots are tensorial, anisotropic objects that feature different coupling to deformation in different directions.

To develop a theory that goes beyond the scalar LHC as a structural predictor in glasses, we set out to calculate the linear response coupling of the LHC to external deformation. That is, given a certain globally imposed deformation $\mathcal{H}(\gamma)$, we aim at calculating analytically $dc_\alpha[\mathcal{H}(\gamma)]/d\gamma$ associated with $\mathcal{H}(\gamma)$. The structure of the differential operator $d/d\gamma$ reflects the intrinsically disordered nature of glasses; it is composed of two contributions [31], one representing the affine response of ordered systems $\partial/\partial\gamma$ and the other representing the additional nonaffine motions associated with disorder-induced forces, $-\mathcal{U}^{\gamma\prime} \cdot \mathcal{M}^{-1} \cdot \partial/\partial\boldsymbol{x}$, where the superscript γ is shorthand notation for $\partial/\partial\gamma$ and $\mathcal{U}^{\gamma\prime}$ are the mismatch forces (that drive the nonaffine motions). Operating with $d/d\gamma = \partial/\partial\gamma - \mathcal{U}^{\gamma\prime} \cdot \mathcal{M}^{-1} \cdot \partial/\partial\boldsymbol{x}$ on c_α, we obtain two terms to the same leading order in $\mathcal{M}^{-1}$ (the complete and exact result, including all orders in $\mathcal{M}^{-1}$, is presented in Ref. [27]),

$$
\begin{aligned}
dc_\alpha d\gamma \simeq &-\mathcal{U}^{\gamma\prime} \cdot \mathcal{M}^{-1} \cdot (\mathcal{U}^{\prime\prime\prime} \cdot \mathcal{M}^{-1} \cdot \mathcal{U}^{\prime\prime\prime} : \mathcal{M}^{-1}) \cdot (\boldsymbol{f}_\alpha \cdot \mathcal{M}^{-1}) \\
&- (\mathcal{U}^{\gamma\prime} \cdot \mathcal{M}^{-1} \cdot \mathcal{U}^{\prime\prime\prime} \cdot \mathcal{M}^{-1}) : (\mathcal{M}^{-1} \cdot \mathcal{U}^{\prime\prime\prime} \cdot \mathcal{M}^{-1} \cdot \boldsymbol{f}_\alpha).
\end{aligned}
\tag{2}
$$

Equation (2), valid for the largest values of $dc_\alpha/d\gamma$, shows that these emerge from a fourth power of $\mathcal{M}^{-1} \sim \omega^{-2}$ (scalingwise), coupled to the energy anharmonicity tensor $\mathcal{U}^{\prime\prime\prime}$, to the internal force vector $\boldsymbol{f}_\alpha$ and to the mismatch force vector $\mathcal{U}^{\gamma\prime}$. Note that similarly to c_α (see the expression

above), the existence of frustration-induced internal forces $\boldsymbol{f}_\alpha$—an intrinsic signature of glassy disorder—is essential for the emergence of abnormally large values of $dc_\alpha/d\gamma$. While the expression for $dc_\alpha/d\gamma$ [in Eq. (2) or its exact counterpart in Ref. [27]] is universal, the specific information regarding
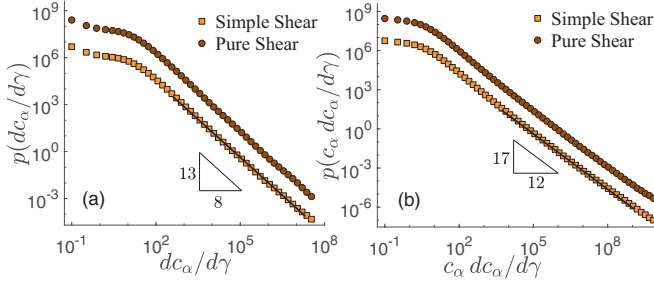
FIG. 2. The probability distribution functions (a) $p(dc_\alpha/d\gamma)$ and (b) $p(c_\alpha\, dc_\alpha/d\gamma)$ for both simple (squares) and pure (circles) shear deformation. The curves in each panel are vertically shifted one with respect to the other for visual clarity, while in fact they perfectly overlap, as expected from the initial glass isotropy. The theoretical power-law predictions are marked by the solid lines and the triangles (see text for details).

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 $\mathcal{U}^{\gamma\prime}$. The validity of the analytic expression for $dc_\alpha/d\gamma$ has been directly verified using numerical simulations [27].

Universal anomalous statistics. To further establish the linear responses $dc_\alpha/d\gamma$ 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, $D_G(\omega)\sim\omega^4$. Considering the eigenrepresentation of $dc_\alpha/d\gamma$ 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 ω dependence emerges only from $\mathcal{M}^{-1}\sim\omega^{-2}$. Consequently, we have $dc_\alpha/d\gamma\sim\omega^{-8}$ and $p(dc_\alpha/d\gamma)$ is predicted to satisfy $p(dc_\alpha/d\gamma)=D_G(\omega)d\omega/d(dc_\alpha/d\gamma)\sim(dc_\alpha/d\gamma)^{-13/8}$ in the large $dc_\alpha/d\gamma$ 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 $dc_\alpha/d\gamma$. The results are presented in Fig. 2(a) and are in great quantitative agreement with the theoretical prediction. We thus conclude that $dc_\alpha/d\gamma$ attains anomalously large values described by universal fat-tailed statistics related to the universal DOS of soft quasilocalized glassy modes, $D_G(\omega)\sim\omega^4$. The relation between $dc_\alpha/d\gamma$ 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_α is a signed scalar whose magnitude $|c_\alpha|$ 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 $dc_\alpha/d\gamma$ 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_α and $dc_\alpha/d\gamma$ 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 $c_\alpha\, dc_\alpha/d\gamma$ to single out a subpopulation of the soft spots (previously defined by $|c_\alpha|$) 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 $c_\alpha\, dc_\alpha/d\gamma$. As we have $c_\alpha\sim\omega^{-4}$ and $dc_\alpha/d\gamma\sim\omega^{-8}$ in the small ω limit, the spatial overlap prediction implies $c_\alpha\, dc_\alpha/d\gamma\sim\omega^{-12}$, which leads to $p(c_\alpha\, dc_\alpha/d\gamma)\sim(c_\alpha\, dc_\alpha/d\gamma)^{-17/12}$ in the large $dc_\alpha/d\gamma$ limit [using $D_G(\omega)\sim\omega^4$]. This prediction is quantitatively verified in Fig. 2(b) for both simple and pure shear, lending strong support to the idea that the product $c_\alpha\, dc_\alpha/d\gamma$ indeed characterizes well-defined soft spots.

We next turn to the spatial properties of $c_\alpha\, dc_\alpha/d\gamma$, and first consider the glass realization shown in Fig. 1, which is shown again in Fig. 3(a). The product $c_\alpha\, dc_\alpha/d\gamma$ 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 $c_\alpha\, dc_\alpha/d\gamma$ (the thickness of the lines quantifies their magnitude). Two major observations can be made: (i) Soft spots that are revealed by $c_\alpha\, dc_\alpha/d\gamma$ indeed overlap those revealed by c_α 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 $c_\alpha\, dc_\alpha/d\gamma$ 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 $c_\alpha\, dc_\alpha/d\gamma$ 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 ξ). A plastic event is assigned a rank λ that corresponds to the fraction of the bins with a higher value than that of the bin in which it
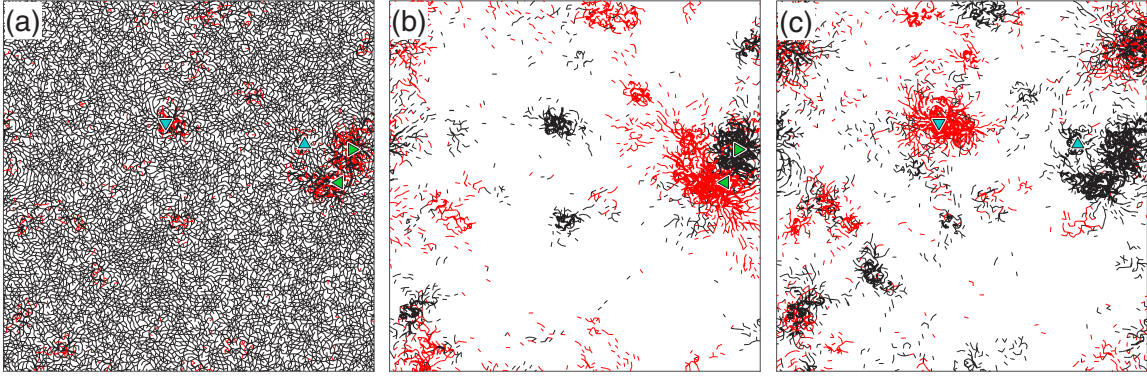
FIG. 3. (a) The same as Fig. 1. (b) $c_\alpha \, dc_\alpha/d\gamma$ [black (red) corresponds to positive (negative) values, and the line thickness represents the magnitude] for simple shear in the positive direction [the right (left) triangles correspond to the first plastic events under positive (negative) simple shear AQS deformation]. (c) The same as (b), but for pure shear in the positive direction [the up (down) triangles correspond to the first plastic events under positive (negative) pure shear AQS deformation].

actually occurred. The best prediction corresponds to $\lambda = 0$ (the event occurred in the highest value bin) and the worst one corresponds to $\lambda \to 1$ (the event occurred in the lowest value bin). When considering the cumulative distribution function $C(\lambda)$, with $0 \leqslant \lambda < 1$, perfect predictability corresponds to $C(\lambda) = \theta(\lambda)$ (Heaviside step function) and no predictability (random guess) corresponds to $C(\lambda) = \lambda$. This metric depends on a single, physically motivated parameter ξ (the quantitative dependence of the results on ξ is discussed in Ref. [27]).

The results are presented in Fig. 4, where $C(\lambda)$ for the absolute value of the LHC $|c_\alpha|$ serves as a reference (circles). In Fig. 4(a) we consider simple shear in the positive direction, and plot $C(\lambda)$ (diamonds) for positive values of $c_\alpha \, dc_\alpha/d\gamma$ (the negative ones are set to zero). It is observed that the predictive power of $c_\alpha \, dc_\alpha/d\gamma$ is significantly larger than that of $|c_\alpha|$. $C(\lambda)$ for negative values of $c_\alpha \, dc_\alpha/d\gamma$ (the positive

ones are set to zero) is also shown (squares), exhibiting essentially no predictive power, i.e., the curve is quite close to $C(\lambda) = \lambda$. Negative values of $c_\alpha \, dc_\alpha/d\gamma$ 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 $c_\alpha \, dc_\alpha/d\gamma$.

Concluding remarks. The results presented above show that $c_\alpha \, dc_\alpha/d\gamma$ 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 $c_\alpha \, dc_\alpha/d\gamma$, 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.
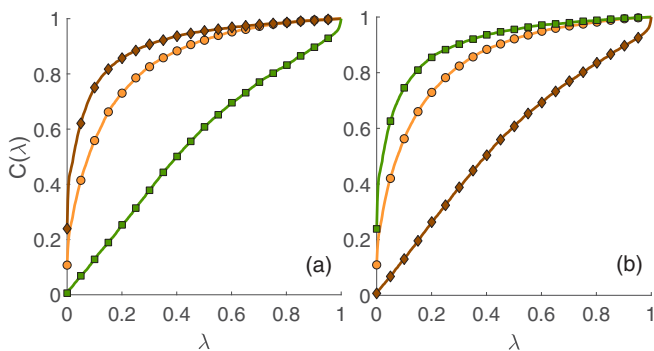

FIG. 4. Quantifying the predictive power of structural indicators with respect to plastic events under AQS deformation through the function $C(\lambda)$ (see text for definitions), where $C(\lambda) = \theta(\lambda)$ (Heaviside step function) corresponds to perfect predictability and $C(\lambda) = \lambda$ to no predictability. (a) Results for positive values (negative ones are set to zero) of $c_\alpha \, dc_\alpha/d\gamma$ (diamonds), for negative values (positive ones are set to zero) of it (squares) and for $|c_\alpha|$ (circles) under simple shear AQS deformation in the *positive* direction. (b) The same as (a), but under simple shear AQS deformation in the *negative* direction.

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