Glass Dynamics at High Strain Rates

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We present a shear-transformation-zone (STZ) theoretical analysis of molecular-dynamics simulations of a rapidly sheared metallic glass. These simulations are especially revealing because, although they are limited to high strain rates, they span temperatures ranging from well below to well above the glass transition. With one important discrepancy, the simplified STZ theory used here reproduces the simulation data, including the way in which those data can be made to collapse approximately onto simple curves by a scaling transformation. The STZ analysis implies that the system’s behavior at high strain rates is controlled primarily by effective-temperature thermodynamics, as opposed to system-specific details of the molecular interactions. The discrepancy between theory and simulations occurs at the lower strain rates for temperatures near the glass transition. We argue that this discrepancy can be resolved by the same multi-species generalization of STZ theory that has been proposed recently for understanding frequency-dependent viscoelastic responses, Stokes-Einstein violations, and stretched-exponential relaxation in equilibrated glassy materials.

I. INTRODUCTION

A remarkable scaling property of viscosity as a function of shear stress has been observed by by Olsson and Teitel [1] in simulations of a strongly overdamped, athermal, two-dimensional, amorphous system near its jamming transition. More recently, an approximately similar scaling behavior has been seen by Guan, Chen and Egami [2] (GCE) in molecular dynamics simulations of a rapidly sheared, three dimensional metallic glass that undergoes a thermodynamic glass transition. These results suggest that there may be a useful, minimal description of the underlying physics for the seemingly complex dynamics of flow in glass-forming fluids, and that some scaling properties of a zero-temperature jamming transition may persist in rapidly deforming systems far from jamming conditions.

The distinguishing feature of the GCE simulations is that they cover an unusually wide range of system parameters. They are carried out at kinetic temperatures ranging from 100 $K$ to 1100 $K$, i.e., from well below to well above an apparent glass transition. Importantly, they are carried out at strain rates from $10^8$ s$^{-1}$ to $10^{12}$ s$^{-1}$, all well above values ordinarily accessible in the laboratory. By driving their systems at large strain rates, GCE achieve steady-state, nonequilibrium shear flows at temperatures far below the glass transition. Under stress, molecular rearrangements become faster and the internal disorder increases, while the kinetic temperatures remain low. GCE are not probing truly glassy behavior in this way; however, they are discovering important features of glass dynamics by looking from this point of view.

In this paper, we explore the implications of the GCE results by comparing them with the predictions of a shear-transformation-zone (STZ) theory [3-5]. This comparison is timely because Voigtmann [6] recently has published an analysis of the same data using an extended version of mode-coupling theory (MCT) [7-9]. These two theories of amorphous plasticity would seem at first sight to have non-overlapping regions of validity.

MCT starts with a liquidlike description of a many-body system. Its strength is that the coupling terms that emerge from its approximate closure of the many-body equations of motion can be evaluated in terms of observable structure factors, thus basing the theory directly upon microscopic dynamics. Its weakness, however, is that this closure approximation is accurate only if the density fluctuations are distributed Gaussianly. Therefore, MCT is effectively a mean-field theory in which motions are determined by averaging over large numbers of weakly correlated events. Such an approximation becomes qualitatively incorrect at low temperatures, where the flow is governed by sporadic, thermally activated events that are rare but intense [10]. Voigtmann’s results may imply that MCT retains some phenomenological validity for flowing states in the low-temperature regime; but they cannot in principle have the same fundamental validity that MCT can claim at higher temperatures.

In contrast, the STZ theory starts with a solidlike picture in which thermally activated, localized flow defects play the central role in controlling plastic deformation. It predicts that, at low temperatures, the yielding transition as a function of stress is an exchange of dynamic stability between jammed and flowing states; and it describes the mechanical behavior of the system on both sides of that transition. When combined with an equation of motion for the effective disorder temperature of the configurational (i.e. structural) degrees of freedom, the STZ theory successfully predicts shear-banding instabilities in agreement with simulations [11, 12] and, as will
We start, in Section II, by presenting the steady-state STZ equations of motion in a more general and compact form than the one that has appeared in previous publications \[3–5\]. Then, in Sec. III, we summarize the primarily kinematic arguments that lead to our equation of motion for the effective temperature. In Sec. IV, we describe the ways in which we use the simulation data to evaluate the theoretical parameters. Section V is devoted to our interpretation of the GCE scaling analysis. We conclude in Sec. VI with a discussion of open questions.
rate at which the system undergoes structural aging. In much of the literature, the quantity $\rho(\theta) \exp\left(-1/\theta\right)/\tau_0$ is defined to be the $\alpha$ relaxation rate $\tau_\alpha^{-1}$. However, as pointed out in [14], this definition is not the same as the conventional assumption that $\tau_\alpha$ is directly proportional to the viscosity.

In analogy to $\rho(\theta)/\tau_0$, the quantity $\Gamma(s, \chi)/\tau_0$ is the contribution to the attempt frequency in Eq. (2.2) due to mechanically generated noise. As in earlier papers [3, 19, 20], we assume that $\Gamma(s, \chi)/\tau_0$ is proportional to the rate of entropy production. In steady flow, all of the work done on the system is dissipated as heat; therefore, the rate of energy dissipation per unit volume is $2 \gamma \dot{\varepsilon}$. To convert this rate to a noise strength with dimensions of energy conveniently written in the form $2 \dot{\varepsilon}$, we multiply by the volume per noise source, i.e. the volume per STZ, $v_0 \exp(1/\chi)$, and divide by an energy conveniently written in the form $2 \epsilon_0 s_0 v_0$. The fact that the stress $s_0$, introduced here for dimensional reasons, turns out to be the low-temperature yield stress has been one of the more interesting surprises in this theory. The resulting formula for the noise strength is

$$\Gamma(s, \chi) = \frac{q s}{\epsilon_0 s_0} e^{1/\chi}. \quad (2.5)$$

The GCE simulations involve only steady-state deformations; thus all of the information that we need is contained in the stationary solutions of Eq. (2.1), obtained by setting $\dot{n}_\chi = 0$ and solving for the $n_\pm$. The resulting expression for the strain rate in Eq. (2.2) is

$$q = \epsilon_0 \tilde{\gamma}(s, \chi) e^{-1/\chi} \left[ \frac{2C(s)}{2C(s) + \tilde{\gamma}(s, \chi)} \right] T(s, \chi), \quad (2.6)$$

where

$$C(s) = \frac{1}{2} \left[ R(+s) + R(-s) \right], \quad (2.7)$$

and

$$T(s, \chi) = \frac{R(+s) - R(-s)}{R(+s) + R(-s)}. \quad (2.8)$$

According to Eq. (2.8), $T(s, \chi)$ is the bias between forward and backward transitions. The second law of thermodynamics (see [13]) requires that

$$T(s, \chi) = \tanh \left( \frac{v_0 s}{\epsilon Z \chi} \right). \quad (2.9)$$

With Eq. (2.9), Eq. (2.10) becomes a quadratic equation that can be solved for $q$ or, more conveniently, for $\Gamma(s, \chi)$. The result is

$$\Gamma(s, \chi) = \frac{1}{2} Q(s, \chi) + \frac{1}{2} [Q(s, \chi)^2 + 4 \rho(\theta) Q(s, \chi)], \quad (2.10)$$

where

$$Q(s, \chi) = 2C(s) T(s, \chi) s - 2C(s) - \rho(\theta). \quad (2.11)$$

Then, knowing $\Gamma(s, \chi)$, we can use Eq. (2.5) to write:

$$q = \frac{\epsilon_0 s_0}{s} e^{-1/\chi} \Gamma(s, \chi). \quad (2.12)$$

Note that $\Gamma(s, \chi)$ is a non-negative, symmetric function of $s$ that vanishes like $s^2$ at $s = 0$.

In the athermal limit where $\rho = 0$, both $\Gamma(s, \chi)$ and $q$ vanish for $s < s_y$ (the jammed state); whereas, for $s > s_y$ (the flowing state),

$$q = 2 \epsilon_0 e^{-1/\chi} C(s) \left[ \frac{T(s, \chi) - s_0}{s} \right]. \quad (2.13)$$

The yield stress $s_y$ is the solution of

$$s_y T(s_y, \chi) = s_0. \quad (2.14)$$

Therefore, $s_y \equiv s_0$ when $s_y$ is large and $T(s_y, \chi) \approx 1$. For small but nonzero $\rho$, according to Eq. (2.11), both $\Gamma(s, \chi)$ and $q$ make smooth transitions near $s = s_y$ between viscous and flowing states.

We also need to examine the viscous limit at small $q$, where $\Gamma(s, \chi) \ll \rho$ and $\chi \approx \theta$. Keeping only the linear term in the relation between $q$ and $s$, we find

$$q = \epsilon_0 \rho(\theta) e^{-1/\theta} \left( \frac{2C(0)}{2C(0) + \rho(\theta)} \right) \frac{v_0 s}{\epsilon Z \theta}. \quad (2.15)$$

The Newtonian linear viscosity, for vanishingly small $q$, and expressed here in units of stress, is $\eta_N = s_0$. Note that this formula produces the conventional result in which $\eta_N \propto \tau_\alpha$ multiplied by a slowly varying function of $\theta$. It is this result that, according to [14], substantially underestimates $\eta_N$ at low temperatures and, therefore, is responsible for the important discrepancy between this theory and the GCE simulations.

$C(s)$ is necessarily a symmetric function of $s$. We write it in the form:

$$C(s) = \exp \left[ -\frac{\theta_E}{\theta} e^{-s^2/2 \theta_E^2} \right] \left[ 1 + \left( \frac{s}{s_B} \right)^2 \right]^{1/4}. \quad (2.16)$$

On the right-hand side, the first factor in square brackets is a thermally activated (Eyring-like) rate in which a barrier of height $\theta_E$ is reduced quadratically by the applied stress via a Gaussian factor, thus preserving the required symmetry. In what follows, we find that we can approximate this term by unity. The second factor is similar to, but not strictly the same as a Bagnold rate, proportional to the square root of the stress at large $s$ for dimensional reasons. It does seem to be significant at low temperatures and high strain rates. The only quantity that can play the role of $s_B$ in this formula is the pressure, which may be roughly proportional to the temperature. Thus, we write $s_B = \left( T/T_B \right) GPa$, where the “Bagnold temperature” $T_B$ is a constant.
Turn now to the effective temperature $\chi$. In general, $\chi$ is the temperature of the configurational degrees of freedom of the system, thus it characterizes the system's state of structural disorder. Its role here is primarily to determine the density of STZ’s, as indicated by its appearance in the Boltzmann factor in Eq. (2.3). We propose that $\chi$ is determined mostly by kinematics rather than by specific details of the molecular interactions, and that this property of $\chi$ is largely responsible for the universality observed at high strain rates.

At low temperatures, where changes in the state of glassy configurational disorder can be induced only by externally driven deformation and not by thermal fluctuations, i.e. where $\rho = 0$, there must be a direct relation between the dimensionless strain rate $q$ and the low-temperature effective temperature, say $\chi = \tilde{\chi}(q)$. When the strain rate is much smaller than any relevant relaxation rate in the system, so that $q \ll 1$, then dimensional analysis based on the fact that there is no intrinsic rate comparable to $q/\tau_0$ requires that $\tilde{\chi}(q)$ be equal to a constant, say $\tilde{\chi}_0$.

In the opposite limit, when $q$ is of the order of unity, we assume that the relation between $q$ and $\tilde{\chi}$ has an Arrhenius form, $q \sim q_0 \exp(-A/\tilde{\chi})$. Both $\tilde{\chi}$ and $A$ are energies measured in units of $e_k$. If $e_k$ is the only energy scale in the system relevant to configurational rearrangements, then we expect that $A$ is of the order of unity. Similarly, if $\tau_0^{-1}$ is the only intrinsic rate, then $q_0$ also should be of the order of unity; but here there is an additional uncertainty about how accurately we have estimated $\tau_0$. We return to these estimates in Sec. [IV].

The relation between strain rate and configurational disorder was discovered in numerical simulations by Haxton and Liu [22]. It was discussed further in [21], where it was written in the form of a glasslike relation between a “viscosity” $q^{-1}$ and the temperature $\tilde{\chi}$:

$$\frac{1}{q} = \frac{1}{q_0} \exp \left[ \frac{A}{\tilde{\chi}} + \alpha_{\text{eff}}(\tilde{\chi}) \right]; \quad (3.1)$$

and, in analogy to Vogel-Fulcher-Tamann (VFT),

$$\alpha_{\text{eff}}(\tilde{\chi}) = \frac{\tilde{\chi}_1}{\tilde{\chi} - \tilde{\chi}_0} \exp \left[ -b \frac{\tilde{\chi} - \tilde{\chi}_0}{\tilde{\chi}_A - \tilde{\chi}_0} \right]. \quad (3.2)$$

Thus, $\tilde{\chi} \to \tilde{\chi}_0$ in the limit of small strain rates, and $\tilde{\chi} \to \infty$ as $q \to q_0$. The exponential cutoff in Eq. (3.2) is needed in order that the VFT divergence at small $\tilde{\chi}$ transform smoothly to the Arrhenius law at large $\tilde{\chi}$. Previous calculations have used $b = 3$. Experience with these formulas, as in [21], leads us to conclude that they are more cleanly reliable than their VFT analog for viscosity as a function of ordinary temperature. (In this connection, see the calculation of the viscosity in [14].)

The equation of motion for $\chi$ itself is a statement of the first law of thermodynamics; it describes entropy flow through the slow, configurational degrees of freedom into the fast thermal motions of the molecules. Near steady state, it has the form

$$\dot{\chi} \propto e^{-1/\chi} \left[ \Gamma(s, \chi) \left( \tilde{\chi}(q) - \chi \right) + \kappa \rho(\theta) (\theta - \chi) \right]. \quad (3.3)$$

The first term in square brackets on the right-hand side is the rate at which $\chi$ is driven toward $\tilde{\chi}(q)$ by the mechanical noise strength $\Gamma(s, \chi)$. The second term, proportional to $\rho(\theta)$, is the rate at which thermal fluctuations drive $\chi$ toward the ambient temperature $\theta$. $\kappa$ is a dimensionless parameter of the order of unity. Since $\chi$ is a measure of configurational disorder, we see here explicitly how $\rho(\theta)$ controls the rate of structural aging in undriven systems. The competition between these two terms in Eq. (3.3) determines the value of $\chi$: it is close to $\tilde{\chi}(q)$ for large $\Gamma(s, \chi)$, and close to $\theta$ when the system is driven slowly so that $\Gamma(s, \chi)$ is small.

There is one complication that must be dealt with at this point. Equation (3.3), as written, implies that the steady-state $\chi$ must lie in the interval between $\tilde{\chi}(q)$ and $\theta$. If we assume that $\tilde{\chi}(q) \approx \tilde{\chi}_0$ is a constant for small enough $q$, then a system initially prepared with $\theta > \tilde{\chi}_0$ would be “cooled” to $\chi < \theta$ when driven at a small strain rate. This behavior seems implausible; so far as we know, it is not seen in other simulations, e.g. [20], in which $\chi$ is measured directly. In [21], this problem was corrected by setting $\tilde{\chi}_0 = \theta$ when $\theta$ exceeds $\chi_0$, and by rescaling $\tilde{\chi}_1$ and $\tilde{\chi}_A$ accordingly. Specifically,

$$\tilde{\chi}_0 = \begin{cases} \chi_0, & \text{for } \theta < \chi_0, \\ \theta, & \text{for } \theta > \chi_0 \end{cases}; \quad (3.4)$$

$$\tilde{\chi}_1 = \begin{cases} \chi_1, & \text{for } \theta < \chi_0, \\ (\chi_0/\chi_0) \chi_1, & \text{for } \theta > \chi_0 \end{cases}; \quad (3.5)$$

and

$$\tilde{\chi}_A = \begin{cases} \chi_A, & \text{for } \theta < \chi_0, \\ (\chi_0/\chi_0) \chi_A, & \text{for } \theta > \chi_0 \end{cases}. \quad (3.6)$$

In summary, we use Eq. (3.3) in the form

$$\chi = \frac{\Gamma(s, \chi) \tilde{\chi}(q) + \kappa \rho(\theta) \theta}{\Gamma(s, \chi) + \kappa \rho(\theta)}, \quad (3.7)$$

along with Eq. (3.3)], to determine one relation between $\chi$, $q$ and $s$. We then use Eq. (2.12) to compute both $q$ and $\chi$ as functions of $s$.

IV. EVALUATION OF PARAMETERS AND COMPARISON WITH THE GCE DATA

In Fig. 11, we show a selection from the raw data of GCE. The data sets shown here are characteristic of the eleven such sets that we have used in these analyses. They are stress versus strain-rate curves for seven different temperatures, $T =$
FIG. 1: (Color online) Log-log plot of stress $s$ versus dimensionless plastic strain rate $q = \tau_0 \dot{\varepsilon}_p$. The top three solid black theoretical curves and the associated open-square data points are for temperatures $T = 100\, K$, $300\, K$, and $500\, K$. The middle dashed blue curves are for $T = 700\, K$ (blue circles), and $840\, K$ (blue triangles). The bottom two solid red theoretical curves and open-circle data points are for $T = 940\, K$ and $1100\, K$. Agreement between theory and simulation is good for the low temperatures (black curves) at the top and for the high temperatures (red curves) at the bottom; but it fails at small strain rates for the intermediate temperatures near the glass transition.

100 $K$, 300 $K$, 500 $K$, 700 $K$, 840 $K$, 940 $K$ and 1100 $K$, reading from top to bottom. The data points are shown here along with our best-fit theoretical curves. The GCE time constant is $\tau_0 = 0.1$ picoseconds; therefore, the dimensional strain rate is $\dot{\varepsilon}_p = 10^{13}\, q\, s^{-1}$. We have arbitrarily chosen the upper limit of validity for the STZ theory to be at $q = 10^{-2}$, i.e. at $\dot{\varepsilon}_p = 10^{11}\, s^{-1}$. Beyond this point, the simulations show strain-rate softening – possibly an indication that the system is liquefying and is no longer consistent with the solidlike STZ model.

Our strategy for choosing parameters has been to look for the simplest possible version of the theory that is consistent with the data. Accordingly, our starting assumption is that the Eyring-like rate factor is negligible; that is, we set $\theta_E = 0$ in Eq. (2.10). We have tried various nonzero values of $\theta_E$, and have found no overall improvement in the results. At low temperatures, only large stresses – above the yield stress – come into play, reducing the activation barrier via the Gaussian factor in Eq. (2.10). Conversely, at high temperatures, the driving stresses may be small, but the activation barriers are small compared to $k_B T$. The exceptions are at the intermediate temperatures, for small stresses and small strain rates, where the theory breaks down for the more interesting reasons mentioned in Sec. II. If we choose $\theta_E$ large enough that, just by itself, it substantially increases the viscosity and improves the fit, for example, at $T = 840\, K$ (the lower dashed curve in Fig. 1), then we qualitatively ruin the fit at $T = 1100\, K$. Ultimately, the Eyring barrier must be important at smaller strain rates and low temperatures; but we see no need for it here.

The dimensionless constants $\epsilon_0$ in Eq. (2.2) and $\kappa$ in Eq. (3.3) should be of the order of unity. In the absence of better information, we set $\epsilon_0 = \kappa = 1$. Similarly, we set $\epsilon_2 / \epsilon_0 = 1$ GPa and, in the next paragraph, use the high-temperature viscosity to check that this estimate is reasonable.

At high temperatures and small stresses, with $C(s) \approx \epsilon_0 \epsilon_2 / \epsilon_2 \approx 1$, Equation (2.13) tells us that the linear Newtonian viscosity is

$$\eta_N = \frac{2 + \rho(\theta)}{2 \rho(\theta)} \theta e^{1/\theta} \approx \frac{3}{2} \theta e^{1/\theta}. \quad (4.1)$$

The last approximation is valid for temperatures high enough that $\rho \approx 1$. At $T = 1100\, K$ (the bottom curve in Fig. 1), the GCE result is $\eta_N = 150$. Then, Eq. (4.1) implies that $\theta = 0.15$, and therefore that $T_Z = \epsilon_2 / k_B \approx 7000\, K$. This means that $\epsilon_2 / \epsilon_2 \approx 10 \, \epsilon_2$, with $\epsilon_2$ measured in cubic nanometers. Thus, a length scale of about half a nanometer gives us $\epsilon_2 / \epsilon_2 \approx 1$ GPa as in the preceding paragraph.

Now consider low-temperature situations for which the GCE data appear to indicate well defined yield stresses $s_0(\theta)$, implying that $\rho(\theta) = 0$ to within the accuracy of the measurements. The temperatures for which this is true are $T = 100\, K$, $300\, K$, $500\, K$ and $600\, K$. In this regime, $T(s, \chi) \approx 1$; thus, in the limit of small $q$, Eq. (2.13) becomes

$$q \approx 2 e^{1/\chi_0} \left(1 - \frac{s_0}{s}\right), \quad (4.2)$$

which we have used to make a first estimate of $\chi_0$. We then have found that we can fit all four of these data sets, for the whole range of strain rates $10^{-5} < q < 10^{-2}$, by using the observed values of $s_0(\theta)$, and by choosing $\chi_0 = 0.1$, $\chi_A = \chi_A = 0.2$, $A = 1.3$, $q_0 = 5$, and $b = 3$ in Eqs. (3.1) and (3.2). We also have chosen the Bagnold term defined following Eq. (2.10) to be $T_B = 100\, K$. With this value of $T_B$, the Bagnold term is negligible for all but the lowest temperatures and largest stresses; but it does seem to be relevant in that regime. We have no reason to believe that this set of temperature-independent parameters is unique or optimal.

The remaining parameters to be chosen are the quantities $\rho(\theta)$ and $s_0(\theta)$ for temperatures near and above the glass transition. Rather than trying to predict these quantities theoretically, we have “measured” them by fitting the GCE data. The results are shown below in Figs. 3 and 4 and are discussed in Sec. V.

The theoretical fits to the data in Fig. 1 are accurate for both the lower and higher temperatures, indicated by solid black and red curves respectively. However, there
are significant discrepancies at small strain rates for the intermediate temperatures $T = 700K$ and $840K$, shown by the dashed blue lines. (There is a similar discrepancy for $T = 800K$, not shown in the figure.) At $T = 840K$, for example, the theoretical stress starts to fall below the observed values when $q$ becomes less than about $10^{-4}$; and the system switches over to linear viscosity at smaller strain rates. The multi-species STZ theory proposed in [13, 14] predicts that the same thing will happen whenever $\rho$ is nonzero, but that the transition will occur at a smaller $q$ and, therefore, at a larger viscosity.

In Fig. 2, we show four examples of our theoretical $\chi$’s as functions of strain rate. All four converge to the same large-$q$ behavior shown in Eq. (4.1). The heavy black curve is the low-temperature function, $\chi = \hat{\chi}(q)$, valid for all cases in the range $100K < T < 600K$ where $\rho = 0$. It decreases logarithmically at small $q$ toward $\chi_0 = 0.1$. The two red curves, for $T = 940K$ and $1100K$, where $\rho$ is large, fall quickly to their corresponding values of $\chi = \theta = T/T_Z$ at small $q$. The blue dashed curve is for $T = 840K$, where $\rho \cong 0.1$. This curve falls substantially below the $\rho = 0$ curve before levelling off at $T/T_Z = 0.12$, again an indication that the small-$q$ behavior is incorrect. The theory predicts too small a viscosity in this case; therefore, it predicts too small a value of $\Gamma(s, \chi)$ relative to $\rho$ in Eq. (5.4), and thus predicts too fast an approach to the thermal limit. Note that the largest values of $\chi$ shown in this figure are still small enough that the STZ density $\sim \exp(-1/\chi)$ remains small.

In principle, an ideal glass transition occurs in an undriven, equilibrated, amorphous system at a Kauzmann temperature $T_0$, below which the viscosity is infinite and, above which, the yield stress vanishes. The GCE scaling analysis implies that something like this transition is occurring in this rapidly deforming system.

To begin to understand this situation, we show in Fig. 3 our “measured” values of the thermal noise strength $\rho$ for the eleven different temperatures included in the GCE data. This function behaves more or less as expected, falling from a value of the order of unity at large $T$ to become unmeasurably small below the glass transition. The most notable feature of this curve is the smooth transition to zero in the region $600K < T < 840K$. For these temperatures, we have optimized the fits to be accurate at the higher strain rates, as shown in Fig. 1 by the dashed curves for $T = 700K$ and $840K$. The single-species STZ theory should be valid at large $q$. Therefore, these measurements of $\rho$ must be taken seriously despite the fact that they appear to be inconsistent with VFT formulas for $\rho$ of the kind developed in [17, 18].

The glass transition emerges more clearly if, instead of plotting $\rho$ itself, we use the $\rho$-dependent formula for $\eta_N$ in Eq. (5.1) to plot the inverse Newtonian viscosity $\eta_N^{-1}$ as a function of temperature. This is the function that GCE suggest can be fit by

$$\eta_N^{-1}(T) = \begin{cases} \text{constant} \times (T - T_0)^\alpha & \text{for } T > T_0, \\ 0 & \text{otherwise}, \end{cases} \quad (5.1)$$

where $T_0 = 860K$ and $\alpha = 1.23$. Our result and the comparison with the GCE fit are shown in Fig. 4. Here, the fact that we are underestimating the viscosities for temperatures less than $T_0$ does not make much difference because $\eta_N^{-1}$ is already very small in that region.

According to the STZ theory, there is no yield stress in the limit of zero strain rate when $\rho$ is nonzero; the system must have a finite linear viscosity at small enough
The analysis presented here makes it appear that the STZ theory of amorphous plasticity is realistic up to strain rates of about $10^{-2}/\tau_0$, i.e. up to about one percent of the underlying molecular relaxation rate. Both the GCE simulations and our theoretical results exhibit an apparently simple scaling behavior across three decades of strain rates below that limit. Our STZ analysis implies, at least in a first approximation, that effective-temperature thermodynamics, rather than material-specific molecular interactions, control the system dynamics at high strain rates.

As discussed in Sec. I these STZ-based results are technically beyond the range of the mode-coupling theory \cite{Teitel85}, because they extend down to temperatures well below the glass transition where plastic flow is governed by other mechanisms.
FIG. 6: (Color online) Scaled inverse viscosity \( \eta^{-1} \), as a function of the scaled stress \( s_T \), computed from the STZ theory using all eleven GCE temperatures. The color scheme is analogous to that used in Fig. 1. The four solid red curves that collapse approximately to a single scaled inverse viscosity are for temperatures in the range 900 K - 1100 K. The four solid black curves that collapse approximately to a single scaled yield stress are for temperatures in the range 100 K - 600 K. The three dashed blue curves, which deviate from the scaling pattern, are for temperatures 700 K, 800 K and 840 K, reading from bottom to top.

by nonperturbative activation mechanisms. However, see \cite{6} for a counter argument. Similarly, we do not see how our results can be reproduced in the framework of soft glassy rheology (SGR) \cite{22,23}, because the STZ and SGR theories are so different kinematically, and because our present analysis is based so strongly on effective-temperature dynamics. Again, however, we point to \cite{22}, which describes progress in reformulating SGR in a thermodynamic framework analogous to that of \cite{15}. It will be interesting to see whether the results presented here do, in fact, distinguish the STZ theory from either of these two other competing points of view.

Our analysis in this paper is based on several, fundamental hypotheses that are potentially falsifiable, and which require further attention. First, in order to check the thermodynamic hypothesis, we need to test our theoretical estimate of the effective temperature \( \chi \) by measuring it directly. Computational measurements of \( \chi \) in \cite{22} served as the basis for the theory developed in \cite{21}, which led to the equations of motion for \( \chi \) presented here in Sec. \ref{theory}. Analogous measurements of an effective temperature for a thermalized hard-sphere system have been reported recently by Haxton \cite{24}, and an STZ analysis of Haxton’s results has revealed interesting insights \cite{27}.

So far, we have not had a similar consistency check for our analysis of the GCE data.

Second, our qualitative explanation of the discrepancies between the GCE data and single-species STZ theory is based on the multi-species reinterpretation of glassy viscosity proposed in \cite{14}. We need now to develop this reinterpretation into a quantitative theory, and test whether that theory corrects the discrepancies. This is our most unconventional effort. We have used large-strain-rate data, instead of the conventional small-strain-rate viscosity, to evaluate the thermal noise strength \( \rho(\theta) \). This procedure produces unexpectedly large values of \( \rho(\theta) \) at temperatures near \( T_0 \), as seen in Fig. 3. The argument in \cite{14} tells us that, because of the statistically inevitable presence of “slow” STZ’s, the viscosity increases more rapidly with decreasing temperature than would be predicted simply by the \( \theta \) dependence of \( \rho \). This argument, when applied to undriven systems near their glass temperatures, accounts for Stokes-Einstein violations and even for the stretched-exponential nature of various relaxation functions. The question is: Does it also account quantitatively for what look like anomalously large viscosities in the GCE data?

A third class of questions arises naturally within the context of the first two. Our analysis indicates that the model of a metallic glass used in the GCE simulations undergoes some kind of glass transition at \( T_0 \approx 860 K \). As \( T \) decreases below \( T_0 \), the configurational noise strength \( \rho \) decreases smoothly and becomes unmeasurably small below about 600 K. As this happens, the system changes from a viscous, glass-forming liquid to a solidlike glass with an apparently well defined yield stress. At nonzero, steady, shear rates, this transition becomes smoother; the liquidlike behavior persists down to arbitrarily low temperatures with continuous increase in the stress level.

Finally, an obvious question from a first-principles point of view is how the STZ transitions appear in the form of changes in the underlying atomic structure. Reference \cite{28}, a sequel to the GCE simulation \cite{2}, is a recent example of work along these lines. It is shown there that, during steady-state flow, the macroscopic Maxwell relaxation time \( \eta/G_\infty \) (where \( G_\infty \) is the instantaneous shear modulus) is essentially the same as the time required for a localized topological change to occur in which an atom gains and/or loses one of its nearest neighbors. We expect that further investigations of this kind will lead to a clearer microscopic picture of the STZ transition mechanism.

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