Thermodynamic interpretation of soft glassy rheology models

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Mesoscopic models play an important role in our understanding of the deformation and flow of amorphous materials. One such description, based on the shear transformation zone theory, has recently been reformulated within a nonequilibrium thermodynamics framework and found to be consistent with it. We show here that a similar interpretation can be made for the soft glassy rheology (SGR) model. Conceptually this means that the "noise temperature" x, proposed phenomenologically in the SGR model to control the dynamics of a set of slow mesoscopic degrees of freedom, can consistently be interpreted as their actual thermodynamic temperature. (Because such modes are slow to equilibrate, this generally does not coincide with the temperature of the fast degrees of freedom and/or heat bath.) If one chooses to make this interpretation, the thermodynamic framework significantly constrains extensions of the SGR approach to models in which x is a dynamical variable. We assess in this light some such extensions recently proposed in the context of shear banding.

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I. INTRODUCTION

Developing predictive theories for the deformation and flow of amorphous materials remains an ongoing challenge. The class of materials concerned is large, including not only colloidal glasses, emulsions, and foams but also molecular glass formers, metallic glasses, and possibly granular materials. The development of such predictive theories would therefore be of great practical significance as well as fundamental theoretical interest [1]. Two key challenges in understanding deformation and flow in amorphous materials are the absence of a reference configuration relative to which strain can be defined (in contrast to ordered crystalline solids), and the difficulty of encapsulating local flow events and/or larger scale stress redistributions in a coarse-grained variable or continuum field (analogous to the dislocation density for crystals).

First-principles approaches to glass rheology have made significant progress [2,3] but entail major approximations (such as mode coupling theory), usually followed by further simplification [4]. In the absence of a comprehensive microscopic theory, a number of models have been developed directly at a mesoscopic level to describe amorphous flow; see, e.g., [5–10]. Two such approaches that have been investigated in some detail are the shear transformation zone (STZ) approach, reviewed recently in [11], and the soft glassy rheology (SGR) model [12–15]. One important feature of the STZ theory is that it can be given an unambiguous thermodynamic interpretation, as discussed in detail in a series of recent papers [16–18].

Our purpose in this paper is to show that the SGR model can likewise be cast consistently within a nonequilibrium thermodynamic framework. This has two main benefits. First, it gives an alternative way of looking at the effective temperature parameter x that was previously proposed within

SGR to set the noise level for stochastic mesoscopic dynamics. This parameter can now, if desired, be interpreted as a genuine nonequilibrium thermodynamic temperature, governing a subset of degrees of freedom whose dynamics causes the system to move among its various "inherent structures" or energy minima. Second, this interpretation significantly constrains how SGR can be extended to allow x to evolve in time and space: It suggests a specific form for the driving term in the dynamical equation for an evolving x. We find that this driving term is consistent, in steady state, with one of two extensions of SGR postulated recently to understand shear banding in amorphous flows [15], but not with the other. Similar remarks apply to two models for shear-thickening fluids developed previously [19].

We begin by reviewing briefly the SGR model and then the nonequilibrium thermodynamic framework of Bouchbinder and Langer [16–18] (Secs. II and III). Section IV has the core of our argument, which shows that in this framework SGR can indeed be written as a thermodynamically consistent model, and that this consistency forces x to be the nonequilibrium thermodynamic temperature of a set of slow degrees of freedom. In Sec. V we discuss to what extent this argument can be made to cover also generalizations of the SGR model. Section VI has a summary of our results and discusses the implications for various extended SGR models that treat x as a dynamical variable.

II. THE SOFT GLASSY RHEOLOGY MODEL

The SGR model [12–15] describes a sample of amorphous material as a collection of mesoscopic elements, chosen large enough that it makes sense to define local strain and stress variables. The local strain l is defined relative to a local equilibrium state [20], and is assumed to change in time by following the change in the macroscopic shear strain γ, so that $\dot{l} = \dot{\gamma}$. We focus here on pure shear strain deformation but note

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that the model can also be extended to general deformation tensors [21]. The local strain cannot grow indefinitely: Once the stored elastic energy, which we write as $(kv_e/2)l^2$ in terms of the local shear modulus k and the element volume v_e, becomes close to a local yield energy E, a yield event will take place. This is assumed to reset l to zero and create a new local equilibrium configuration with correspondingly a new yield energy, drawn from a distribution $\rho(E)$. The final ingredient is that this yield process is taken as activated by some effective temperature x. This is based on the intuition that every yield event elsewhere in the material causes a "kick" locally via the associated stress redistribution, and that many such kicks add up to an effectively thermal noise. We assume here and below that x is spatially homogeneous, but we relax this assumption in some of the model extensions discussed in Sec. V.

The resulting equation of motion for the distribution of strain and yield energy across elements, $P(E,l,t)$, is

$$\dot{P}(E,l,t) = -\dot{\gamma}\partial_l P - \Gamma(E,l)P + Y(t)\rho(E,l). \quad (1)$$

Here $\Gamma(E,l)$ is the yield rate from a given local configuration, which represents the thermal activation in the form $\Gamma(E,l) = \Gamma_0 \exp[-(E - kv_e l^2/2)/x]$. [We will leave $\Gamma(E,l)$ general whenever possible in the following, to assess the scope of our arguments.] We use $k_B = 1$ throughout, so that all temperatures are measured in energy units. The time-dependent total yield rate is $Y(t) = \int dE\, dl\, \Gamma(E,l)P(E,l,t)$. Finally, $\rho(E,l)$ is the probability density of E and l for elements after a jump. This is equal to $\rho(E)\delta(l)$ in the standard SGR model as described above. By choosing $\rho(E)$ to have an exponential tail $\sim e^{-E/E_0}$ one finds that a glass transition arises at $x/E_0 = 1$. For $x/E_0 > 1$ the material is ergodic (albeit with infinite viscosity for $1 < x/E_0 < 2$) whereas for $x/E_0 < 1$ the material is a nonergodic amorphous solid with time-dependent (aging) material properties [14].

The SGR shear stress is an average over elements,

$$\sigma(t) = \int dE\, dl\, kl P(E,l,t) \quad (2)$$

with k the local shear modulus as defined above. Starting from some initial condition $P(E,l,0)$, this together with the evolution equation for $P(E,l,t)$ in principle determines the stress $\sigma(t)$ for any imposed shear history $\gamma(t)$.

III. NONEQUILIBRIUM THERMODYNAMICS FRAMEWORK

We next review the nonequilibrium thermodynamic framework of Bouchbinder and Langer [16–18]. The basic premise is that the degrees of freedom of the material can be divided up, conceptually, into two weakly interacting subsystems. The degrees of freedom in the configurational subsystem (C) encode which of the exponentially many local energy minima, known in the glass community as inherent structures, the system finds itself in; this subsystem is "slow" because at low temperature any motion between inherent structures has to be thermally activated or driven by external deformation. The kinetic-vibrational subsystem (K) gathers the remaining degrees of freedom that describe fast motion around these inherent structures. This subsystem is taken as strongly

coupled to a thermal reservoir (R) that sets the thermodynamic equilibrium temperature θ_R. The total internal energy of the system is then written as

$$U_{\text{tot}} = U_C(S_C,\Lambda) + U_K(S_K) + U_R(S_R),$$

where S_C, S_K, and S_R are the entropies of the configurational and kinetic-vibrational subsystems and of the thermal reservoir. In U_C, Λ denotes a set of internal state variables of the configurational subsystem. In the STZ context these are taken as concentrations of shear transformation zones in different orientations. For our application to SGR, we will take Λ as the distribution $P(E,l)$. Note that because this distribution already contains information on all local elastic strains, we do not include separately in U_C a state variable for strain as was done in [17,18].

The thermodynamic analysis is based on the first and second laws. The first law, i.e., energy conservation, takes the following form:

$$\dot{U}_C + \dot{U}_K + \dot{U}_R = V\sigma\dot{\gamma}. \quad (3)$$

The right-hand side is the external work rate that arises from shearing a system of volume V at shear rate $\dot{\gamma}$ against a shear stress σ. In terms of the subsystem entropies S_C, S_K, and S_R, and the corresponding temperatures $\chi = \partial U_C/\partial S_C|_\Lambda$ and $\theta = \partial U_K/\partial S_K$ [22], the first law can be rewritten as

$$\chi\dot{S}_C + \theta\dot{S}_K + \dot{U}_R - V\sigma\dot{\gamma} + \dot{\Lambda}\left.\frac{\partial U_C}{\partial \Lambda}\right|_{S_C} = 0. \quad (4)$$

We only write one Λ-derivative term here, as a shorthand for a sum over the relevant derivatives with respect to the state variables gathered in Λ.

The second law states that total entropy must remain constant or increase; i.e.,

$$\dot{S}_C + \dot{S}_K + \dot{S}_R \geqslant 0. \quad (5)$$

One can eliminate $\dot{S}_C$ from this using the first law and write $\dot{S}_R = \dot{U}_R/\theta_R$ to get

$$-\left(1 - \frac{\theta}{\theta_R}\right)\dot{U}_R - \left(1 - \frac{\chi}{\theta}\right)\left(\theta\dot{S}_K + \frac{\theta}{\theta_R}\dot{U}_R\right) + W \geqslant 0, \quad (6)$$

where

$$W = V\sigma\dot{\gamma} - \dot{\Lambda}\left.\frac{\partial U_C}{\partial \Lambda}\right|_{S_C}. \quad (7)$$

In [17] it was argued that time variations in U_R, S_K, and Λ are in principle arbitrary, and that the three terms on the left-hand side of (6) should therefore be separately nonnegative. To allow easier comparison with [16–18], we follow this reasoning here. However, it turns out that this separate nonnegativity is sufficient but not necessary for (6): in the Appendix we present a less restrictive but still sufficient set of conditions for nonnegative entropy production. Setting that aside and assuming separate nonnegativity of all three terms, one obtains the conditions $W \geqslant 0$ and

$$\dot{U}_R = -B(\theta_R - \theta), \qquad \theta\dot{S}_K + \frac{\theta}{\theta_R}\dot{U}_R = -A(\theta - \chi), \quad (8)$$

where A and B are positive but can depend on the state of the system, e.g., via the various temperatures. Our notation for A

follows Ref. [11], and we define B by analogy; the coefficients in [16–18] differ by factors of temperature. To simplify further, one assumes strong coupling between the thermal reservoir and the fast subsystem. In the corresponding limit of large B, one then obtains $\theta = \theta_R$, but this equality is approached in such a way that the product $B(\theta_R - \theta)$ stays finite. In fact it is only in this limit that (6) leads to (8), as discussed further in the Appendix.

One can now write the first law as

$$\chi \dot{S}_C = -(\theta \dot{S}_K + \dot{U}_R) + W. \qquad (9)$$

Because of the assumption of strong coupling between the reservoir and the fast subsystem, one can exploit $\theta = \theta_R$ to write the first term on the right-hand side as $-(\theta \dot{S}_K + \frac{\theta}{\theta_R} \dot{U}_R) = A(\theta - \chi)$, so

$$\chi \dot{S}_C = W + A(\theta - \chi). \qquad (10)$$

The upshot of this analysis [17,18] is thus twofold. First, one obtains an equation of motion for the configurational entropy; this also then determines the dynamics of the nonequilibrium temperature χ as discussed in Sec. VI. The second result is the constraint $W \geqslant 0$. In (10), W is the dissipation rate, i.e., the difference between the external work (rate) and the (rate of) increase in the free energy of the Λ degrees of freedom.

To go beyond the formal expression for W above requires further assumptions on the relation between the state variables Λ and the other configurational degrees of freedom [16–18]. The slow subsystem as a whole is described via $U_C(S_C, \Lambda) = U_\Lambda(\Lambda) + U_1(S_C - S_\Lambda(\Lambda))$ where $U_1(S_1)$ captures all the slow degrees of freedom other than Λ. Then

$$\left. \frac{\partial U_C}{\partial \Lambda} \right|_{S_C} = \frac{\partial U_\Lambda}{\partial \Lambda} - \frac{\partial U_1}{\partial S_1} \frac{\partial S_\Lambda}{\partial \Lambda}. \qquad (11)$$

One can now simplify using $\chi = \partial U_C / \partial S_C |_\Lambda = \partial U_1 / \partial S_1$. Assuming further that the number of internal state variables in Λ is much smaller than the overall number of slow degrees of freedom [16], one has $S_\Lambda \ll S_C$ and χ can be regarded as a function of this vast majority of configurational degrees only. The final expression for the dissipation rate is then

$$W = V \sigma \dot{\gamma} - \dot{\Lambda} \left(\frac{\partial U_\Lambda}{\partial \Lambda} - \chi \frac{\partial S_\Lambda}{\partial \Lambda} \right), \qquad (12)$$

and this shows that external work that is not dissipated is indeed stored in the free energy $U_\Lambda - \chi S_\Lambda$ of the internal state variables Λ.

IV. APPLICATION TO THE SGR MODEL

As mentioned earlier, in applying the above formalism to our SGR approach we take the distribution $P(E,l)$ as the set of internal state variables Λ. The arguments above rely on the number of degrees of freedom in Λ being much smaller than the overall number of degrees of freedom in the configurational subsystem. This can be ensured by initially considering, instead of $P(E,l)$, the total probability this distribution assigns to each of a finite number of bins covering the (E,l) plane. The number of bins is then sent to infinity more slowly than the system volume V in the thermodynamic limit of large V. We assume below that this limit has been taken appropriately and

write directly the version of the calculations in the large system limit, i.e., without binning.

Clearly for SGR one should choose as the internal energy associated with $\Lambda \equiv P(E,l)$,

$$U_\Lambda(\Lambda) = N_e \int dE \, dl \, P(E,l) u(E,l), \qquad (13)$$

where $N_e = V/v_e$ is the number of SGR elements with volume v_e each, and $u(E,l)$ is some function of the local yield energy and strain. The corresponding choice of entropy is less obvious. A natural proposal is $S_\Lambda(\Lambda) = -N_e \int dE \, dl \, P(E,l)[\ln P(E,l) - 1]$. However, it turns out that this choice does not give a thermodynamically consistent interpretation of the SGR model, essentially because every yield event resets $l = 0$ and so loses an infinite amount of entropy. We argue instead that, because the l dynamics is deterministic apart from jumps when rearrangements occur, these degrees of freedom are "slaved." Hence we should only consider the entropy of the distribution across yield energies. We also allow for a prior distribution (or density of states) $R(E)$ in the entropy, and take

$$S_\Lambda(\Lambda) = -N_e \int dE \, dl \, P(E,l) \left[\ln \frac{P(E)}{R(E)} - 1 \right], \qquad (14)$$

where $P(E) = \int dl \, P(E,l)$. The argument that the entropy should be independent of the l degrees of freedom is somewhat analogous to the observation made in STZ that in order to derive the standard STZ equations of motion one has to assume that the shear transformation zone orientation makes no contribution to the entropy [11].

To ensure that SGR fits into the thermodynamic framework described above, we need to show that with the above choices for U_Λ and S_Λ, one has $W \geqslant 0$ always, which means that the rate at which external work is performed is never less than the rate at which free energy is stored reversibly in the internal state variables $\Lambda \equiv P(E,l)$.

The inequality $W \geqslant 0$ has to hold at all times t, so we drop all time arguments in the following. If we unpack our shorthand notation, the contribution to W in (12) from the change of $P(E,l)$ becomes

$$\dot{\Lambda} \frac{\partial (U_\Lambda - \chi S_\Lambda)}{\partial \Lambda} \equiv \int dE \, dl \, \dot{P}(E,l) \frac{\delta (U_\Lambda - \chi S_\Lambda)}{\delta P(E,l)}, \qquad (15)$$

where the functional derivative on the right-hand side is

$$\frac{\delta (U_\Lambda - \chi S_\Lambda)}{\delta P(E,l)} = N_e \left[u(E,l) + \chi \ln \frac{P(E)}{R(E)} \right]. \qquad (16)$$

In combination with the external work, this gives for the dissipation rate

$$W = V \sigma \dot{\gamma} - N_e \int dE \, dl \, \dot{P}(E,l) \left[u(E,l) + \chi \ln \frac{P(E)}{R(E)} \right]. \qquad (17)$$

We now want to check that $W \geqslant 0$, for the SGR equation of motion (1). Inserting the latter into the expression above and

also expressing σ as an average over $P(E,l)$ from (2) shows

$$\frac{W}{N_e} = \int dE\, dl\, \left\{ \dot{\gamma} k v_e l P + \dot{\gamma} \partial_l P \left[u + \chi \ln \frac{P(E)}{R(E)} \right] \right.$$
$$\left. + (\Gamma P - Y\rho) \left[u + \chi \ln \frac{P(E)}{R(E)} \right] \right\}, \qquad (18)$$

where P, ρ, u are short for $P(E,l)$, $\rho(E,l)$, $u(E,l)$, respectively. Now integrate by parts over l:

$$\frac{W}{N_e} = \int dE\, dl\, \left\{ \dot{\gamma} P(k v_e l - \partial_l u) \right.$$
$$\left. + \chi(\Gamma P - Y\rho) \ln \frac{e^{u/\chi} P(E)}{R(E)} \right\}. \qquad (19)$$

Since the sign of $\dot{\gamma}$ is arbitrary, and so is the shape of $P(E,l)$, the first term must vanish pointwise; i.e., $\partial_l u = k v_e l$. This of course makes sense: The stress contribution of each element is just the strain derivative of its internal energy density. So $u(E,l) - k v_e l^2/2$ is a function of E only, and from the meaning of the yield energy we expect this function to be $-E$ itself so that $u(E,l) = k v_e l^2/2 - E$.

With the flow term thereby eliminated,

$$\frac{W}{N_e \chi Y} = \int dE\, dl\, \left(\frac{\Gamma P}{Y} - \rho \right) \ln \frac{e^{u/\chi} P(E)}{R(E)}. \qquad (20)$$

We now specialize to the conventional SGR model where $\rho(E,l) = \rho(E)\delta(l)$, $\Gamma(E,l) = \Gamma_0 e^{u/\chi}$, and the obvious prior to use in the definition of S_Λ is $R(E) = \rho(E)$. Also, the quantity

$$\pi(E,l) \equiv \Gamma(E,l) P(E,l)/Y \qquad (21)$$

is a normalized distribution by definition of Y. So we can express W as

$$\frac{W}{N_e \chi Y} = \int dE\, dl\, \pi(E,l) \ln \frac{\Gamma(E,l) P(E)}{\Gamma_0 \rho(E)}$$
$$- \int dE\, \rho(E) \ln \frac{e^{-E/\chi} P(E)}{\rho(E)}. \qquad (22)$$

To show that this is nonnegative we now use a cross-entropy inequality of the form

$$\int dz\, P(z) \ln[P(z)/Q(z)] \geqslant -\ln \int dz\, Q(z). \qquad (23)$$

This is valid if P is a probability distribution and Q is a measure (i.e., a probability distribution but not necessarily normalized to 1). The second term in W is already of the form (23). In the first term, we can get $\pi(E,l)$ inside the log by writing $P(E)P(l|E) = P(E,l)$; here $P(l|E)$ is the conditional distribution of l given E. This yields

$$\frac{W}{N_e \chi Y} = \int dE\, dl\, \pi(E,l) \ln \frac{Y\pi(E,l)}{\Gamma_0 P(l|E)\rho(E)}$$
$$+ \int dE\, \rho(E) \ln \frac{\rho(E)}{e^{-E/\chi} P(E)} \qquad (24)$$

$$\geqslant -\ln(\Gamma_0/Y) - \ln \int dE\, e^{-E/\chi} P(E) \qquad (25)$$

$$= \ln \frac{\int dE\, dl\, e^{(k v_e l^2/2 - E)/\chi} P(E,l)}{\int dE\, dl\, e^{-E/\chi} P(E,l)} \geqslant 0. \qquad (26)$$

This shows that the SGR model is thermodynamically consistent in the sense that its equation of motion ensures that the dissipation rate W is nonnegative as it should be. Note that this argument worked only because in writing down the yield rate $\Gamma(E,l)$ above we had already identified the SGR effective temperature x with the thermodynamic temperature χ of the slow degrees of freedom. This is an important conclusion, which we will develop in more detail in the next section.

We remark finally that the three inequalities used above to prove $W \geqslant 0$ (two cross-entropy ones, and the last one that the average of $\exp[k v_e l^2/(2\chi)]$ over any distribution is $\geqslant 1$) all become equalities if (and only if) $P(E,l) \propto \rho(E)e^{E/\chi}\delta(l)$. As is physically sensible, this is the (Boltzmann) steady state of the SGR model without flow, where $W = 0$. In all other situations, W will be positive.

V. CONSTRAINTS ON MORE GENERAL SGR-LIKE MODELS

The natural question arising from the results of the previous section is whether more general versions of the SGR equation of motion are thermodynamically consistent in the same sense.

A. Deviations from local linear elasticity

Consider first deviations from local linear elasticity, as contemplated, e.g., in the tensorial version of SGR [21]. These are simple to incorporate: One could have $u(E,l) = u(l) - E$, with the stress then the average of $u'(l)/v_e$ over all elements. As long as $u(l)$ has its global minimum $u(0) = 0$ at $l = 0$, the argument above goes through, and the model is thermodynamically consistent.

B. More general yield rates $\Gamma(E,l)$

One can also generalize the SGR model by allowing a more general dependence of the yield rates $\Gamma(E,l)$ on the state of the local element, i.e., E and l. (This was done, for instance, in [19] in the context of shear thickening models; we discuss the implications for that work in Sec. VI.) To understand the behavior of W in this case, we start from the still general expression (20), choosing again $R(E) = \rho(E)$ as the prior in the entropy:

$$\frac{W}{N_e \chi Y} = \int dE\, dl\, \pi(E,l) \ln \frac{e^{u/\chi} P(E)}{\rho(E)}$$
$$- \int dE\, \rho(E) \ln \frac{e^{-E/\chi} P(E)}{\rho(E)}. \qquad (27)$$

As before, one can now write $P(E) = P(E,l)/P(l|E) = Y\pi(E,l)/[\Gamma(E,l)P(l|E)]$ so that the first term becomes

$$\int dE\, dl\, \pi(E,l) \ln \frac{e^{u/\chi} P(E)}{\rho(E)}$$
$$= \int dE\, dl\, \pi(E,l) \ln \frac{Y\pi(E,l)e^{u/\chi}}{\Gamma(E,l)P(l|E)\rho(E)}. \qquad (28)$$

Similarly, the second term is

$$\int dE\, \rho(E) \ln \frac{\rho(E)}{e^{-E/\chi} P(E)}$$
$$= \int dE\, dl\, P(l|E)\rho(E) \ln \frac{\Gamma(E,l) P(l|E)\rho(E)}{e^{-E/\chi} Y \pi(E,l)}. \quad (29)$$

Putting these two together and isolating the cross-entropy terms gives

$$\frac{W}{N_e \chi Y} = \int dE\, dl\, [\pi(E,l) - P(l|E)\rho(E)] \ln \frac{\pi(E,l)}{P(l|E)\rho(E)}$$
$$+ \int dE\, dl\, \pi(E,l) \ln \frac{e^{u/\chi}}{\Gamma(E,l)}$$
$$+ \int dE\, dl\, P(l|E)\rho(E) \ln \frac{\Gamma(E,l)}{e^{-E/\chi}}. \quad (30)$$

To say something about necessary conditions on $\Gamma(E,0)$ first, consider $P(l|E) = \delta(l)$, so that $\pi(E,l) \propto \Gamma(E,0)P(E)\delta(l)$. Choose a $P(E)$ such that this is $\rho(E)[1 + \eta r(E)]\delta(l)$ with some small η and a so far unspecified function $r(E)$ satisfying $\int dE\, \rho(E)r(E) = 0$. Then the first contribution to W is $O(\eta^2)$. The second and third combine to

$$-\eta \int dE\, \rho(E)r(E) \ln \frac{e^{-E/\chi}}{\Gamma(E,0)}. \quad (31)$$

Unless the argument of the logarithm is a constant, one can arrange the sign of $r(E)$ so as to make the integral positive, and thus W negative to leading order in η. To avoid this contradiction to $W \geqslant 0$, $\Gamma(E,0)$ must be proportional to $e^{-E/\chi}$, say $\Gamma(E,0) = \Gamma_0 e^{-E/\chi}$. This argument shows that for thermodynamic consistency, the effective temperature x of SGR *must* be equal to the nonequilibrium thermodynamic temperature χ of the configurational subsystem, as promised in the previous section.

Turning next to l dependence of $\Gamma(E,l)$, we can write this as $\Gamma(E,l) = \Gamma_0 e^{-E/\chi} g(E,l)$ with $g(E,0) = 1$, and hence

$$\frac{W}{N_e \chi Y} = \int dE\, dl\, [\pi(E,l) - P(l|E)\rho(E)] \ln \frac{\pi(E,l)}{P(l|E)\rho(E)}$$
$$+ \int dE\, dl\, \pi(E,l) \ln \frac{e^{k v_e l^2/(2\chi)}}{g(E,l)}$$
$$+ \int dE\, dl\, P(l|E)\rho(E) \ln g(E,l). \quad (32)$$

The first term here is of cross-entropy form, while the second and third terms are (pointwise) nonnegative if $1 \leqslant g(E,l) \leqslant e^{k v_e l^2/(2\chi)}$. In summary, a sufficient condition for $W \geqslant 0$ is

$$\Gamma(E,l) = \Gamma_0 e^{-E/\chi} g(E,l) \quad \text{with} \quad 1 \leqslant g(E,l) \leqslant e^{k v_e l^2/(2\chi)}. \quad (33)$$

The first part is also necessary as shown above. Whether the second part is likewise necessary is not clear; i.e., there may be other $g(E,l)$ that also keep $W \geqslant 0$. But physically the sufficient condition given here is broad enough: The yield rate should increase with l^2; i.e., it should never go below its $l = 0$ value for given E; on the other hand the yield rate should not go up more quickly with l^2 than prescribed by the reduction in activation energy. If anything, one might expect the rate to

level off when the barrier $-u = E - k v_e l^2$ becomes small or even negative, for example

$$\Gamma(E,l) = \Gamma_0 \min(e^{u/\chi}, 1) \quad (34)$$

or

$$\Gamma(E,l) = \Gamma_0/(1 + e^{-u/\chi}). \quad (35)$$

It is clear from the above arguments that one could also allow a shear-rate dependence of the attempt rate Γ_0, e.g., replacing Γ_0 by $(\Gamma_0^2 + \dot{\gamma}^2)^{1/2}$ in the spirit of recent proposals [23] on how the relaxation time varies with shear rate. These more general forms can of course be combined with deviations from local linear elasticity as in the previous subsection, with the upper limit on $g(E,l)$ then $e^{u(l)/\chi}$ instead of $e^{k v_e l^2/(2\chi)}$.

C. Frustration

We had previously considered [13] the possibility that the local strain l might not relax fully to zero after a yield event because disordered stresses from the environment prevent this, and termed this effect "frustration." Mathematically, it would correspond to $\rho(E,l) = \rho(E)r(l|E)$ with $r(l|E) \neq \delta(l)$. For an SGR model extended in this way it is doubtful that an argument can still be made that l should be left out of the entropy S_Λ as l no longer has deterministic dynamics slaved to the last yield time of each element. If one nevertheless leaves S_Λ as before, then one can show that the extended model does not guarantee $W \geqslant 0$; i.e., there are choices for $P(E,l)$ which lead to $W < 0$. It is possible that the model would still be thermodynamically consistent because such distributions might not be accessible by the dynamics starting from reasonable initial conditions, but we have not been able to establish this.

VI. SUMMARY AND DISCUSSION

With plausible expressions for the entropy and internal energy of the $P(E,l)$ degrees of freedom, we have been able to show that the SGR model admits an interpretation consistent with the nonequilibrium thermodynamics framework of Bouchbinder and Langer. This interpretation requires that the effective temperature x, postulated phenomenologically within the SGR model, is in fact the thermodynamic temperature χ of a configurational subsystem describing mesoscopic degrees of freedom. (These degrees of freedom have slow energy transfer to the thermal bath of "fast modes" and hence their temperature is a nonequilibrium one.)

We believe this represents significant progress in understanding the conceptual foundation of the SGR model. The original empirical motivation for introducing x was (and remains) intuitively reasonable: an effective noise temperature arising ultimately from yield events throughout the sample. However a more solid formal grounding is now provided by the link to a thermodynamically defined temperature that governs the exploration of the system's inherent structures, through the dynamics of its mesoscopic degrees of freedom. These degrees of freedom are taken to be in thermal equilibrium with each other, but not with the rest of the system. If one accepts that, the thermodynamic interpretation gives new insights by showing, for example, that x should be viewed as intrinsic to the state of the system, and thereby to its history, as opposed to

being set directly by the current rate of external driving. (The latter would make x-mediated effects second order, and hence negligible, within linear response theory [24].)

Of course, the consistency of the thermodynamic interpretation of x does not make this the only possible interpretation of the SGR model. It would be perfectly defensible to believe that, in many soft glasses, mesoscopic degrees of freedom do not equilibrate sufficiently *even among themselves* for their nonequilibrium temperature to ever be defined. In that case, however, it remains useful to know that the empirical choice made for the noise-driven dynamics in the standard formulation of SGR is exactly equivalent to having such a temperature. Anyone convinced that such a temperature does not exist would probably wish to add terms to break this equivalence—which amounts to an implicit assumption of detailed balance among slow modes in the quiescent state.

We have further shown that the thermodynamic consistency extends from the original SGR model to more general versions that allow for nonlinear local elasticity. Likewise, a fairly broad class of yield rates $\Gamma(E,l)$ is allowed. This includes most physically plausible choices, but always maintaining the thermally (χ-)activated dependence on yield energy.

Beyond checking consistency, the thermodynamic framework gives an equation of motion (10) for the entropy S_C of the configurational subsystem; see the Appendix for a generalization of this. As discussed in [11], one can convert Eq. (10) to an equation of motion for the effective temperature (a more involved scenario is discussed in [25]). To see this, recall the decomposition we assumed for the energy of the slow subsystem, $U_C(S_C,\Lambda) = U_\Lambda(\Lambda) + U_1(S_C - S_\Lambda(\Lambda))$. The effective temperature $\chi = \partial U_C/\partial S_C|_\Lambda = \partial U_1/\partial S_1$ depends then on $S_1 = S_C - S_\Lambda$. Assuming as before that $S_\Lambda \ll S_C$ because Λ only contains a vanishing fraction of the slow degrees of freedom, χ becomes a function of $S_1 \approx S_C$ only. Conversely, χ then fully determines S_C, so that $\dot{S}_C = (\partial S_C/\partial\chi)\dot{\chi}$ and Eq. (10) becomes

$$C_V^{\text{eff}}\dot{\chi} = W + A(\theta - \chi). \qquad (36)$$

Here $C_{\text{eff}}^V(\chi)$ is an effective heat capacity at constant volume, and $A(\chi,\theta)$ is as before positive but otherwise unspecified; in principle, A could also depend on other quantities like $\dot{\gamma}$ and possibly even $P(E,l)$. What is interesting, however, is that the form of the driving term W is not negotiable; this is from (20)

$$\frac{W}{N_e\chi} = \int dE\, dl\, \Gamma(E,l)P(E,l)\ln\frac{e^{u/\chi}P(E)}{\rho(E)}$$

$$- Y\int dE\, \rho(E)\ln\frac{e^{-E/\chi}P(E)}{\rho(E)}. \qquad (37)$$

While not obvious from this expression, it is clear from the definition (7) that in steady state, where $\Lambda \equiv P(E,l)$ does not change, $W = V\sigma\dot{\gamma}$ is just the external work rate. This is the form that was assumed recently in exploring the extension of the SGR approach to understand shear banding: It was called "model 1" in [15]. Steady state flow curves of shear stress σ versus $\dot{\gamma}$ as predicted from (36) would therefore look exactly like those found in [15]. On the other hand, "model 2" from that paper, where the driving term for χ is taken as proportional to

the yield rate Y, is not thermodynamically consistent. We note that even with model 1, Eq. (37) predicts different transient behavior, because the driving term W generally differs from the external work rate $V\sigma\dot{\gamma}$. It would be interesting to explore in future work what effects this thermodynamically motivated form for W has on the predicted shear banding dynamics.

We can similarly assess the thermodynamic consistency of two extensions of the SGR model that were proposed some time ago to model shear thickening effects [19]. That paper considered two models. In one, the effective temperature $x \equiv \chi$ is taken as a function $x(\sigma)$ of the overall stress. The second model, in a more radical departure from the standard SGR picture, takes $x(l)$ as a function of the local strain in each element. The first case can feasibly be accommodated within the thermodynamic framework, at least if we consider situations where the effective temperature x, in its time evolution according to (36), has reached a steady state. Assuming that $\theta \ll x$ for simplicity, so that the effective temperature is much larger than the bath temperature, the steady state condition is $x = W/A = V\sigma\dot{\gamma}/A$. Given that A is only restricted to be positive but allowed to depend on, e.g., $\dot{\gamma}$ and σ, there are certainly choices for this dependence that would give a steady state x depending (only) on σ.

The second model in [19], where $x = x(l)$, is different. Having a temperature that differs from one element to the next is a significant departure from the idea of a single effective temperature for the slow configurational subsystem. Consistent with this, one can check that a generically chosen $x(l)$ will violate the conditions (33). This does not of itself prove $W < 0$, but we have checked that there are indeed conditions under which negative W does arise, e.g., for the specific (stepwise decreasing) form of $x(l)$ considered in [19]. Accordingly this second model of [19] is not consistent with a thermodynamic interpretation of the effective temperature as described here.

Finally, we should note again that our thermodynamic viewpoint for SGR was developed above by closely following the route first taken for STZ by Bouchbinder and Langer [16–18]. These authors used the second law to infer conditions on the dynamics of macroscopic variables. However these conditions, while both plausible and sufficient, are not in fact necessary: In the Appendix we describe a less restrictive, but still sufficient, set. The true value of thermodynamic consistency as a constraint on mesoscopic rheological models will not be known without a set of necessary, as well as sufficient, conditions for compliance of such models with the second law. It is possible that an explicitly model-independent formalism, such as that developed in [26], could prove useful in identifying such conditions.

It is worth reemphasizing also that the thermodynamic interpretation [16–18] adopted here is subject to a number of nontrivial assumptions. These include weak coupling between the slow configurational subsystem and the fast vibrational subsystem, and an assumption that the contribution of the order parameters Λ to the entropy of the slow subsystem is negligible. Where we have used the phrase "thermodynamic consistency" above, we mean consistency with thermodynamic constraints *as they result from these assumptions*. This

is also why we would not advocate rejecting out of hand the variants of the SGR model identified here as "thermodynamically inconsistent": They may well be consistent with thermodynamics per se, just not within the set of assumptions that we have taken from the work of Bouchbinder and Langer [16–18].

A further interesting direction for future work, beyond the study of more general thermodynamic constraints, will be to explore thermodynamic effects on the aging behavior predicted by the SGR model. An important feature of the model is that aging effects are present [13,14] even when χ is constant, as long as its value lies below the glass transition at $\chi = E_0$. It is clear that allowing χ to evolve dynamically according to (36) in line with the thermodynamic picture will contribute additional aging effects. However, what quantitative form these take can only be fully assessed once physically motivated parameter dependencies have been established for the effective heat capacity C_V^{eff} and the coefficient A determining the rate of heat transfer between the slow and fast subsystems.

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APPENDIX: MORE GENERAL CONDITIONS FOR SECOND LAW

We have in the main part of the paper adopted the argument of [17] that the three terms in (6) must be separately nonnegative. This is a sufficient condition for the overall sum to be nonnegative and is also physically plausible. However it is not mathematically necessary. We discuss here a somewhat more general set of conditions that are also sufficient. These admit the thermodynamic consistency of a wider class of models than allowed by the conditions of [17] as used above.

If we write the first law as in (4) and insert $\dot{U}_R = \theta_R \dot{S}_R$, we obtain

$$\chi \dot{S}_C + \theta \dot{S}_K + \theta_R \dot{S}_R = W. \tag{A1}$$

This needs to be satisfied together with the second law

$$\dot{S}_C + \dot{S}_K + \dot{S}_R \geqslant 0. \tag{A2}$$

With the reversible degrees of freedom (Λ) of the slow system already treated separately in W, these two equations are completely symmetric in the roles played by the slow, fast, and reservoir subsystems. A more general set of conditions for the second law to hold then suggests itself as $W \geqslant 0$ and

$$\chi \dot{S}_C = \alpha_C W + A(\theta - \chi) + C(\theta_R - \chi), \tag{A3}$$

$$\theta \dot{S}_K = \alpha_K W + A(\chi - \theta) + B(\theta_R - \theta), \tag{A4}$$

$$\theta_R \dot{S}_R = \alpha_R W + B(\theta - \theta_R) + C(\chi - \theta_R), \tag{A5}$$

where A, B, C, α_C, α_K, α_R are all nonnegative and $\alpha_C + \alpha_K + \alpha_R = 1$. The last condition is enough to ensure that the first law is satisfied, as the heat flow terms all cancel in pairs. For the total rate of entropy change one gets

$$\dot{S}_C + \dot{S}_K + \dot{S}_R = W \left(\frac{\alpha_C}{\chi} + \frac{\alpha_K}{\theta} + \frac{\alpha_R}{\theta_R} \right) + A \frac{(\chi - \theta)^2}{\chi \theta}$$
$$+ B \frac{(\theta - \theta_R)^2}{\theta \theta_R} + C \frac{(\chi - \theta_R)^2}{\chi \theta_R}, \tag{A6}$$

which is evidently nonnegative. The more restrictive condition of [17] corresponds essentially to $\alpha_K = \alpha_R = C = 0$. One has then $\dot{U}_R = \theta_R \dot{S}_R = -B(\theta_R - \theta)$ as in (8), and

$$\theta \dot{S}_K + \frac{\theta}{\theta_R} \dot{U}_R = A(\chi - \theta) + \frac{B}{\theta_R}(\theta_R - \theta)^2. \tag{A7}$$

This generalizes the second equality of (8) to the case where $\theta \neq \theta_R$. In the limit where B becomes large at constant $B(\theta_R - \theta)$, the term on the right-hand side proportional to B goes to zero so (8) is retrieved. That a term proportional to $B(\theta_R - \theta)^2$ should be present in general here is clear because $\theta \dot{S}_K + (\theta/\theta_R)\dot{U}_R = \theta(\dot{S}_K + \dot{S}_R)$ and this rate of entropy change should have a nonzero contribution when there is heat transfer at a finite rate between the reservoir and the kinetic-vibrational subsystem.

Returning to our more general conditions above, the effect of the coefficient C is probably unimportant so long as we are indeed interested only in the limit $\theta = \theta_R$ because of strong thermal coupling to the reservoir: One can then just combine A and C.

The effect of the α coefficients is more subtle. In soft materials, where the reservoir should include the solvent degrees of freedom, it seems plausible that a substantial amount of the work performed on the system (minus the amount stored reversibly in Λ, which gives W) could be dissipated directly in the solvent. For instance, in a dense emulsion, work could be dissipated directly in the solvent films between droplets without ever passing through the Λ degrees of freedom. In soft glasses, therefore, α_R could be substantial, and correspondingly $\alpha_C < 1$. An alternative strategy might be to exclude this contribution from the work input, by separating off a global Newtonian solvent contribution. However, as the example of thin films between emulsion droplets shows, the mesoscopic and solvent contributions cannot be treated as simply additive: In principle the droplet organization controls both the thicknesses of films and the shear rates within them. It therefore seems advisable not to assume that kind of separation within a thermodynamic description, at least in the case of soft glasses.

Despite the above generalizations the condition that $W \geqslant 0$ remains in place. So far we have not been able to show mathematically that $W \geqslant 0$ is necessary (or indeed that there are not broader conditions on the heat flows that would satisfy the second law), although physically it is certainly plausible that the dissipation rate should be nonnegative. The main

possible generalization arising from the arguments above is then in the equation of motion for χ, which would become [replacing (36)]

$$C_V^{\mathrm{eff}} \dot{\chi} = \alpha_{\mathrm{C}} W + A(\theta - \chi). \tag{A8}$$

Here one now has an additional undetermined factor $0 \leqslant \alpha_{\mathrm{C}} \leqslant 1$ in front of the driving term W. One could speculate whether there is a difference between, e.g., metallic and soft glasses in the size of α_{C}, which could be of order unity for the former and small for the latter.

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