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Emergence of Long-Ranged Stress Correlations at the Liquid to Glass Transition

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A theory for the nonlocal shear stress correlations in supercooled liquids is derived from first principles. It captures the crossover from viscous to elastic dynamics at an idealized liquid to glass transition and explains the emergence of long-ranged stress correlations in glass, as expected from classical continuum elasticity. The long-ranged stress correlations can be traced to the coupling of shear stress to transverse momentum, which is ignored in the classic Maxwell model. To rescue this widely used model, we suggest a generalization in terms of a single relaxation time τ for the fast degrees of freedom only. This generalized Maxwell model implies a divergent correlation length $\xi \propto \tau$ as well as dynamic critical scaling and correctly accounts for the far-field stress correlations. It can be rephrased in terms of generalized hydrodynamic equations, which naturally couple stress and momentum and furthermore allow us to connect to fluidity and elastoplastic models.

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In 1867, Maxwell described the phenomenology of viscoelasticity in quiescent glass-forming liquids [1]. A viscoelastic liquid possesses a slow structural process characterized by the (final) relaxation time τ and behaves like a solid with (shear) elastic modulus G_∞ in rapid deformations. Only on long time scales does the liquid flow with a finite viscosity η, which follows as $\eta = G_\infty \tau$ according to Maxwell. The approach to vitrification is modeled by an increase in the relaxation time, and a glass state is probed when τ exceeds the observation time. This model for the macroscopic stress response close to equilibrium has been the basis for phenomenological extensions to flowing complex fluids [2], yet it was overlooked that the model errs in the solid state. Equilibrium correlations in fluids are short ranged, whereas elastic stress correlations in solids are long ranged according to classical continuum mechanics [3], and the emergence of shear rigidity in glass is still a topic of active research [4–9].

Local stresses in amorphous solids have been a topic of strong interest in recent years, focusing on the rheology of viscoplastic materials [10–14]. The yielding of soft glassy materials is well accounted for in phenomenological models, which combine elastic deformations at small stresses with plastic deformations at large stresses. Well inside the glassy phase, the latter have been identified with activation processes between metastable states [10–12], or with localized excitations, such as shear deformation zones [13] and quadrupolar energy fluctuations [14]. Their coupling via long-ranged elastic stress fields has been incorporated in elastoplastic models [15–18].

Here we focus on the liquid to glass transition. Following Maxwell in assuming that correlations at finite frequencies cross over smoothly at a glass transition, the emergence of elasticity thus requires the buildup of long-lived and long-ranged spatial correlations in supercooled states. We consider spatial correlations of the shear stress within the Zwanzig-Mori (ZM) formalism [19], and thereby generalize Maxwell's macroscopic description to finite wave vectors $\mathbf{q}$. We recover the far-field solutions of elasticity theory in isotropic solids [20] in the limit of large τ and identify their precursors in the fluid phase. The generalized Maxwell model contains a correlation length ξ diverging at the glass transition and possesses a nonanalytic limit for small $\mathbf{q}$ in glass.

We start from the conservation of total momentum in a fluid of N particles with mass m which introduces the stress tensor $\boldsymbol{\sigma}$ as the momentum current: $\partial_t m\mathbf{v}(\mathbf{q},t) = i\mathbf{q}\cdot\boldsymbol{\sigma}(\mathbf{q},t)$. The fluid velocity field is $\mathbf{v}(\mathbf{q},t) = (1/\sqrt{N})\sum_{i=1}^{N} e^{i\mathbf{q}\cdot\mathbf{r}_i(t)}\mathbf{v}_i(t)$ [21], where $\mathbf{r}_i(t)$ and $\mathbf{v}_i(t)$ denote the position and velocity of particle i. Using Newton's equation of motion, $m\dot{\mathbf{v}}_i(t) = \mathbf{F}_i(t)$, the stress tensor contains a kinetic term, which we will neglect, and a potential term, whose expression was determined by Irving and Kirkwood [22]. The potential term dominates in supercooled and glassy states [23] and its qualitative change at the glass transition is the topic we want to address. Fluctuations are decomposed into plane-wave contributions at $\mathbf{q}$ by Fourier transformation.

The crucial difference between a fluid and a solid concerns the response under volume-conserving shear deformations; a fluid flows with a viscosity while a solid deforms dominantly elastically. In both cases, the force transmitted by the stress through a planar element is coplanar to it, which is captured by an off-diagonal element of the stress tensor [3]. We chose $\sigma_{xy}(\mathbf{q})$, and consider its autocorrelation function

$$C_\sigma(\mathbf{q},t) = \frac{n}{k_B T}\langle\sigma_{xy}(\mathbf{q},t)^*\sigma_{xy}(\mathbf{q})\rangle, \tag{1}$$

where any other set of orthogonal directions other than the $\hat{\mathbf{x}}$ and $\hat{\mathbf{y}}$ directions would be equivalent; here n is the particle density and k_BT the thermal energy. Also, we will consider the limit of an incompressible and isothermal fluid in order to simplify the presentation, postponing the compressible case to a future submission [24]. The hydrodynamic conservation laws cause slow dynamics in the shear stress autocorrelation function $C_\sigma(\mathbf{q},t)$, which can be brought out by projections onto the conserved variables according to the ZM formalism [19]. A projection operator $\mathcal{P}$ captures the overlaps between general fluctuations and fluctuations of the (relevant) conserved quantities, which are the transversal momenta in the present (incompressible, isothermal) case; $\mathcal{P}=(m/k_BT)\mathbf{v}^\perp(\mathbf{q})\rangle\cdot\langle\mathbf{v}^\perp(\mathbf{q})^*$, with $\mathbf{v}^\perp(\mathbf{q})=\mathbf{q}\times[\mathbf{q}\times\mathbf{v}(\mathbf{q})]/q^2$. Nonlocal and non-Markovian effects follow from integrating out internal degrees of freedom and introduce memory kernels, which reduce to transport coefficients in the hydrodynamic limit of both small frequencies and small wave vectors [25]. Götze and Latz formulated a general ZM decomposition which is aimed at the limit of generalized hydrodynamics (GH) in viscoelastic fluids and retains the possibility of a large structural relaxation time τ [26]. Applied to correlations of the shear stress, we find for the Laplace transform of the stress autocorrelation

$$C_\sigma^{\mathrm{GH}}(\mathbf{q},s)=G_0^\perp(s)-\left((q_x^2+q_y^2)-4\frac{q_x^2q_y^2}{q^2}\right)\frac{[G_0^\perp(s)]^2}{nk_BT}K_q^\perp(s). \tag{2}$$

[We have used the convention $C(s)=\int_0^\infty dte^{-st}C(t)$ and only kept the leading order in q; for details of the derivation, see the Supplemental Material [27].] The GH representation of Eq. (2) shows explicitly the decomposition of stress relaxation into a contribution of the hydrodynamic modes and the fast dynamics in the space complementary to the hydrodynamic modes. The latter are captured by the time- or frequency-dependent memory kernel $G_0^\perp(s)$, whereas the former enter Eq. (2) via the transverse current correlation function $\langle\mathbf{v}^\perp(\mathbf{q},t)^*\mathbf{v}^\perp(\mathbf{q})\rangle=(\mathbf{1}-\mathbf{q}\mathbf{q}/q^2)\mathrm{K}_q^\perp(t)$. In the GH-limit, $K_q^\perp(s)$ is conveniently represented [21],

$$K_q^\perp(s)=\frac{k_BT/m}{s+\frac{q^2}{mn}G_0^\perp(s)}, \tag{3}$$

in terms of the same memory kernel $G_0^\perp(s)$. The kernel is thus identified as generalized shear viscosity, called shear modulus in rheometry [2], which is the autocorrelation of the fluctuating transverse force with ZM-reduced dynamics at $q=0$ [19,21].

Equation (2) is the central result of our Letter and will be discussed in the following for supercooled liquids, glasses, and the transition in between. It is valid for arbitrary frequencies and contains microscopic motion on short time scales. A related function, the transverse force autocorrelation with real dynamics, which does not exhibit elastic correlations [28], can be recovered from $C_\sigma(\mathbf{q},t)$ for specific wave vector directions (viz., $\mathbf{q}=q\hat{\mathbf{x}}$ and $\mathbf{q}=q\hat{\mathbf{y}}$). The crucial differences between fluids and glasses are observed for small frequencies or long time scales. We hence focus on stress correlations on hydrodynamic scales. This is also the regime where the ZM formalism develops its full power because the relaxation of selected variables can be well separated from microscopic scales. We already used this in Eq. (3), where the hydrodynamic pole in the (conserved) momentum fluctuations was identified, which captures shear diffusion in the limit of small frequencies and wave vectors [19,21].

For the global shear stress in fluid states, the difference between ZM-projected and full dynamics vanishes as expected [19],

$$C_\sigma(\mathbf{q}=\mathbf{0},t)=G_0^\perp(t),\qquad\text{(fluid)}. \tag{4}$$

The global memory kernel reduces to the shear viscosity for vanishing frequency, so Eq. (4) is equivalent to the Green-Kubo relation for the viscosity [21],

$$\begin{aligned}\eta&=G_0^\perp(s=0)\\&=\frac{n}{k_BT}\int_0^\infty dt\langle\sigma_{xy}(\mathbf{q}=\mathbf{0},t)\sigma_{xy}(\mathbf{q}=\mathbf{0})\rangle.\end{aligned} \tag{5}$$

The hydrodynamics of fluids is recovered by replacing the memory kernels by wave vector–and frequency-independent transport coefficients. Here this means $G_0^\perp(s\to 0)\to\eta$, and gives for the shear stress correlation function in a liquid

$$C_\sigma^{\mathrm{fluid}}(\mathbf{q},s)=\eta+\left(\frac{q_x^2q_y^2}{q^2}-\frac{q_x^2+q_y^2}{4}\right)\frac{4\eta^2}{nms+\eta q^2}. \tag{6}$$

Clearly, the hydrodynamic velocity correlator introduces anisotropic terms into the stress correlator. In the corresponding time-dependent correlation, $C_\sigma^{\mathrm{fluid}}(\mathbf{q},t)$, these are in fact dominant for finite times [see Fig. 1 (left, lower panel)].

Maxwell‘s model of viscoelasticity introduces a single relaxation time τ for the stress, so that its correlation is given by $C_\sigma^{\mathrm{Max}}(\mathbf{q},s)=G_\infty\tau/(1+s\tau)$. (Maxwell ignored the wave vector dependence.) Solidlike behavior emerges, when the deformations are rapid relative to τ and elastic correlations dominate at low frequencies. This implies $\lim_{s\to 0}\lim_{\tau\to\infty}sC_\sigma^{\mathrm{Max}}(\mathbf{q},s)=G_\infty$ and a persistent contribution of the time-dependent function: $\lim_{t\to\infty}\lim_{\tau\to\infty}C_\sigma^{\mathrm{Max}}(\mathbf{q},t)=G_\infty$. For the generalized hydrodynamics, entailed in the full Eq. (2), we still expect a divergence of the relaxation time τ; however, the small q

dependence is completely different than envisioned by Maxwell due to the coupling to correlations of transverse momentum which are long ranged in an elastic solid. To elucidate this we follow the suggestion by the microscopic mode-coupling theory that an idealized glass state is obtained when the relaxation time τ is infinite and correlation functions do not decay to zero [29]. This should hold for collective density fluctuations and the fluctuating forces entering the memory kernel $G_0^\perp(t)$ [30], implying a time-persistent contribution to the shear stress correlation,

$$C_\sigma(\mathbf{q},t)\to C_\sigma^\infty(\mathbf{q}),\quad \text{for } t\to\infty \text{ in glass.} \tag{7a}$$

In an incompressible glass, its small wave vector limit is solely determined by $G_{0,\infty}^\perp = \lim_{t\to\infty} G_0^\perp(t)$ and follows from Eq. (2),

$$C_\sigma^\infty(\mathbf{q}) = \left(4\frac{q_x^2 q_y^2}{q^4}+\frac{q_z^2}{q^2}\right)G_{0,\infty}^\perp + \mathcal{O}(q^2). \tag{7b}$$

The system is characterized by a finite resistivity to shear deformations and Maxwell's shear elastic constant can be identified with $G_\infty = G_{0,\infty}^\perp$. In contrast to Maxwell's macroscopic description of a glass, however, generalized hydrodynamics predicts that the limit of $q\to 0$ is nonanalytic and depends on the direction $\mathbf{q}$ is taken to zero. Such nonanalytic behavior points to the existence of a length scale that diverges in the supercooled regime when approaching the glass transition. It will be discussed below. Eshelby's result for the response of an elastic (isotropic) medium to a point force can be recognized in Eq. (7b) [15,20]. It predicts the existence of long-ranged stress correlations,

$$C_\sigma^\infty(\mathbf{r})\to\frac{3}{4\pi}\frac{G_{0,\infty}^\perp}{r^3}\frac{10x^2y^2-r^2(x^2+y^2)}{r^4},\quad \text{for } r\to\infty. \tag{7c}$$

In d dimensions, the elastic correlations decay like r^{-d}, as seen recently in flowing dense quasi-two-dimensional emulsions [31].

How can we generalize Maxwell's theory to account for the frequency- and wave vector–dependence of stress correlations at the glass transition and in the elastic solid? Generalized hydrodynamics, as entailed in Eq. (2), suggests to approximate the memory kernel instead of the correlations of the stress itself. Thus we model the memory kernel by a single relaxation time τ

$$G_0^\perp(s)\approx G^{\mathrm{gM}}(s)=\frac{G_\infty \tau}{1+s\tau} \tag{8a}$$

in the spirit of Maxwell but correctly accounting for the coupling of the stress to conserved momentum fluctuations. Substitution into Eq. (2) then captures the far-field shear stress correlation function in the incompressible limit, to be denoted $C_\sigma^{\mathrm{gM}}(\mathbf{q},s)$. The generalized Maxwell model has a rather rich content. $C_\sigma^{\mathrm{gM}}(\mathbf{q},s)$ contains an isotropic term and an anisotropic term whose angular dependence $4\hat{q}_x^2\hat{q}_y^2+\hat{q}_z^2$ agrees with the one of the elastic Green's function in Eq. (7b). The distance to the idealized glass transition is controlled by the divergence of τ, which we do not specify explicitly. Introducing a characteristic length scale $\xi^2 = G_\infty\tau^2/mn = v_T^2\tau^2$ (with v_T the transversal sound velocity of glass [29,32]), we observe that the time- and wave number–dependent stress correlation obeys scaling

$$C_\sigma^{\mathrm{gM}}(\mathbf{q},t) = G_\infty \mathcal{F}(t/\tau,\mathbf{q}\xi); \tag{8b}$$

i.e., all dependence on the distance to the critical point is absorbed in the time scale τ and the length scale ξ. The correlation length $\xi\propto\tau$ determines the spatial extent of solidlike regions within the viscoelastic fluid. It diverges strongly as the glass transition is approached, signaling the appearance of long-ranged stress correlations in the glassy state, and may be related to the length scale seen in four-point density correlations in simulations [33,34]. The hydrodynamic excitations are determined by the poles of $C_\sigma^{\mathrm{gM}}(\mathbf{q},s)$ in the complex s plane: $s\tau(1+s\tau)+q^2\xi^2=0$. The hydrodynamics of the fluid is recovered in the limit

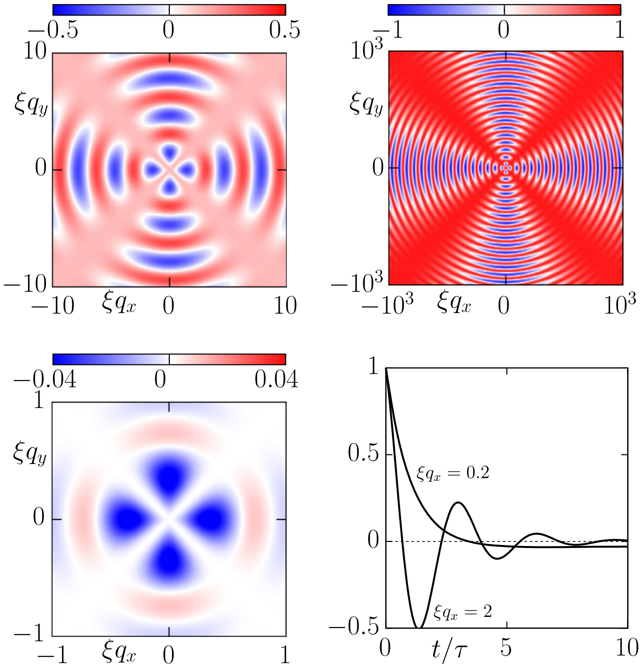

FIG. 1. Scaling function $C_\sigma^{\mathrm{gM}}(\mathbf{q},t)/G_\infty = \mathcal{F}(t/\tau,\mathbf{q}\xi)$ of the generalized Maxwell model in the q_x and q_y plane for three rescaled times t/τ. Top left panel is in the viscoelastic regime for $t/\tau = 2$, top right panel is in the glassy regime for $t/\tau = 10^{-1}$, and bottom left panel is in the fluid regime for $t/\tau = 10^1$; $v_T = 1$ is chosen. Bottom right panel: Time dependence of $\mathcal{F}(t/\tau,\mathbf{q}\xi)$ along the axis $q_y = 0$ for two ξq_x as labeled.

$s\tau \ll 1$, implying $s\tau = -q^2\xi^2$. Glassy hydrodynamics is recovered in the opposite limit $s\tau \gg 1$, implying $s\tau = \pm iq\xi$. The critical dynamics is contained in the roots of the dispersion relation for $q\xi \gg 1$. In this limit, one finds again $s\tau = \pm iq\xi$, i.e., small-wavelength, high-frequency sound in the fluid phase [29,32].

Figure 1 summarizes the different behaviors, which could be tested in scattering from photoelastic materials [35]. For short rescaled times, the elastic limit Eq. (7b) dominates. [Note that the rapid oscillations along the axes would be damped if an instantaneous dissipation rate was added in Eq. (8a), which is an easy way to capture sound damping [24].] For long times, the decay of shear oscillations is evident, but the stress correlations remain anisotropic. For intermediate times, $t/\tau \approx 1$, the complete anisotropic and oscillatory pattern of the scaling function $\mathcal{F}(t/\tau, \mathbf{q}\xi)$ reveals itself. As function of time, $\mathcal{F}(t/\tau, \mathbf{q}\xi)$ is nonmonotonic along the axes (see Fig. 1) and an exponential along the diagonals.

The stress correlation tensor can be measured in a linear response experiment by applying a weak shear velocity gradient $\partial_y \mathrm{v}_x^{\mathrm{ext}}$ to the liquid [19]. The inhomogeneous flow gives rise to a shear stress, which is given in linear response by

$$\langle \sigma_{xy}(\mathbf{r},t)\rangle^{\mathrm{lr}} = 2\int_{-\infty}^{t} dt' \int d^d r' C_\sigma(\mathbf{r}-\mathbf{r}', t-t')\bar{\kappa}_{xy}^{\mathrm{ext}}(\mathbf{r}', t'). \tag{9}$$

Because of the symmetry of the Irving-Kirkwood stress tensor, only the symmetric component of the external velocity gradient enters, with $\bar{\kappa}_{\alpha\beta}(\mathbf{r},t) = \frac{1}{2}[\partial_\alpha \mathrm{v}_\beta(\mathbf{r},t) + \partial_\beta \mathrm{v}_\alpha(\mathbf{r},t)]$. In the fluid phase, a uniform stationary shear flow gives rise to uniform stationary stress [21]: $\sigma_{xy} = 2\eta\bar{\kappa}_{xy}^{\mathrm{ext}}$. This global constitutive equation is generalized by Eq. (2) to finite wave vectors and frequencies, $\sigma_{xy}(\mathbf{q},\omega) = 2C_\sigma(\mathbf{q}, s=-i\omega)\bar{\kappa}_{xy}^{\mathrm{ext}}(\mathbf{q},\omega)$. Here, a periodic external flow rate with frequency ω was assumed, which leads to a periodic stress with the same frequency.

The linear response relation allows for an intuitive interpretation of generalized hydrodynamics. Momentum conservation is expressed as usual in terms of the force balance with the local stress and pressure p,

$$mn\partial_t \mathbf{v}(\mathbf{r},t) = \nabla \cdot [\boldsymbol{\sigma}^{\mathrm{gM}}(\mathbf{r},t) - p(\mathbf{r},t)\mathbf{1}]. \tag{10a}$$

We consider the incompressible limit (viz., $\nabla \cdot \mathbf{v} = 0$), so that the velocity is purely transverse $\mathbf{v}^\perp$ and does not couple to the pressure. The constitutive equation for $\boldsymbol{\sigma}^{\mathrm{gM}}(\mathbf{r},t)$,

$$\left(\frac{1}{\tau} + \partial_t\right)\boldsymbol{\sigma}^{\mathrm{gM}}(\mathbf{r},t) = 2G_\infty[\bar{\boldsymbol{\kappa}}^{\mathrm{ext}}(\mathbf{r},t) + \bar{\boldsymbol{\kappa}}(\mathbf{r},t)], \tag{10b}$$

is built on Maxwell's insight on glassy relaxation but includes the full velocity gradient, which is the sum of the externally applied one and the internal flow as computed from Eq. (10a), resulting in a linear but spatially nonlocal differential equation. Fourier transformation of Eqs. (10a) and (10b) reproduces the linear response relation with $C_\sigma^{\mathrm{gM}}(\mathbf{q}, s)$ as given by Eq. (2) with the Maxwell approximation for $G_0^\perp(s)$.

While the classic Maxwell model only has the external flow gradient on the right-hand side of Eq. (10b), the generalized model also includes the internal $\bar{\boldsymbol{\kappa}}$ arising from the nonlocal velocity field, which necessarily is induced by the imposed flow. Only for finite frequencies and relaxation times τ does the transverse momentum diffusion allow for an (anisotropic) gradient expansion leading to Eq. (6). In solid states where $1/\tau = 0$, however, the gradient expansion breaks down and the nonlocal strain field induces the nonanalytic small-$\mathbf{q}$ expansion in Eq. (7), which signals long-ranged elastic stress fields that are at the heart of the elastoplastic models [15–17]. The strain field considered in these models follows here as the time integral of the (symmetrized) velocity gradient tensor [36]: $\varepsilon(\mathbf{r},t) = \int_{t_0}^{t_0+t} dt' \bar{\boldsymbol{\kappa}}(\mathbf{r},t')$.

In summary, within the Zwanzig-Mori approach we have obtained the nonlocal correlations of the shear stress in the long-wavelength limit. This result holds generally in viscoelastic liquids including, e.g., polymeric systems, where the q dependence of the transverse force correlation function has been studied [37,38]. We specifically addressed glass-forming melts. We have shown that the simplest generalization of Maxwell's model including spatial variations of the stress recovers the long-ranged elastic fields expected in solids. The shear-stress memory kernel plays the role attributed by Maxwell to the global shear stress. The generalization implies the rapid growth of a correlation length ξ, which opens the window in wave vector space for the nonanalytic small-$\mathbf{q}$ behavior of the shear stress autocorrelator expected in solids. The far-field decay of the frozen-in stress fluctuations $C_\sigma^\infty(\mathbf{r}) \propto r^{-d}$ agrees with the one deduced from Goldstone modes in solids with quenched disorder in $d = 2$ and $d = 3$ [39]. Our approach to neglect the wave vector dependence of the generalized viscosity kernel $G_q^\perp(t)$ is at odds with some simulation results [40–42] which appear to find a strong wave vector dependence of the viscosity when supercooling. (Whether this can be related to the nonanalytic $\mathbf{q}$-dependent stress correlations we find should be clarified in future.) Yet, our approach may be useful for nonlocal rheological models, where *ad hoc* transport equations are formulated including diffusive terms [43] or considering the inverse Maxwell relaxation time as an independent state variable [44–46]. The generalized Maxwell model Eq. (10) implies that nonlocality of the stress relaxation is generated by the internal velocity field accompanying the externally imposed flow, which constrains the building of nonlocal rheological models.

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