THE JOURNAL OF CHEMICAL PHYSICS **149**, 084502 (2018)

Stress auto-correlation tensor in glass-forming isothermal fluids: From viscous to elastic response

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(Received 14 June 2018; accepted 8 August 2018; published online 27 August 2018)

We develop a generalized hydrodynamic theory, which can account for the build-up of long-ranged and long-lived shear stress correlations in supercooled liquids as the glass transition is approached. Our theory is based on the decomposition of tensorial stress relaxation into fast microscopic processes and slow dynamics due to conservation laws. In the fluid, anisotropic shear stress correlations arise from the tensorial nature of stress. By approximating the fast microscopic processes by a single relaxation time in the spirit of Maxwell, we find viscoelastic precursors of the Eshelby-type correlations familiar in an elastic medium. The spatial extent of shear stress fluctuations is characterized by a correlation length ξ which grows like the viscosity η or time scale $\tau \sim \eta$, whose divergence signals the glass transition. In the solid, the correlation length is infinite and stress correlations decay algebraically as r^{-d} in d dimensions. *Published by AIP Publishing.* https://doi.org/10.1063/1.5044662

I. INTRODUCTION

Correlation functions provide important information on the structural and transport properties of simple and complex fluids.[1,2] At long wavelengths, the continuum mechanics theory of fluctuating hydrodynamics rationalizes slow wave-like or diffusive processes in the correlation functions of density, momentum, and energy fluctuations.[3] These are the fields of conserved variables and considerations of, e.g., the entropy production rate provide the (Newtonian, Markovian) constitutive equations required to close the set of equations and to explain the hydrodynamic modes.[4] For example, transverse momentum relaxes diffusively with the ratio of shear viscosity η to mass density determining the diffusion coefficient. The viscosity enters from the constitutive equation valid for Newtonian fluids that transverse stresses are proportional to the shear element of the velocity gradient tensor, $\sigma_{xy} = \eta \, \partial_y v_x$.[5] Microscopic theories can build on the projection operator formalism developed by Zwanzig and Mori[6,7] to derive the Markovian equations of fluctuating hydrodynamics by coarse-graining.[1,2,8] This provides exact microscopic expressions for the determination of the correlation functions also beyond the hydrodynamic regime.

While the correlation functions of the conserved fields in liquids have amply been studied experimentally, numerically, and theoretically, far less is known about correlation functions of non-conserved fields such as the stress tensor. On the experimental side, first measurements of the stress redistribution in a flowing emulsion have been performed[9] and techniques are being developed in order to measure stress correlations in quiescent complex fluids.[10] On the theoretical side, the Green-Kubo relation states that the shear viscosity can be obtained by integrating the auto-correlation function of the total shear stress over all times from zero to infinity; the total shear stress itself is given by the integral over the whole volume of the system.[1] By exploiting the law of transverse momentum conservation, Evans[11] could connect specific elements of the stress tensor correlations to the well-studied momentum correlations. This relation has been used in molecular dynamics simulations.[12] To the best of our knowledge, however, the correlations of the local stress have not been analyzed within the formalism of (generalized) hydrodynamics.

In this contribution, we provide the complete tensorial analysis of long-wavelength stress correlations in glass-forming liquids within the Zwanzig-Mori projection operator formalism. As sole approximation, we neglect energy fluctuations which corresponds to assuming rapid heat transport. The analysis extends a short exposition of transverse correlations in Ref. 13 and addresses the emergence of rigidity at the glass transition.[14–18] Moreover, we discuss nonlocal correlations in the far-field and generalize Maxwell's widely used model of viscoelasticity.[19] Equilibrium correlations in fluids are short-ranged, while elastic stresses are correlated over large distances in solids.[5] We unify their description across the glass transition following Maxwell in assuming that correlations at finite frequencies and wavevectors crossover smoothly.

The paper is organized as follows. In Sec. II, the relevant dynamical variables in liquids are introduced and the conservation laws are summarized. In Sec. III, the auto-correlation tensor of stress fluctuations is defined and its symmetry properties and slow dynamics owing to conservation laws are analyzed. The Zwanzig-Mori projection operator formalism is applied, and results valid in arbitrary isotropic and homogeneous viscoelastic fluids at fixed temperature are obtained. Section IV focuses on the nonlocal and retarded auto-correlation function of shear stresses because these show the most interesting phenomena when approaching a glass transition. We include spatial resolution into Maxwell's picture of viscoelasticity and discuss the shear stress evolution at large distances and long times. The discussion draws on the picture of an idealized glass transition developed by mode coupling theory (MCT).[8] Section V A provides (for completeness) the formal argument from linear response theory that the considered stress auto-correlation tensor can be measured in rheological experiments for small deformation rates. This leads to a formulation of the generalized Maxwell model, developed in Sec. V B, in terms of hydrodynamic equations, familiar from rheological modeling. We summarize our results and give a perspective to future developments of our approach in Sec. VI. Technical details are delegated to Appendixes A–C.

II. DYNAMIC VARIABLES AND CONSERVATION LAWS

The stress tensor $\boldsymbol{\sigma}$ arises in the law of momentum conservation,[1]

$$\partial_t \mathbf{v}(\mathbf{q},t) = i\mathcal{L}\mathbf{v}(\mathbf{q},t) = i\mathbf{q}\cdot\boldsymbol{\sigma}(\mathbf{q},t)/m, \tag{1}$$

where the velocity field of N point-particles in a volume V is

$$\mathbf{v}(\mathbf{q},t) = \frac{1}{\sqrt{N}}\sum_{i=1}^{N} e^{i\mathbf{q}\cdot\mathbf{r}_i(t)}\,\dot{\mathbf{r}}_i(t). \tag{2}$$

Note that the thermodynamic limit will be taken at the end. Here, $\mathbf{r}_i(t)$ is the position of particle i depending on time according to classical dynamics and $\dot{\mathbf{r}}_i(t)$ denotes its time derivative, viz., velocity. Because all particles have mass m, particle momentum and velocity will be treated as equivalent. In Eq. (1), the time-derivative is written with the Liouville operator, $\mathcal{L} = -i\partial_t$, which is Hermitian.[20] In Eq. (2), the fluctuations are given in Fourier space depending on the wavevector $\mathbf{q}$ in order to exploit translational invariance later on.

Newton's equation $i\mathcal{L}\mathbf{v}_i(t) = \mathbf{F}_i(t)/m$ is assumed to hold, where $\mathbf{F}_i$ is the force acting on particle i. Using this in Eq. (1), the kinetic and potential contributions to the divergence of the stress tensor follow

$$\begin{aligned} i\mathbf{q}\cdot\boldsymbol{\sigma}(\mathbf{q}) &= \frac{1}{\sqrt{N}}\sum_{i=1}^{N}\left[im\mathbf{q}\cdot\dot{\mathbf{r}}_i\dot{\mathbf{r}}_i + \mathbf{F}_i\right]e^{i\mathbf{q}\cdot\mathbf{r}_i} \\ &= \text{kinetic term} + \mathbf{F}(\mathbf{q}). \end{aligned} \tag{3}$$

The last equality defines a force density, that we will later use to interpret the linear combination of stress tensor elements, which determines its divergence. We will neglect the kinetic term and focus on the potential part because it dominates in supercooled and glassy states.[21] Its qualitative change at the glass transition is the topic we want to address, but see footnotes for the changes required to correctly include the kinetic contributions.

The potential part of the divergence of the stress tensor can be obtained from the microscopic Irving-Kirkwood expression that takes the following form when simplifying to pair interactions $U(r)$ (with $\mathbf{r}_{jk} = \mathbf{r}_j - \mathbf{r}_k$):[1,22]

$$\boldsymbol{\sigma}(\mathbf{q}) = \frac{1}{\sqrt{N}}\sum_{\langle j,k\rangle}\frac{\mathbf{r}_{jk}\mathbf{r}_{jk}}{r_{jk}}U'(r_{jk})\frac{1-e^{-i\mathbf{q}\cdot\mathbf{r}_{jk}}}{i\mathbf{q}\cdot\mathbf{r}_{jk}}\,e^{i\mathbf{q}\cdot\mathbf{r}_j}, \tag{4}$$

where the sum counts all different pairs j, k once.[23] The stress in Eq. (4) is symmetric $\sigma_{\alpha\beta} = \sigma_{\beta\alpha}$, where (here and throughout the text) Greek indices denote spatial directions. The average of the stress tensor gives the negative pressure p[24] in equilibrium $\sqrt{N}\langle\boldsymbol{\sigma}(\mathbf{q})\rangle/V = -p\,\mathbf{1}\,\delta_{\mathbf{q},\mathbf{0}}$, where equilibrium averaging is done with the canonical Gibbs-Boltzmann distribution. In the following, only the fluctuations $\delta\boldsymbol{\sigma}(\mathbf{q}) = \boldsymbol{\sigma}(\mathbf{q}) - \langle\boldsymbol{\sigma}(\mathbf{q})\rangle$ will be of interest.

In real space, the stress tensor reduces to the form

$$\boldsymbol{\sigma}(\mathbf{r}) = \frac{1}{\sqrt{N}}\sum_{\langle j,k\rangle}\frac{\mathbf{r}_{jk}\mathbf{r}_{jk}}{r_{jk}}U'(r_{jk})\int_0^1 ds\,\delta(\mathbf{r}-\mathbf{r}_k - s\mathbf{r}_{jk}). \tag{5}$$

For hard spheres, the replacement $\frac{\mathbf{r}_{ij}}{r_{ij}}U'(r_{jk}) \to -m\Delta\mathbf{v}_{ij}\delta(t-t_{ij})$ would allow the calculation of the force between particles i and j from the momentum transfer $m\Delta\mathbf{v}_{ij}$ in a collision at time t_{ij}.[25,26]

In order to study the dynamics of fluctuations in liquids, it is necessary to account for conservation laws because these imply slow relaxation processes at long wavelength. The Fourier-modes of the relevant conserved variables are the density

$$\varrho(\mathbf{q},t) = \frac{1}{\sqrt{N}}\sum_{i=1}^{N} e^{i\mathbf{q}\cdot\mathbf{r}_i(t)}, \tag{6}$$

which obeys the conservation law

$$\partial_t\,\varrho(\mathbf{q},t) = i\mathcal{L}\,\varrho(\mathbf{q},t) = i\mathbf{q}\cdot\mathbf{v}(\mathbf{q},t), \tag{7}$$

and the momentum $m\mathbf{v}(\mathbf{q},t)$ from Eq. (2). Energy conservation will be neglected for simplicity, assuming that heat transport is fast on the slow time scales of interest. The equilibrium magnitude of density fluctuations $\delta\varrho(\mathbf{q}) = \varrho(\mathbf{q}) - \langle\varrho(\mathbf{q})\rangle$ around the homogeneous average density n (viz., $\sqrt{N}\langle\varrho(\mathbf{q})\rangle/V = n\delta_{\mathbf{q},\mathbf{0}}$) is measured by the static structure factor $S_q = \langle|\delta\varrho(\mathbf{q})|^2\rangle$, while velocity fluctuations obey $\langle\mathbf{v}^*(\mathbf{q})\ \mathbf{v}(\mathbf{q})\rangle = \frac{k_BT}{m}\mathbf{1}$, because of the Maxwellian velocity distribution in equilibrium; here k_BT is the thermal energy, * denotes complex conjugation, and $\langle\mathbf{v}(\mathbf{q},t)\rangle = 0$ holds in quiescent equilibrium.

III. STRESS AUTO-CORRELATION TENSOR

The dynamics in liquids is traditionally described using correlation functions.[1,2] A correlation function of two variables A and B with fluctuations like $\delta A = A - \langle A\rangle$ takes the form $\langle\delta A(t)^*\delta B(0)\rangle = \langle\delta A(0)^*\mathcal{U}(-t)\delta B(0)\rangle$, with the unitary time evolution denoted by $\mathcal{U}(t) = e^{i\mathcal{L}t}$. It satisfies $\partial_t\mathcal{U}(t) = i\mathcal{L}\mathcal{U}(t)$ and the time-dependence of a general variable $A(t)$ can be rewritten as $A(t) = \mathcal{U}(t)A(0)$.[20] Time-translational invariance of the system is assumed throughout, enabling us to choose $\delta B(t')$ at $t' = 0$ in the above definition.

The central object of our study is the correlation tensor $\mathbf{C}^{(\sigma)}$ of the local potential stresses. It shall be studied in wavevector space exploiting homogeneity. Using the stress-fluctuations from Eq. (4), it is defined as

$$\mathrm{C}^{(\sigma)}_{\alpha\beta\gamma\delta}(\mathbf{q},t) = \frac{n}{k_BT}\,\langle\delta\sigma^*_{\alpha\beta}(\mathbf{q})\,e^{-i\mathcal{L}t}\,\delta\sigma_{\gamma\delta}(\mathbf{q})\rangle. \tag{8}$$

A number of symmetries under exchanges of indices hold for $C^{(\sigma)}_{\alpha\beta\gamma\delta}$,

$$C^{(\sigma)}_{\alpha\beta\gamma\delta}(\mathbf{q},t) = C^{(\sigma)}_{\beta\alpha\gamma\delta}(\mathbf{q},t) = C^{(\sigma)}_{\gamma\delta\alpha\beta}(\mathbf{q},-t)^* = C^{(\sigma)}_{\gamma\delta\alpha\beta}(-\mathbf{q},t). \tag{9}$$

These relations hold because the stress tensor is real and symmetric [see Eq. (5)], time-reversal symmetry is valid, and the Liouville operator is hermitian. The correlation tensor at $\mathbf{q} = 0$ has been well studied in liquids[1,2] including in supercooled states,[8] but its behavior at finite $\mathbf{q}$ has only been discussed for specific directions of the wavevector.[11]

A. Consequences of isotropy

In an isotropic and achiral system, a fourth order autocorrelation tensor is required to satisfy the following behavior under rotation or inversion of the wavevector $\mathbf{q}' = \mathbf{R}\cdot\mathbf{q}$ with a general orthogonal matrix $\mathbf{R}$ (obeying $\mathbf{R}^T\cdot\mathbf{R} = \mathbf{1}$),

$$\mathbf{C}^{(\sigma)}(\mathbf{q},t) = \mathbf{R}^T\mathbf{R}^T : \mathbf{C}^{(\sigma)}(\mathbf{R}\cdot\mathbf{q},t) : \mathbf{R}\mathbf{R}. \tag{10}$$

By inspection, or by generalizing the proof in Ref. 8 to tensors,[27] the general solution for the stress correlation tensor $\mathbf{C}^{(\sigma)}$ compatible with its index-symmetries in Eq. (9) follows:

$$\begin{aligned} C^{(\sigma)}_{\alpha\beta\gamma\delta}(\mathbf{q},t) = {}& C^{(1)}(q,t)\,\delta_{\alpha\beta}\delta_{\gamma\delta} + C^{(2)}(q,t)\,(\delta_{\alpha\gamma}\delta_{\beta\delta}+\delta_{\alpha\delta}\delta_{\beta\gamma}) \\ &+ C^{(3)}(q,t)\,(\delta_{\alpha\beta}q_\gamma q_\delta + q_\alpha q_\beta\delta_{\gamma\delta}) + C^{(4)}(q,t) \\ &\times(\delta_{\alpha\gamma}q_\beta q_\delta + \delta_{\alpha\delta}q_\beta q_\gamma + \delta_{\beta\gamma}q_\alpha q_\delta + \delta_{\beta\delta}q_\alpha q_\gamma) \\ &+ C^{(5)}(q,t)\,(q_\alpha q_\beta q_\gamma q_\delta). \end{aligned} \tag{11}$$

Quite conveniently, isotropy dictates that the five functions $C^{(i)}(q, t)$ only depend on the modulus $q = |\mathbf{q}|$. The $C^{(1)}(q, t)$ and $C^{(2)}(q, t)$ functions are the generalizations of the Lame coefficients λ and μ, respectively. The $C^{(3)}(q, t)$ and $C^{(4)}(q, t)$ functions are their leading anisotropic second order gradients, while $C^{(5)}(q, t)$ is the next-order anisotropic contribution. See Ref. 28 for their connection to spherical tensors and a more general decomposition including the case where $A_{\alpha\beta\gamma\delta} \neq A_{\gamma\delta\alpha\beta}$.

B. Consequences of conservation laws

The hydrodynamic conservation laws cause slow dynamics in $\mathbf{C}^{(\sigma)}$ that can be brought out by projections onto the conserved variables according to the Zwanzig-Mori formalism.[3] A projection operator $\mathcal{P}$ captures the overlaps between general fluctuations and fluctuations of the conserved quantities, which are density and momentum in the present case

$$\mathcal{P} = \frac{1}{S_q}\delta\varrho(\mathbf{q})\rangle\langle\delta\varrho(\mathbf{q})^* + \frac{m}{k_BT}\mathbf{v}(\mathbf{q})\rangle\cdot\langle\mathbf{v}(\mathbf{q})^*. \tag{12}$$

The complementary projector $\mathcal{Q} = 1 - \mathcal{P}$ decouples the fluctuations from the conserved hydrodynamic ones and obeys

$$\mathcal{P}\,\mathcal{Q} = 0 \quad \text{and} \quad \mathcal{Q}\mathcal{L}\delta\varrho(\mathbf{q}) = 0. \tag{13}$$

With these projectors, the general decomposition can conveniently be done in Laplace space, where the resolvent captures the (Laplace-) frequency dependence: $R(s) = \int_0^\infty dt\; e^{-st}e^{-i\mathcal{L}t} = (s+i\mathcal{L})^{-1}$. The general decomposition established by Götze and Latz,[29] which is summarized in Appendix A in order for the presentation to be self-contained, gives

$$C^{(\sigma)}_{\alpha\beta\gamma\delta}(\mathbf{q},s) = \frac{n}{k_BT}\Big\{\langle\delta\sigma^*_{\alpha\beta}(\mathbf{q})R'(s)\delta\sigma_{\gamma\delta}(\mathbf{q})\rangle + \langle\delta\sigma^*_{\alpha\beta}(\mathbf{q})\big[(1-iR'(s)\mathcal{L})\mathcal{P}R(s)\mathcal{P}(1-i\mathcal{L}R'(s))\big]\delta\sigma_{\gamma\delta}(\mathbf{q})\rangle\Big\}. \tag{14}$$

The reduced resolvent $R'(s) = \mathcal{Q}(s+i\mathcal{Q}\mathcal{L}\mathcal{Q})^{-1}\mathcal{Q}$ describes the time-dependence devoid of the slow conserved dynamics.[3,29] It appears at a number of places in Eq. (14), most prominently in the projected stress-memory kernel, viz., the first term on the right-hand side of Eq. (14),

$$M_{\alpha\beta\gamma\delta}(\mathbf{q},t) = \frac{n}{k_BT}\,\langle\delta\sigma^*_{\alpha\beta}(\mathbf{q})\mathcal{Q}\,e^{-i\mathcal{Q}\mathcal{L}\mathcal{Q}t}\,\mathcal{Q}\,\delta\sigma_{\gamma\delta}(\mathbf{q})\rangle. \tag{15}$$

This fourth order tensor obeys the same index exchange-relations (9), behavior under rotation (10), and decomposition into anisotropic terms (11) as the full stress correlation tensor because the projections in the reduced resolvent do not affect the relevant symmetries. It generalizes the familiar force memory kernel $\mathbf{G}(\mathbf{q}, t)$[1,8] which appears in the velocity correlation tensor because of momentum conservation; see Eq. (24) below.[30]

The coupling of the hydrodynamic poles into the result is made explicit in the term $\mathcal{P}R(s)\mathcal{P}$ in Eq. (14). The projector $\mathcal{P}$ from Eq. (12) leads to the appearance of the Laplace-transform of the auto-correlation functions of the conserved variables. On the one hand, the density correlator (intermediate scattering function) $\Phi_q(t) = \langle\delta\varrho(\mathbf{q}, t)^*\ \delta\varrho(\mathbf{q})\rangle/S_q$ enters; it is traditionally rescaled to unity at zero time. On the other hand, the collective velocity correlation functions enter, which shall be collected in the second order tensor $\mathbf{K}(\mathbf{q}, t)$. It is defined as

$$\mathbf{K}(\mathbf{q},t) = \langle\mathbf{v}(\mathbf{q},t)^*\ \mathbf{v}(\mathbf{q})\rangle. \tag{16}$$

Its initial value is diagonal and has already been stated: $\mathbf{K}(\mathbf{q},0) = \frac{k_BT}{m}\mathbf{1}$. In an isotropic system, the tensor of velocity correlations splits into a longitudinal (connected to compressional motion) and a transverse velocity autocorrelator,

$$\mathbf{K}(\mathbf{q},t) = \hat{\mathbf{q}}\hat{\mathbf{q}}\ K^{\parallel}_q(t) + (\mathbf{1}-\hat{\mathbf{q}}\hat{\mathbf{q}})\ K^{\perp}_q(t), \tag{17}$$

where both longitudinal and transverse correlators depend on the magnitude q of the wavevector only; the direction vector is denoted as $\hat{\mathbf{q}} = \mathbf{q}/q$. This follows from the analog of Eq. (10), which states that for a second rank tensor $\mathbf{K}(\mathbf{q},t) = \mathbf{R}^T \cdot \mathbf{K}(\mathbf{R}\mathbf{q},t)\cdot\mathbf{R}$ holds in isotropic systems.[1] Equivalent decompositions hold for all the following tensors of second rank.

C. General decomposition of stress auto-correlation tensor

Entering the correlators of the conserved variables and the memory kernel (15) into Eq. (14), the stress-correlation tensor can be rewritten using Einstein's sum convention

$$\begin{aligned} \mathrm{C}^{(\sigma)}_{\alpha\beta\gamma\delta}(\mathbf{q},s) = \mathrm{M}_{\alpha\beta\gamma\delta}(\mathbf{q},s) - \frac{1}{sS_q}\Big[\delta_{\alpha\beta}q_\zeta \mathrm{K}_{\zeta\eta}(\mathbf{q},s)q_\xi \mathrm{M}_{\eta\xi\gamma\delta}(\mathbf{q},s) + \mathrm{M}_{\alpha\beta\eta\xi}(\mathbf{q},s)q_\eta \mathrm{K}_{\xi\zeta}(\mathbf{q},s)q_\zeta\delta_{\gamma\delta}\Big] \\ + \frac{nk_BT}{S_q}\,\Phi_q(s)\,\delta_{\alpha\beta}\delta_{\gamma\delta} - \frac{1}{k_BTn}\Big\{\mathrm{M}_{\alpha\beta\zeta\xi}(\mathbf{q},s)q_\zeta\ \mathrm{K}_{\xi\xi'}(\mathbf{q},s)\ \mathrm{M}_{\zeta'\xi'\gamma\delta}(\mathbf{q},s)q_{\zeta'}\Big\}. \end{aligned} \tag{18}$$

Here we used the conservation laws, Eqs. (1) and (7), the relation $(s+i\mathcal{L})^{-1} = (1 - i\mathcal{L}(s+i\mathcal{L})^{-1})/s$, time reversal symmetry, and the result for the coupling between density and pressure fluctuations, $\langle\delta\varrho(\mathbf{q})^*\ \delta\boldsymbol{\sigma}(\mathbf{q})\rangle = k_BT\mathbf{1}$. It can easily be found using the conservation laws[1]

$$\frac{\mathbf{q}}{m}\cdot\langle\delta\varrho^*(\mathbf{q})\delta\boldsymbol{\sigma}(\mathbf{q})\rangle = \langle\delta\varrho^*(\mathbf{q})\mathcal{L}\mathbf{v}(\mathbf{q})\rangle = \mathbf{q}\cdot\langle\mathbf{v}^*\mathbf{v}\rangle = \frac{k_BT}{m}\mathbf{q}.$$

Equation (18) captures the coupling of longitudinal stress to density fluctuations, directly via $\Phi_q(t)$ and also via the longitudinal velocity correlator. It also leads to the appearance of the transverse currents in its last term, entering the form of $\mathbf{K}(\mathbf{q},s)$ from Eq. (17). The fourth order tensor $\mathbf{M}$ can be decomposed in complete analogy to Eq. (11) so that the components of the stress correlations can be expressed in terms of functions which only depend on the wavevector modulus q,

$$\begin{aligned} \mathrm{C}^{(1)}(q,s) = \mathrm{M}^{(1)}(q,s) + \frac{nk_BT}{S_q}\Phi_q(s) - \frac{q^2\mathrm{K}^{\parallel}_q(s)}{nk_BT} \\ \times\Big[\frac{2nk_BT}{sS_q}\mathrm{M}^{\parallel(1)}(q,s) + (\mathrm{M}^{\parallel(1)}(q,s))^2\Big], \end{aligned} \tag{19a}$$

$$\mathrm{C}^{(2)}(q,s) = \mathrm{M}^{(2)}(q,s), \tag{19b}$$

$$\begin{aligned} \mathrm{C}^{(3)}(q,s) = \mathrm{M}^{(3)}(q,s) - \frac{\mathrm{K}^{\parallel}_q(s)}{nk_BT}\,\mathrm{M}^{\parallel(2)}(q,s) \\ \times\Big[\mathrm{M}^{\parallel(1)}(q,s) + \frac{nk_BT}{sS_q}\Big], \end{aligned} \tag{19c}$$

$$\mathrm{C}^{(4)}(q,s) = \mathrm{M}^{(4)}(q,s) - \frac{\mathrm{K}^{\perp}_q(s)}{nk_BT}(\mathrm{G}^{\perp}_q(s))^2, \tag{19d}$$

$$\begin{aligned} \mathrm{C}^{(5)}(q,s) = \mathrm{M}^{(5)}(q,s) + \frac{4\mathrm{K}^{\perp}_q(s)}{nq^2k_BT}(\mathrm{G}^{\perp}_q(s))^2 \\ - \frac{\mathrm{K}^{\parallel}_q(s)}{nq^2k_BT}(\mathrm{M}^{\parallel(2)}(q,s))^2. \end{aligned} \tag{19e}$$

Here we introduced the following abbreviations:

$$\mathrm{M}^{\parallel(1)}(q,s) = \mathrm{M}^{(1)}(q,s) + q^2\mathrm{M}^{(3)}(q,s), \tag{20a}$$

$$\begin{aligned} \mathrm{M}^{\parallel(2)}(q,s) = 2\mathrm{M}^{(2)}(q,s) + q^2\mathrm{M}^{(3)}(q,s) \\ + 4q^2\mathrm{M}^{(4)}(q,s) + q^4\mathrm{M}^{(5)}(q,s), \end{aligned} \tag{20b}$$

which couple longitudinal velocity fluctuations into the stress fluctuations. Another combination of stress kernels appears twice in Eq. (19), which will play an important role later on, the generalized shear modulus,

$$\mathrm{G}^{\perp}_q(s) = \mathrm{M}^{(2)}(q,s) + q^2\mathrm{M}^{(4)}(q,s). \tag{21a}$$

Its familiar definition will be given in Eq. (24) below. There, the longitudinal modulus will also be obtained and consists of the sum

$$\mathrm{G}^{\parallel}_q(s) = \mathrm{M}^{\parallel(1)}(q,s) + \mathrm{M}^{\parallel(2)}(q,s). \tag{21b}$$

$\mathrm{C}^{(2)}(q,s)$ is the only stress component that is not coupled to the conserved densities; it approaches the frequency-dependent macroscopic shear stress correlator at vanishing wavevector [see Eqs. (23b) and (30) below].

D. Force auto-correlations

Only the divergence of the stress tensor enters momentum conservation Eq. (1). The projections along the wavevector simplify the stress to the force density defined in Eq. (3). Thus, the elements of $\mathbf{C}^{(\sigma)}$ combining to its divergence correspond to force auto-correlations (rescaled by $1/q^2$) and are of special interest.[31] They combine to a 2nd order tensor

$$\begin{aligned} \mathbf{Z}(\mathbf{q},t) = \hat{\mathbf{q}}\cdot\mathbf{C}^{(\sigma)}\cdot\hat{\mathbf{q}} = \frac{n}{k_BT\,q^2}\langle\mathbf{F}(\mathbf{q},t)^*\ \mathbf{F}(\mathbf{q})\rangle \\ = \hat{\mathbf{q}}\hat{\mathbf{q}}\ \mathrm{Z}^{\parallel}_q(t) + (\mathbf{1} - \hat{\mathbf{q}}\hat{\mathbf{q}})\ \mathrm{Z}^{\perp}_q(t), \end{aligned} \tag{22}$$

where the second line expresses the decomposition into longitudinal and transverse correlation functions. Newton's action-reaction law enforces $\mathbf{F}(\mathbf{q}\to 0)\to 0$ and cures the apparent divergence on the first line. From Eq. (11), it follows that

$$\begin{aligned} \mathrm{Z}^{\parallel}_q(t) = \mathrm{C}^{(1)}(q,t) + 2\mathrm{C}^{(2)}(q,t) + q^2(2\mathrm{C}^{(3)}(q,t) \\ + 4\mathrm{C}^{(4)}(q,t)) + q^4\mathrm{C}^{(5)}(q,t) \\ = \mathrm{C}^{\parallel(1)}(q,t) + \mathrm{C}^{\parallel(2)}(q,t), \end{aligned} \tag{23a}$$

$$\mathrm{Z}^{\perp}_q(t) = \mathrm{C}^{(2)}(q,t) + q^2\mathrm{C}^{(4)}(q,t), \tag{23b}$$

with $\mathrm{C}^{\parallel(1)}(q,t)$ and $\mathrm{C}^{\parallel(2)}(q,t)$ defined analogously to Eq. (20). The divergence of the stress memory tensor $\mathbf{M}$ corresponds to a correlation function of the fluctuating forces with reduced dynamics[32]

$$\mathbf{G}(\mathbf{q},t) = \hat{\mathbf{q}} \cdot \mathbf{M} \cdot \hat{\mathbf{q}} = \frac{n}{k_B T\, q^2} \langle \mathbf{F}(\mathbf{q})^* \mathcal{Q}\, e^{-i\mathcal{Q}\mathcal{L}\mathcal{Q}t} \mathcal{Q}\, \mathbf{F}(\mathbf{q})\rangle$$
$$= \hat{\mathbf{q}}\hat{\mathbf{q}}\, \mathrm{G}_q^{\parallel}(t) + (\mathbf{1} - \hat{\mathbf{q}}\hat{\mathbf{q}})\, \mathrm{G}_q^{\perp}(t). \tag{24}$$

Because of isotropy, the same relations as Eq. (23) connect the longitudinal and transverse components $\mathrm{G}_q^{\parallel}$ and $\mathrm{G}_q^{\perp}$ to the isotropic functions $\mathrm{M}^{(i)}$ with $i = 1, \ldots, 5$ of the reduced stress tensor $\mathbf{M}$; these relations were already identified in Eq. (21).

As announced when introducing the complete tensorial stress memory kernel $\mathbf{M}$, the force memory kernel $\mathbf{G}$ is familiar from the theory of liquids.[1,2] It is called (generalized) modulus and determines the correlation functions of the conserved variables; see Appendix A. The correlation of longitudinal velocity fluctuations follows from the longitudinal modulus

$$\mathrm{K}_q^{\parallel}(s) = \frac{k_B T/m}{s + \frac{\Omega_q^2}{s} + \frac{q^2}{mn}\mathrm{G}_q^{\parallel}(s)}, \tag{25a}$$

where the frequency $\Omega_q = \sqrt{\frac{q^2}{mn\kappa_q^T}}$ characterizes the initial decay; here, $\kappa_q^T = \frac{S_q}{nk_BT}$ denotes the generalized q-dependent isothermal compressibility. Some formulas will be shorter when this constant contribution is added to the modulus, which motivates the abbreviation: $\tilde{\mathrm{G}}_q^{\parallel}(t) = \frac{1}{\kappa_q^T} + \mathrm{G}_q^{\parallel}(t)$. Transverse velocity fluctuations are connected to the memory kernel of transverse fluctuating forces

$$\mathrm{K}_q^{\perp}(s) = \frac{k_B T/m}{s + \frac{q^2}{mn}\mathrm{G}_q^{\perp}(s)}. \tag{25b}$$

The memory kernel $\mathrm{G}_q^{\perp}$ is called shear modulus and generalizes the shear viscosity to finite frequencies and wavevectors.[1,2] Density fluctuations couple to longitudinal velocity fluctuations via the conservation law Eq. (7), which leads to the relation

$$\Phi_q(s) = \frac{1}{s}\left(1 - \frac{q^2}{sS_q}\,\mathrm{K}_q^{\parallel}(s)\right) = \left(s + \frac{\Omega_q^2}{s + \frac{q^2}{mn}\mathrm{G}_q^{\parallel}(s)}\right)^{-1}. \tag{25c}$$

In theoretical approaches, such as MCT, a rescaled memory kernel m_q is often used instead of the longitudinal one: $m_q(t) = \kappa_q^T\, \mathrm{G}_q^{\parallel}(t)$.

Besides the correlation functions of the conserved variables, the force correlation functions are also determined by the longitudinal and transverse moduli only. Using Eqs. (19) and (23)–(25), they are as follows:

$$\mathrm{Z}_q^{\parallel}(s) = \tilde{\mathrm{G}}_q^{\parallel}(s) - \frac{q^2\left(\tilde{\mathrm{G}}_q^{\parallel}(s)\right)^2}{mns + q^2\tilde{\mathrm{G}}_q^{\parallel}(s)}, \tag{26a}$$

$$\mathrm{Z}_q^{\perp}(s) = \mathrm{G}_q^{\perp}(s) - \frac{q^2(\mathrm{G}_q^{\perp}(s))^2}{mns + q^2\mathrm{G}_q^{\perp}(s)}. \tag{26b}$$

The transverse force auto-correlation function can be rewritten as found by Evans[11]

$$\mathrm{Z}_q^{\perp}(s) = \left[\frac{q^2}{mns} + \frac{1}{\mathrm{G}_q^{\perp}(s)}\right]^{-1}. \tag{27}$$

It is discontinuous at the origin of frequency and wavevector, viz., at q, $s = 0$.[20] Following Mori,[7,33] the force auto-correlations can be calculated bypassing the general decomposition in Eq. (18); see Appendix B. It serves as a check of the above derivation and provides the basis for proving that the force auto-correlations cannot possess time-persistent parts in glass; the proof follows Götze's[8] arguments.

IV. EMERGENCE OF LONG-RANGED STRESS CORRELATIONS

Section III C contains our principal result, the Zwanzig-Mori decomposition of the stress auto-correlation tensor into correlations of the conserved fields, viz., $\Phi_q(t)$, $\mathrm{K}_q^{\perp}(t)$, and $\mathrm{K}_q^{\parallel}(t)$, and stress memory kernels, viz., $\mathbf{M}(\mathbf{q}, t)$, defined in Eq. (15) and built with the reduced dynamics decoupled from the conservation laws. The achieved separation into conserved and non-conserved dynamics allows us to generalize Maxwell's insight into the glass transition to the wave number and frequency dependent stress correlations: only the non-conserved degrees of freedom encoded in $\mathbf{M}$ allow for a simple approximation, after the dynamics of the conserved densities has been split off.

The crucial difference between a fluid and a solid concerns the response under volume-conserving shear deformations; a fluid flows with a viscosity η, while a solid deforms dominantly elastically. In each case, the force transmitted by the stress through a surface element is tangential to the plane, viz., perpendicular to the surface normal. Thus in the following, we only consider off-diagonal elements of the cartesian stress auto-correlation tensor and postpone a discussion of the longitudinal elements to the future. The considered function $\mathrm{C}_{xyxy}^{(\sigma)}(\mathbf{q}, t)$[14,34] is the auto-correlation of the xy-component of the spatial Fourier-mode of the stress tensor, where any other off-diagonal element would be equivalent according to Eq. (11). The choice of the $\mathbf{x}$ and $\mathbf{y}$ direction, however, sets the coordinate system in the following.

A. Shear stress tensor auto-correlation

The shear stress auto-correlation, $\mathrm{C}_{xyxy}^{(\sigma)}(\mathbf{q}, s)$, can be obtained from the full tensorial analysis in Sec. III C. Equation (11) gives

$$\mathrm{C}_{xyxy}^{(\sigma)}(\mathbf{q}, t) = \mathrm{C}^{(2)}(q,t) + (q_x^2 + q_y^2)\mathrm{C}^{(4)}(q,t) + q_x^2 q_y^2 \mathrm{C}^{(5)}(q,t).$$

Combining it with the results for the $\mathrm{C}^{(i)}$ in Eq. (19), one finds

$$\mathrm{C}_{xyxy}^{(\sigma)}(\mathbf{q}, s) = \mathrm{M}_{xyxy}(\mathbf{q}, s) - \left(\frac{q_x^2 + q_y^2}{4} - \frac{q_x^2 q_y^2}{q^2}\right)\frac{(2\,\mathrm{G}_q^{\perp}(s))^2}{nk_BT}$$
$$\times\, \mathrm{K}_q^{\perp}(s) - \frac{q_x^2 q_y^2}{q^2}\frac{(\mathrm{M}^{\parallel(2)}(q,s))^2}{nk_BT}\,\mathrm{K}_q^{\parallel}(s). \tag{28}$$

Shear stress fluctuations are coupled to velocity fluctuations via stress memory kernels $\mathrm{G}_q^{\perp}(s)$ and $\mathrm{M}^{\parallel(2)}(q, s)$ [defined in Eqs. (21a) and (20b), respectively], which follows from the equation of momentum conservation. The auto-correlation functions of the velocities $\mathrm{K}^{\perp}$ and $\mathrm{K}^{\parallel}$ are given in Eq. (25). Even in isotropic materials, transverse motion and longitudinal motion are involved at finite wavevectors.

Maxwell describes the viscoelastic behavior of fluids undergoing a glass transition by a single relaxation time approximation for the global shear stress. In terms of stress correlations, this corresponds to

$$\mathrm{C}^{(\sigma)\,M}_{xyxy}(\mathbf{q}=0,s)=\frac{G_\infty\tau}{1+s\tau}. \tag{29}$$

Note that in this equation, Maxwell ignored the dependence on the wavenumber. Fluid behavior results, if $s\tau \ll 1$, so that $\lim_{s\to 0}\mathrm{C}^{(\sigma)\,M}_{xyxy}(\mathbf{q}=0,s)=G_\infty\tau=\eta^{\mathrm{M}}$ accounting for a viscous response. Solid-like behavior results, when the limit $s\tau \gg 1$ holds, implying $\lim_{s\to 0} s\mathrm{C}^{(\sigma)\,M}_{xyxy}(\mathbf{q}=0,s)=G_\infty$ accounting for an elastic response with a finite shear modulus G_∞. The transition from the viscous fluid to the elastic solid is signaled by a divergence of the relaxation time τ.

Even though this simple model has been very helpful for a qualitative understanding of viscoelasticity, it misses the hydrodynamic stress transport originating from the coupling to momentum fluctuations identified by the Zwanzig-Mori decomposition. Thus, as we will show, it misses the emergence of long-ranged stress correlations at the glass transition as well as the anisotropic stresses in the solid. These were discussed by Eshelby and have attracted a lot of attention in recent experiments on and simulations of glasses. In the following, we show how these deficiencies can be overcome by a generalization of Maxwell's model, using a single relaxation time approximation for the fast degrees of freedom only. Before doing so in Sec. IV E, we elaborate on the predictions of the general result [Eq. (28)] in the hydrodynamic limit $qa \ll 1$, where the length a can be taken as average particle distance.

B. Global stress

The global shear stress is included in the general result and recovered at vanishing wavevector, $\mathbf{q}=\mathbf{0}$. Assuming finite memory kernels, Eq. (28) then recovers the expected result of the Zwanzig-Mori formalism that the difference between projected and full dynamics vanishes[3]

$$\mathrm{C}^{(\sigma)}_{xyxy}(\mathbf{q}=\mathbf{0},t)=\mathrm{M}^{(2)}(q=0,t)=\mathrm{G}_0^{\perp}(t)\quad\text{(fluid)}. \tag{30}$$

In the last equality, we have introduced the macroscopic shear modulus [defined in Eq. (24) and related to $\mathbf{M}$ in Eq. (21a)] that is familiar from rheometry. Its Laplace-transform determines the transverse velocity correlator in the long-wavelength regime according to Eq. (25b) and reduces to the shear viscosity for vanishing frequency.[1,2] Equation (30) thus leads to the Green-Kubo relation for the viscosity,

$$\eta=\mathrm{G}_0^{\perp}(s=0)=\frac{n}{k_BT}\int_0^\infty dt\,\langle\delta\sigma_{xy}(\mathbf{q}=\mathbf{0},t)\delta\sigma_{xy}(\mathbf{q}=\mathbf{0})\rangle. \tag{31}$$

This result holds for fluid states, where memory kernels remain finite even in the limit $s\to 0$.

C. Hydrodynamics of fluids

Equation (28) is valid for arbitrary frequencies and wavevectors and contains microscopic motion on short time and length scales. For example, the initial value of the stress recovers the "high-frequency elastic moduli" determined by Zwanzig and Mountain.[35] Yet, the Zwanzig-Mori formalism is tailored to smooth and slow fluctuations, where approximation schemes for the memory kernels have been developed.[2,3,8] To recover the hydrodynamics of fluids, memory kernels like the tensor $\mathbf{M}$ from Eq. (15) can be replaced by wavevector and frequency independent transport coefficients denoted as viscosities, $\boldsymbol{\eta}=\mathbf{M}(\mathbf{q}\to 0,\ s\to 0)$. This rests on the property of the reduced Liouvillian $\mathcal{QLQ}$ which is decoupled from the slow dynamics of conserved variables.[3] In an isotropic medium, only two components of the viscosity tensor survive in the limit $\mathbf{q}\to 0$, namely, the shear viscosity $\eta=M^{(2)}(q\to 0,\ s\to 0)$ and the bulk viscosity $\eta_B=M^{(1)}(q\to 0,\ s\to 0)$. The shear stress correlation function reads in this limit

$$\begin{aligned}\mathrm{C}^{(\sigma)\mathrm{fluid}}_{xyxy}(\mathbf{q},s)=\eta+\Big(\frac{q_x^2q_y^2}{q^2}-\frac{q_x^2+q_y^2}{4}\Big)\frac{4\,\eta^2}{nms+\eta q^2}\\ -\frac{q_x^2q_y^2}{q^2}\frac{4\,s\,\eta^2}{nms^2+q^2/\kappa_q^T+s\eta^{\parallel}q^2},\end{aligned} \tag{32}$$

where we have introduced the longitudinal viscosity $\eta^{\parallel}=\eta_B+4\eta/3$. Clearly, the hydrodynamic velocity correlators introduce slow shear momentum diffusion (first line) and damped compressional wave motion (second line) into the shear stress correlation. Most remarkable is the strong anisotropy in $\mathbf{q}$-space of both contributions. Even though the fluid is isotropic, the shear stress correlation at any finite frequency is strongly anisotropic, reflecting the tensorial character of the observable σ_{xy}. Furthermore the stress correlations become long-ranged as the glass transition is approached, signalled by a divergence of the shear viscosity η. The spatial decay at fixed frequency ω is characterized by the diffusive length $\sqrt{\eta/(mn\omega)}$, which grows as the glass transition is approached because the viscosity grows.

The real part of the frequency dependent correlation, viz., $\Re\{\mathrm{C}^{(\sigma)\mathrm{fluid}}_{xyxy}(\mathbf{q},s=-i\omega)\}$, can be considered a viscosity-like quantity; it is shown in Fig. 1. For finite frequencies, it approaches the Newtonian viscosity for vanishing wavevector as expected from the Green-Kubo relation, $\lim_{\omega\to 0}\lim_{q\to 0}\Re\{\mathrm{C}^{(\sigma)\mathrm{fluid}}_{xyxy}(\mathbf{q},s=-i\omega)\}\ \to\ \eta$. It is reduced relative to the Newtonian viscosity by both transport processes, vorticity diffusion and compressional waves, because particles move with the velocity fields to reduce stresses. Along the diagonals ($\mathbf{q}=\pm q(\hat{\mathbf{x}}\pm\hat{\mathbf{y}})/\sqrt{2}$), stress fluctuations are reduced only by compressional waves. For all other directions, vorticity diffusion lowers the frequency-dependent correlation for increasing wavevector. Along the x- or y-direction, the stress correlation vanishes for vanishing frequency at finite q, for example $\mathrm{C}^{(\sigma)\mathrm{fluid}}_{xyxy}(\mathbf{q}=q\hat{\mathbf{x}},s)=\frac{nms\,\eta}{nms+\eta q^2}$. This explains the decay of $\mathrm{C}^{(\sigma)\mathrm{fluid}}_{xyxy}$ with increasing $q\sqrt{\eta/(mn\omega)}$ along the axes. Appendix C provides additional details on the origin of the angular variations. To demonstrate these anisotropies, we plot in Fig. 1 the frequency dependent shear stress correlation at fixed frequency ω.

D. Hydrodynamics of glasses

1. Elasticity of glasses

The q-dependence of the stress correlations as predicted by generalized hydrodynamics [Eq. (28)] is completely

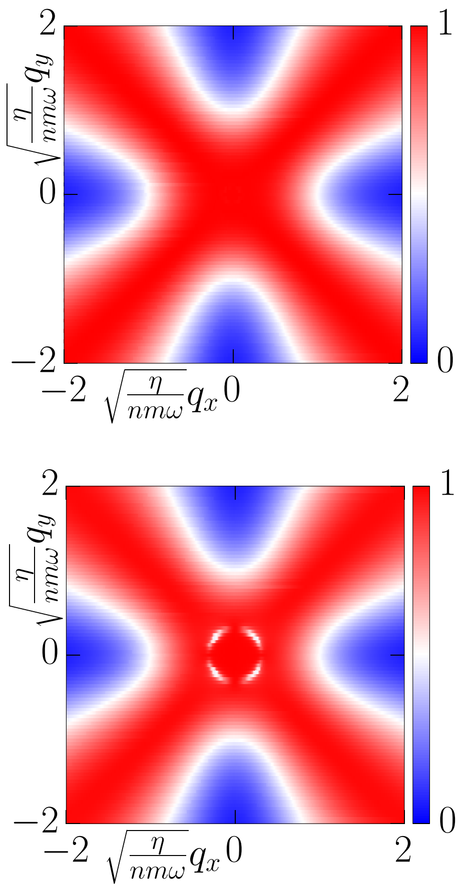

FIG. 1. Rescaled real part of the frequency dependent shear stress autocorrelation $\Re\{C^{(\sigma)\text{fluid}}_{xyxy}(\mathbf{q}, s = -i\omega)\}/\eta$ [from Eq. (32)] of a fluid state in the q_x and q_y plane (viz., at $q_z = 0$); the rescaled wavevector $\tilde{\mathbf{q}} = \sqrt{\eta/(mn\omega)}\mathbf{q}$ contains the shear diffusion length at the given frequency. The isothermal compressibility is chosen as $\kappa_0^T = 0.01/(\omega\eta)$ (upper panel) so that longitudinal corrections are negligible and as $\kappa_0^T = 0.1/(\omega\eta)$ (lower panel) so that they are visible mainly along the diagonals. Here compressional waves reduce the correlations, giving rise to the small white stripes. Throughout, $\eta^\parallel = \frac{4}{3}\eta$ is set.

ignored in the simple ansatz of Maxwell [Eq. (29)]. Yet, his basic idea of a diverging relaxation time at the glass transition is widely confirmed, *inter alia* in the microscopic approach of MCT,[36,37] where a diverging relaxation time has been identified as the indicator of an idealized glass transition. It arises from an arrest of the structural relaxation in fluids.[8] As a consequence of the infinite relaxation time in the glassy state, structural correlation functions no longer decay to zero. This should hold for collective density fluctuations $\Phi_q(t)$ and the fluctuating force memory kernels $\mathbf{G}(\mathbf{q}, t)$. We extend this idea for the definition of an idealized glass state to the stress memory tensor

$$\mathbf{M}(\mathbf{q}, t) \to \mathbf{M}_\infty(\mathbf{q}) \quad \text{for } t \to \infty \quad \text{(ideal glass state)} \tag{33}$$

corresponding to $\mathbf{M}(\mathbf{q}, s) \to \frac{1}{s}\mathbf{M}_\infty(\mathbf{q})$ in the limit $s \to 0$ of the Laplace transformation. As a consequence, the shear stress correlation tensor acquires a time persistent contribution: $C^{(\sigma)}_{xyxy}(\mathbf{q}, t) \to C^{(\sigma)\infty}_{xyxy}(\mathbf{q})$ for $t \to \infty$. Isolating this contribution in Eq. (28) and making use of Eqs. (25), (20b), and (21a), we find for the time persistent part of the shear stress correlation

$$\begin{aligned} C^{(\sigma)\infty}_{xyxy}(\mathbf{q}) &= G^\perp_{q,\infty} - q_z^2\, M^{(4)}_\infty(q) + q_x^2 q_y^2\, M^{(5)}_\infty(q) \\ &\quad + \left(4\frac{q_x^2 q_y^2}{q^4} - \frac{q_x^2 + q_y^2}{q^2}\right) G^\perp_{q,\infty} - \left(\frac{q_x^2 q_y^2}{q^4}\right) \\ &\quad \times \frac{(M^{\parallel(2)}_\infty(q))^2}{\tilde{G}^\parallel_{q,\infty}}. \end{aligned} \tag{34}$$

The elasticity of the glass phase is entailed in the long-wavelength limit of the time persistent stress correlations, which can be obtained from Eq. (34) in the limit $q \to 0$

$$\begin{aligned} C^{(\sigma)\infty}_{xyxy}(\mathbf{q}) &= \left(4\frac{q_x^2 q_y^2}{q^4} + \frac{q_z^2}{q^2}\right) G^\perp_{0,\infty} - \left(4\frac{q_x^2 q_y^2}{q^4}\right) \\ &\quad \times \frac{(G^\perp_{0,\infty})^2}{\tilde{G}^\parallel_{0,\infty}} + \mathcal{O}((qa)^2). \end{aligned} \tag{35}$$

The naive expectation, which relates Maxwell's constant shear modulus, G_∞, to the frozen-in shear stress correlations, clearly fails, because the limit $q \to 0$ is non-analytic. The result on the direction $\mathbf{q}$ is taken to zero. The non-analytic behavior can be traced back to the conserved momentum correlations, which are long-ranged in an elastic solid. This non-analytic behavior points to the existence of a length scale, which is infinite in the glassy phase and diverges in the supercooled regime when approaching the glass transition. It will be discussed in Sec. IV E.

Fourier transformation of Eq. (35) reproduces Eshelby's result for the response of an elastic (isotropic) medium to a point shear at the origin.[38,39] It predicts the existence of long-ranged stress correlations

$$\begin{aligned} C^{(\sigma)\infty}_{xyxy}(\mathbf{r}) \to{}& \frac{3}{4\pi}\,\frac{G^\perp_{0,\infty}}{r^3}\,\frac{10x^2y^2 - r^2(x^2+y^2)}{r^4} \\ &- \frac{3}{4\pi}\,\frac{(G^\perp_{0,\infty})^2}{\tilde{G}^\parallel_{0,\infty}}\,\frac{10x^2y^2}{r^7} \quad \text{for } r \to \infty. \end{aligned} \tag{36}$$

In d dimensions, the elastic correlations decay like r^{-d};[39] see Appendix C for details.

2. *Shear stress dynamics in glass*

While glass possesses frozen-in stress correlations, local motion of particles causes stress fluctuations. Shear stresses participate in the transport by sound waves, by longitudinal and transverse ones, and in relaxation processes which contribute to the sound damping. In this paragraph, we show that generalised hydrodynamics is rich enough to account for these dynamic phenomena, going beyond solid elasticity as discussed in the last paragraph. However, our focus is shear stress correlations near the glass transition. Hence we just quote the results and postpone a careful discussion to future work.

Temporal correlation functions describing non-ergodic states shall be denoted by a caret such that $\hat{\mathbf{C}}^{(\sigma)}(\mathbf{q}, t) = \mathbf{C}^{(\sigma)}(\mathbf{q}, t) - \mathbf{C}^{(\sigma)\infty}(\mathbf{q})$ captures the decorrelation of stress fluctuations in the glass. Similarly, $\hat{\mathbf{M}}(\mathbf{q}, t)$ and $\hat{\mathbf{G}}(\mathbf{q}, t)$ denote the memory kernels devoid of frozen-in terms. Equation (28) can then be rewritten in order to describe the shear stress dynamics in glass on all length and time scales (not shown). Especially, the limit of long wavelengths is of interest, where longitudinal and transverse sound waves are expected according to elasticity theory. Their sound speeds will be denoted as $v_\parallel$ and v_T, respectively, and are given by the limits: $v_\parallel^2 = \tilde{\mathrm{G}}_0^\parallel(t \to \infty)/(mn)$ and $v_T^2 = \mathrm{G}_0^\perp(t \to \infty)/(mn)$. Transport coefficients follow from the low frequency limit of the (reduced) memory kernels at vanishing wavevector. Only $\zeta^\parallel = \hat{\mathrm{G}}_0^\parallel(s \to 0)/(mn)$ and $\zeta^\perp = \hat{\mathrm{G}}_0^\perp(s \to 0)/(mn)$ enter the hydrodynamic stress correlations close to the sound poles, when $s/(qv)$ is of order unity

$$\hat{\mathbf{C}}_{xyxy}^{(\sigma)\mathrm{glass}}(\mathbf{q}, s) = mn\zeta^\perp + \left(mnv_T^2\right)\left(1 - \frac{4q_x^2q_y^2}{q^4} - \frac{q_z^2}{q^2}\right) \times \frac{\left(s - \zeta^\perp q^2\right)}{s^2 + q^2v_T^2 + sq^2\zeta^\perp} + \frac{4q_x^2q_y^2}{q^4}\frac{\left(\frac{v_T^2}{v_\parallel^2}(s + \zeta^\parallel q^2) - 2\zeta^\perp q^2\right)}{s^2 + q^2v_\parallel^2 + sq^2\zeta^\parallel}. \quad (37)$$

In the glass, the shear stress correlations differ from the transport coefficient $\zeta^\perp$ because of the coupling to wave motion. Compressional and transverse sound waves decorrelate stresses of the strength $\mathrm{G}_{0,\infty}^\perp$, while the more rapid decay processes, which contribute the constant $\zeta^\perp$ to the spectrum, are not resolved in the hydrodynamic description.

E. Generalized Maxwell model

The Maxwell model [Eq. (29)] captures the fluid and solid behavior of the macroscopic stress response. Intriguingly, the finite-wavevector results obtained from the Zwanzig-Mori projection operator formalism show that Maxwell's model misses the long-ranged stress correlations of an elastic solid. Therefore, Maxwell's model cannot be applied to the full shear-stress correlation function $\mathrm{C}_{xyxy}^{(\sigma)}(\mathbf{q}, t)$—not even for $q \to 0$. The generalized hydrodynamics approach pioneered by Ref. 40 and transferred to supercooled liquids by Ref. 29 allows us to separate the fast relaxation processes from the slow dynamics of conserved variables. The natural extension of Maxwell's theory is then the single relaxation time approximation for the fast degrees of freedom only.

The following discussion simplifies in the limit of an incompressible fluid, where the longitudinal velocity correlations can be neglected relative to the transverse ones. This holds, because the longitudinal velocity correlator obeys $\mathrm{K}_q^\parallel(s) \leq \frac{ns}{q^2}\kappa_q^T$, and thus can be neglected relative to the transverse one in the hydrodynamic limit if the isothermal compressibility κ_0^T is small. Then, dropping the local contributions of higher order in wavevector, e.g., in $\mathrm{M}_{xyxy}(\mathbf{q}, s) \approx \mathrm{G}_0^\perp(s) + \mathcal{O}((qa)^2)$, a single memory kernel, namely, the global shear stress modulus $\mathrm{G}_0^\perp(s)$ [defined in Eq. (24)], determines the model in the hydrodynamic limit. We suggest a generalised Maxwell model, marked by the suffix $^{\mathrm{gM}}$, for the global shear stress modulus,

$$\mathrm{G}_0^\perp(s) \approx \mathrm{G}^{\mathrm{gM}}(s) = \frac{G_\infty \tau}{1 + s\tau}. \quad (38a)$$

It encodes the structural relaxation which dominates the fluctuating forces in glass-forming fluids. Let us reiterate the crucial difference to the classic Maxwell model: The generalized model first accounts for the coupling of the stress to hydrodynamic modes and then approximates the remaining fluctuating forces. Maxwell's model, by contrast, neglects the coupling of the stress to transverse momenta and approximates the correlations of the full stress fluctuations.

The model predicts the far-field shear stress correlation function in the incompressible fluid as follows from the simplification of Eq. (28):

$$\mathrm{C}_{xyxy}^{(\sigma)\mathrm{gM}}(\mathbf{q}, s) \approx \left(\frac{q^2}{mns} + \frac{1}{\mathrm{G}^{\mathrm{gM}}(s)}\right)^{-1} + \left(4\frac{q_x^2q_y^2}{q^4} + \frac{q_z^2}{q^2}\right)\frac{q^2\,(\mathrm{G}^{\mathrm{gM}}(s))^2}{nms + q^2\mathrm{G}^{\mathrm{gM}}(s)}. \quad (38b)$$

In the spirit of Maxwell, we take the relaxation time to diverge at the glass transition and remain infinite in the solid phase. The generalised model then includes both, the fluid limit, as given in the first line of Eq. (32), for $s\tau \ll 1$ and the elasticity of the solid [Eq. (35)] for $s\tau \gg 1$. It misses sound damping as described in Sec. IV D 2.

The term on the first line is the Laplace-transform of the long-wavelength limit of the transverse force correlation function $\mathrm{Z}_q^\perp(t)$, defined in Eq. (22); it will be denoted as $\mathrm{Z}_q^{\mathrm{gM}}(s)$. It was expressed in terms of the shear modulus in Eq. (27) and takes the given form when only the explicit q-dependence arising from momentum conservation is retained. Equation (38b) shows that the transverse force correlation function describes the stress correlation function only if the wavevector lies along one of the directions of the stress tensor itself: $\mathrm{C}_{xyxy}^{(\sigma)}(\mathbf{q} = q\hat{\mathbf{x}}, t) = \mathrm{Z}_q^\perp(t)$ holds for $\mathbf{q}$ along the x- or y-direction. This relation is also valid according to the exact Zwanzig-Mori decomposition Eq. (28). In general directions, the term on the second line in Eq. (38b) appears, which originates in the hydrodynamic transverse velocity correlator given in Eq. (25b). Along the diagonals in the x, y-plane, the stress correlations reduce to the Maxwell modulus, $\mathrm{C}_{xyxy}^{(\sigma)}(\mathbf{q}^T = q(1, \pm 1, 0)/\sqrt{2}, t) = \mathrm{G}^{\mathrm{gM}}(t)$, and are independent of the modulus of the wavevector.

1. Critical behavior at the glass transition

The generalised Maxwell model not only captures the correct long wavelength behavior of both, the fluid and the glass, it furthermore fully accounts for the transition between the two phases. The distance to the idealised glass transition is controlled by the divergence of τ which we do not specify explicitly. Introducing a characteristic length scale, $\xi^2 = \frac{G_\infty \tau^2}{mn} = v_T^2\tau^2$, written in terms of the transverse sound velocity v_T of the glass (Refs. 8 and 41), we observe that the time and wavenumber dependent stress correlation obeys scaling

$$\mathrm{C}_{xyxy}^{(\sigma)\mathrm{gM}}(\mathbf{q}, t) = G_\infty\, \mathcal{F}(t/\tau, \mathbf{q}\xi), \quad (39)$$

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 solid like 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.[42,43] In the glass, the correlation length remains infinite in accordance with the algebraic decay of stress correlations.

The hydrodynamic excitations are determined by the poles of $C^{(\sigma)\mathrm{gM}}_{xyxy}(\mathbf{q}, s)$ 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 $s\tau \ll 1$, 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 and high frequency sound in the fluid phase.[8,41] The sound velocity remains approximately constant through the glass transition because of the linear scaling of frequency and wavenumber and $\xi \propto \tau$.

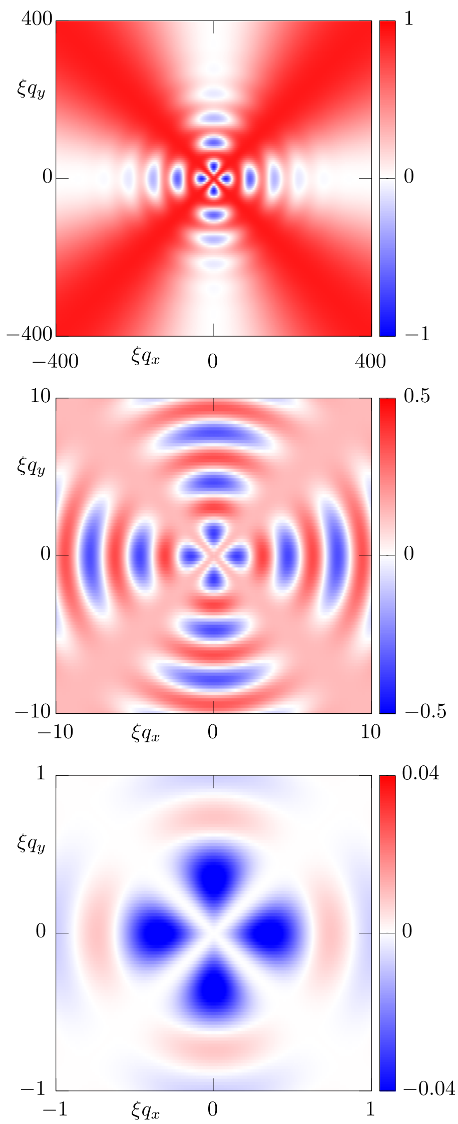

FIG. 2. Scaling function $C^{(\sigma)\mathrm{gM}}_{xyxy}(\mathbf{q}, t)/G_\infty = \mathcal{F}(t/\tau, \mathbf{q}\xi, \tau/\tau_0)$ of the generalized Maxwell model in the q_x and q_y plane at $q_z = 0$ for three rescaled times t/τ and including an instantaneous damping. Top panel in the glassy regime for $t/\tau = 10^{-1}$, middle panel in the viscoelastic regime for $t/\tau = 2$, and bottom panel in the fluid regime for $t/\tau = 10^1$. The microscopic time is chosen as $\tau/\tau_0 = 10^3$.

Damping of transverse sound modes in the solid state is neglected in the ansatz (38a), which disagrees with the contents of the full theory, as discussed in Sec. IV D 2. One can simply introduce an instantaneous damping of strength Γ in order to account approximately for dissipation in the solid state: $G^{\mathrm{gM}}(s) = \frac{G_\infty \tau}{1+s\tau} + \Gamma$. Defining $\tau_0 = \Gamma/G_\infty$ shows that Γ actually introduces a time scale τ_0 encoding fast dissipative processes; obviously, $\tau \gg \tau_0$ needs to hold. Figure 2 shows the rescaled stress correlation functions $C^{(\sigma)\mathrm{gM}}_{xyxy}(\mathbf{q}, t)/G_\infty = \mathcal{F}(t/\tau, \mathbf{q}\xi, \tau/\tau_0)$ at fixed rescaled times t/τ in the q_x and q_y plane at $q_z = 0$. For short times, e.g., $t = 0.1\tau$ (upper panel), stresses behave like in a solid. They are high along the diagonals because $G^{\mathrm{gM}}(\tau_0 \ll t \ll \tau) \approx G_\infty$. Along the axes, stresses have been relaxed by particle motion in (damped) transverse sound waves. For long times, e.g., $t = 10\tau$ (lower panel), stress correlations characteristic for a fluid state are visible. Along the diagonals, shear stresses have been relaxed by the structural processes encoded in the shear modulus $G^{\mathrm{gM}}(t)$. Along the axes, a wavevector dependence originates from the transverse momentum diffusion which crosses over to transverse waves around $q\xi \approx \frac{1}{2}$. For intermediate times, $t = 2\tau$ is chosen in the middle panel of Fig. 2, the oscillatory dependence on $q\xi$ originating from the sound waves is visible along the axes. Along the diagonals, the Maxwell modulus has partially decayed relative to its initial value. (Note the different wavevector scales in the panels of Fig. 2.) Comparing Fig. 2 with damping to Fig. 1 in Ref. 13 without damping, qualitative differences are observed only in the glassy state: Transverse sound waves, which arise along the axes $\mathbf{q} = q\hat{\mathbf{x}}$ and $\mathbf{q} = q\hat{\mathbf{y}}$, now are damped so that stresses decorrelate in these directions.

2. Spectra

Rheological measurements can record frequency dependent spectra, which are obtained by Fourier-transformation

$$C^{(\sigma)\mathrm{gM}\prime}_{xyxy}(\omega) + iC^{(\sigma)\mathrm{gM}\prime\prime}_{xyxy}(\omega) = i\omega C^{(\sigma)\mathrm{gM}}_{xyxy}(\mathbf{q}, s = -i\omega).$$

Note the conventional prefactor $i\omega$ which renders the storage part $C^{(\sigma)\prime}$ (the loss part $C^{(\sigma)\prime\prime}$) in phase with the strain (strain rate). In order to compute such spectra from our generalised hydrodynamic model, we again include the instantaneous damping to account for dissipation in the solid state.

Three physical quantities characterize $C^{(\sigma)\mathrm{gM}}_{xyxy}(\mathbf{q}, t)$ for finite $\Gamma > 0$: the generalized transverse sound frequency $\omega^s(q)$, a damping rate $\gamma(q)$, and the generalized elastic modulus $G(q)$. They are given by

$$\omega^s(q) = \frac{\sqrt{(2\xi q)^2 - 1 + 2x - x^2}}{2\tau}, \quad x = \frac{\Gamma q^2 \xi^2}{\eta^{\mathrm{M}}}, \tag{40a}$$

$$\gamma(q) = \frac{1}{2}\left(\frac{1}{\tau} + \frac{\Gamma q^2}{mn}\right), \tag{40b}$$

$$G(q) = G_\infty - \frac{\Gamma^2 q^2}{mn}, \tag{40c}$$

and enter the generalized shear stress modulus and the transverse force correlation function

$$G^{gM}(t) = G_\infty\, e^{-\frac{t}{\tau}} + 2\Gamma\, \delta(t), \tag{40d}$$

$$Z_q^{gM}(t) = 2\Gamma\, \delta(t) + G(q) \exp\{-\gamma(q)\, t\} \times [\cos(\omega^s(q)\, t) - \alpha(q) \sin(\omega^s(q)\, t)], \tag{40e}$$

with

$$\alpha(q) = \frac{\gamma(q)}{\omega^s(q)} + \frac{\Gamma q^2}{nm}\left(G_\infty + \frac{\Gamma}{\tau}\right)\frac{1}{\omega^s(q) G(q)}. \tag{40f}$$

The full tensorial form of the stress correlations in the generalized Maxwell model then takes the form

$$C_{xyxy}^{(\sigma)gM}(\mathbf{q}, t) = Z_q^{gM}(t) + \left(4\frac{q_x^2 q_y^2}{q^4} + \frac{q_z^2}{q^2}\right)\left[G^{gM}(t) - Z_q^{gM}(t)\right]. \tag{40g}$$

While the shear stress modulus $G^{gM}(t)$ only contains relaxational contributions and is wavevector independent, the transverse force correlation function $Z_q^{gM}(t)$ shows differing behavior depending on the wavevector. For $2q\xi \geq 1$ (at $\Gamma = 0$), it exhibits transverse sound waves. For smaller q, it is non-monotonic and a superposition of exponential decays.[13]

Typical rheological spectra of supercooled liquids for finite wavevectors are shown in Fig. 3. They depend on the modulus q and the direction $\hat{\mathbf{q}}$ of the wavevector even in the limit of $q \to 0$ for glassy states. To demonstrate the two dynamic contributions, we choose $q_x = q_y = q/\sqrt{2}$ in the upper panel so that $C_{xyxy}^{(\sigma)gM}(\mathbf{q}, t) = G^{gM}(t)$. Purely relaxational dynamics is observed, and the low-frequency peaks in the different curves could be collapsed to a single one if plotted versus $\omega\tau$. In the lower panel, we have chosen $q_x = q_z = 0$, so that $C_{xyxy}^{(\sigma)gM}(\mathbf{q}, t) = Z_q^{gM}(t)$, and the spectrum of the transverse force correlation function Z_q^{gM} can be seen. A damped resonance develops in the latter when the Maxwell relaxation hump at $\omega\tau \approx 1$ shifts down to the frequency of the transverse sound wave.

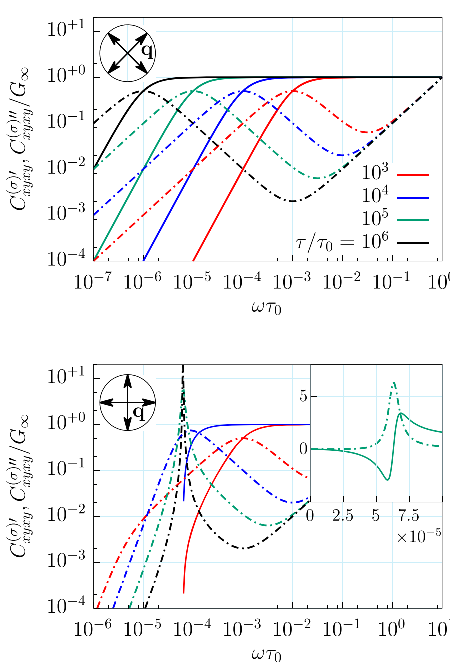

FIG. 3. Frequency dependent rheological shear stress spectra obtained from the generalized Maxwell model at finite wavevector: storage part $C_{xyxy}^{(\sigma)gM\prime}$ (solid lines) and loss part $C_{xyxy}^{(\sigma)gM\prime\prime}$ (dashed lines) with $q_z = 0$ are shown for different Maxwell relaxation times τ (see legend) and along different wavevector directions: (upper panel) $q_x = q_y = \frac{q}{2}$ and (lower panel) $q_x = 0$ (C' for $\tau/\tau_0 = 10^5$ is shown only in the linear plot given as inset). Frequency is rescaled by the time scale of instantaneous dissipation, $\tau_0 = \Gamma/G_\infty$. The wavevector magnitude q is set to $v_T q \tau_0 = 2\pi \cdot 10^{-5}$.

TABLE I. Parameters of the generalized Maxwell model and their relation to limits of the shear modulus; the last column gives additional notations used in the text.

gM model	ZM kernel	Symbol
η^{gM}	$G_0^\perp(s = 0)$	η
G_∞	$G_0^\perp(t \to \infty)$	$G_{0,\infty}^\perp$
Γ	$\hat{G}_0^\perp(s = 0)$	$\zeta^\perp$

It is straightforward to connect the generalized Maxwell model to the exact Zwanzig-Mori decomposition. Table I contains the mapping of the parameters of the model to the limiting values of the memory kernels collected and discussed in Secs. IV B–IV D 2.

3. Real space structure

The shear stress correlations in real space and as a function of time are of interest to spatially resolved investigations. The scaling law Eq. (39) in real space takes the form (neglecting the short-time part, viz., $\Gamma = 0$)

$$C_{xyxy}^{(\sigma)gM}(\mathbf{r}, t) = \frac{G_\infty}{\xi^3}\, \mathcal{F}(t/\tau, \mathbf{r}/\xi). \tag{41}$$

The scaling function combines oscillatory with decaying structures. Anticipating, that the correlation length ξ can be made large, so that a window opens between the microscopic and the correlation length, $a \ll r \ll \xi$, the limit $\xi \to \infty$ can be taken in Eq. (41). One finds

$$C_{xyxy}^{(\sigma)gM}(\mathbf{r}, t) \to \frac{3}{4\pi}\frac{G_\infty}{r^3}\frac{10x^2y^2 - r^2(x^2 + y^2)}{r^4}\, e^{-t/\tau}. \tag{42}$$

For distances small compared to the correlation length, solid-like correlations extend according to Eq. (36) (viz., like a power-law, $\propto r^{-d}$ for $r \gg a$), with an amplitude that decays as a function of time according to the structural relaxation process.

V. LINEAR RESPONSE AND HYDRODYNAMIC EQUATIONS

So far, we have discussed the stress auto-correlation and have shown that it reveals the emergence of long-ranged stress correlations as the glass transition is approached. An equivalent, alternative description is the linear response function that is accessible to experiment by applying a weak inhomogeneous velocity field to the liquid. Discussing the response to an externally applied velocity, $\mathbf{v}^{ext}$, furthermore allows for an intuitive interpretation of our results in terms of hydrodynamic equations.

A. Linear response

Following textbook and older approaches,[20,44] the linearized Liouville equation for a perturbation of the distribution function $\delta\Psi$ in phase space is

$$(\partial_t + i\mathcal{L})\delta\Psi = \frac{1}{k_B T}\dot{\mathrm{H}}\Psi_{\mathrm{eq}}. \tag{43}$$

It contains the Liouville operator $\mathcal{L}$ and the canonical equilibrium distribution Ψ_{eq} of the unperturbed system. Kinetic contributions shall be neglected because they are negligible in supercooled liquids, which gives $\dot{\mathrm{H}} = \dot{\mathrm{U}}$ with the total potential energy U. The change $\dot{\mathrm{U}}$ arises from the applied velocity field $\mathbf{v}^{\mathrm{ext}}(\mathbf{r}, t)$ as it displaces particles

$$\dot{\mathbf{r}}_j = \int d^d r \phi(\mathbf{r}_j - \mathbf{r})\mathbf{v}^{\mathrm{ext}}(\mathbf{r}, t). \tag{44}$$

Here, the symmetric function $\phi(\Delta\mathbf{r})$ describes the region of influence of the applied flow field on the particles.[45] The choice of a Dirac delta, $\phi(\Delta\mathbf{r}) = \delta(\Delta\mathbf{r})$ will lead to the Irving-Kirkwood microscopic expression of the stress tensor, while a more spread-out weighting function is often used in non-equilibrium molecular dynamics simulations.[20] The change in potential energy thus becomes

$$\begin{aligned}\dot{\mathrm{U}} &= \frac{d}{dt}\sum_{\langle j,k\rangle} U(r_{jk}) = \sum_{\langle j,k\rangle} U'(r_{jk})\frac{\mathbf{r}_{jk}}{r_{jk}}\cdot\left(\dot{\mathbf{r}}_j - \dot{\mathbf{r}}_k\right)\\ &= \sum_{\langle j,k\rangle} U'(r_{jk})\hat{\mathbf{r}}_{jk}\cdot\int d^d r\mathbf{v}^{\mathrm{ext}}(\mathbf{r},t)\int_0^1 ds\partial_s\Phi(\mathbf{r}-\mathbf{r}_k-s\mathbf{r}_{jk})\\ &= \sum_{\langle j,k\rangle} U'(r_{jk})\hat{\mathbf{r}}_{jk}\mathbf{r}_{jk}\colon\int d^d r\int_0^1 ds\Phi(\mathbf{r}-\mathbf{r}_k-s\mathbf{r}_{jk})\nabla\mathbf{v}^{\mathrm{ext}}(\mathbf{r},t)\\ &= \sqrt{N}\int d^d r\,\boldsymbol{\sigma}(\mathbf{r}) : \nabla\mathbf{v}^{\mathrm{ext}}(\mathbf{r},t).\end{aligned} \tag{45}$$

In the second line, Eq. (44) was employed. In the last but one line, $\partial_s\Phi(\mathbf{r} - s\mathbf{r}_{jk}) = -\mathbf{r}_{jk}\cdot\nabla_{\mathbf{r}}\Phi(\mathbf{r} - s\mathbf{r}_{jk})$ was used and a partial integration performed neglecting surface terms at infinity. In the last line, the (generalized) Irving-Kirkwood expression for the spatially resolved stress tensor was identified from Eq. (5); the unfamiliar prefactor $\sqrt{N}$ follows from its definition in Eq. (5), which is aimed to simplify the equations for the correlation functions. The formal solution for the perturbation of the distribution function $\delta\Psi$ follows now from Eqs. (43) and (45) and can immediately be applied to obtain the linear response of the stress tensor to the external velocity gradient. Note that we follow the standard convention on normalizing the linear-response stress to be energy per volume (and denote the phase space volume element as $d\Gamma$)

$$\begin{aligned}\langle\delta\boldsymbol{\sigma}(\mathbf{r},t)\rangle^{\mathrm{lr}} &= \frac{\sqrt{N}}{Vk_B T}\int_{-\infty}^{t}dt'\int d\Gamma\delta\boldsymbol{\sigma}(\mathbf{r})e^{-i\mathcal{L}(t-t')}\dot{\mathrm{U}}(\mathbf{r}',t')\Psi_{\mathrm{eq}}\\ &= \int_{-\infty}^{t}dt'\int d^d r'\,\mathbf{C}^{(\sigma)}(\mathbf{r}-\mathbf{r}',t-t') : \bar{\boldsymbol{\kappa}}^{\mathrm{ext}}(\mathbf{r}',t').\end{aligned} \tag{46}$$

Because of the symmetry of the Irving-Kirkwood stress tensor, only the symmetric component of the external velocity gradient enters, with $\bar{\boldsymbol{\kappa}}(\mathbf{r},t) = \frac{1}{2}\left(\nabla\mathbf{v}(\mathbf{r},t) + (\nabla\mathbf{v}(\mathbf{r},t))^T\right)$. This calculation verifies the expected fluctuation dissipation relation. The stress auto-correlation tensor defined in Eq. (8) describes the linear response of the local stress at time t to the (symmetric) velocity gradients applied at earlier times t' at distances $\mathbf{r} - \mathbf{r}'$ apart. It can thus be measured in rheological investigations in the limit of small applied deformation rates. This was used in the discussion of Fig. 3.

B. Generalised hydrodynamic equations

The stress correlations of the generalized Maxwell model, which were derived within the Mori-Zwanzig formalism, can be reformulated in terms of simple hydrodynamic equations. This approach provides an alternative derivation, clearly points to the generalization of the Maxwell model, and facilitates the interpretation considerably.

We start again from the Maxwell model, recapitulated in Sec. IV A, and formulated as a differential equation for the stress.[5] The linear response relation (46) and Maxwell's ansatz (29) are equivalent to the differential equation relating the shear stress and strain-rate

$$\tau\ \partial_t\sigma_{xy}^{\mathrm{M}}(t) + \sigma_{xy}^{\mathrm{M}}(t) = \eta^M\ \dot{\gamma}^{\mathrm{ext}}(t). \tag{47}$$

This formulation can also serve as the starting point for phenomenological generalizations in cases of strong flow rates $\boldsymbol{\kappa} = (\nabla\mathbf{v})^T$, where the partial time-derivative is replaced by the upper convected derivative $\partial_t\boldsymbol{\sigma} \to \partial_t\boldsymbol{\sigma} + \mathbf{v}\cdot\nabla\boldsymbol{\sigma} - \boldsymbol{\kappa}\cdot\boldsymbol{\sigma} - \boldsymbol{\sigma}\cdot\boldsymbol{\kappa}^T$,[46] while approaches within Zwanzig-Mori formalism more directly lead to integral formulations.[47]

The generalized Maxwell model naturally leads to partial differential equations which include elastic stress transport over large length scales. Given an externally imposed velocity field $\mathbf{v}^{\mathrm{ext}}(\mathbf{r}, t)$, we decompose the total velocity $\mathbf{v}(\mathbf{r}, t) = \mathbf{v}^d(\mathbf{r}, t) + \mathbf{v}_p(\mathbf{r}, t)$ into the streaming $\mathbf{v}^d(\mathbf{r}, t)$ and the peculiar velocity $\mathbf{v}_p(\mathbf{r}, t)$ and suppose that the streaming (or drift) flow is the externally imposed one, $\mathbf{v}^d(\mathbf{r}, t) = \mathbf{v}^{\mathrm{ext}}(\mathbf{r}, t)$. The peculiar velocity field is conserved and hence can be computed from the microscopic conservation law

$$mn\,\partial_t\mathbf{v}_p(\mathbf{r},t) = \nabla\cdot\left(\boldsymbol{\sigma}^{\mathrm{gM}}(\mathbf{r},t) - p(\mathbf{r},t)\mathbf{1}\right). \tag{48a}$$

We consider the incompressible limit (viz., $\nabla\cdot\mathbf{v}_p = 0$) so that the velocity is purely transverse and does actually not couple to the pressure p. The constitutive equation for the viscoelastic part $\boldsymbol{\sigma}^{\mathrm{gM}}(\mathbf{r}, t)$ is built on Maxwell's insight in glassy relaxation

$$(1 + \tau\partial_t)\boldsymbol{\sigma}^{\mathrm{gM}}(\mathbf{r},t) = 2\eta^{\mathrm{M}}\ \bar{\boldsymbol{\kappa}}(\mathbf{r},t), \tag{48b}$$

with the important difference that stresses are generated from the inhomogeneity of the flow field including both, local peculiar and externally imposed streaming motion

$$\bar{\boldsymbol{\kappa}}(\mathbf{r},t) = \frac{1}{2}\left(\nabla(\mathbf{v}_p + \mathbf{v}^{\mathrm{ext}}) + [\nabla(\mathbf{v}_p + \mathbf{v}^{\mathrm{ext}})]^T\right). \tag{48c}$$

Solving Eq. (48) for the flow induced stress reproduces the linear response relation in the incompressible case, viz., Eq. (38). The proof proceeds by the calculation of $\boldsymbol{\sigma}^{\mathrm{gM}}(\mathbf{q},\omega) = \int d^d r\,dt\,e^{i(\omega t+\mathbf{q}\cdot\mathbf{r})}\boldsymbol{\sigma}^{\mathrm{gM}}(\mathbf{r}, t)$ in Fourier space for an external perturbation $\bar{\boldsymbol{\kappa}}^{\mathrm{ext}}(\mathbf{q},\omega) = \frac{1}{2}\left((\nabla\mathbf{v}^{\mathrm{ext}}) + (\nabla\mathbf{v}^{\mathrm{ext}})^T\right) = \frac{\dot{\gamma}(\omega)}{2}(\hat{\mathbf{x}}\hat{\mathbf{y}}+\hat{\mathbf{y}}\hat{\mathbf{x}})$. See Appendix C for a sketch of the calculation which also provides additional details on the origin of stress anisotropy. The stress correlation function then is the Green's function $\mathrm{C}_{xyxy}^{(\sigma)\mathrm{gM}}(\mathbf{q}, s = -i\omega) = \sigma_{xy}^{\mathrm{gM}}(\mathbf{q},\omega)/\dot{\gamma}(\omega)$. In order to include sound damping in solids, an instantaneous dissipative contribution can be added: $\boldsymbol{\sigma}^{\mathrm{gM}}(\mathbf{r},t) \to \boldsymbol{\sigma}^{\mathrm{gM}}(\mathbf{r},t) + 2\Gamma\,\bar{\boldsymbol{\kappa}}(\mathbf{r},t)$. Then the model of Sec. IV E 2 is obtained.

While the classic Maxwell model would only have the external flow gradient on the right-hand side of Eq. (48b) [see Eq. (47)], the internal velocity gradient arises from the nonlocal (peculiar) velocity field which necessarily is induced by the imposed flow within a coarse-graining volume inherent to a hydrodynamic description.[20] This coupling ensures that the stationary stress is divergence free, which may fail in approaches in the class of classic Maxwell models in confinement.[48] Only for finite frequencies and relaxation times τ, does the transverse momentum diffusion allow for an (anisotropic) gradient expansion leading to Eq. (32). In solid states where $1/\tau = 0$, however, the gradient expansion breaks down and the nonlocal strain field induces the non-analytic small $\mathbf{q}$-expansion in Eq. (38b), which signals long-ranged elastic stress fields that are at the heart of elasto-plastic models.[39,49–51]

VI. CONCLUSIONS AND OUTLOOK

We presented the general Zwanzig-Mori decomposition of the stress tensor auto-correlations in fluids based solely on considerations of symmetry and conservation laws. Focusing on shear stress, we obtained its hydrodynamic spectrum in a fluid. We also showed that a single relaxation time approximation for one memory kernel, viz., the generalized shear modulus at zero wavevector, provides a consistent picture of shear stress correlations at long wavelengths in supercooled liquids and glasses and at the transition in between. The long-ranged correlations of elastic shear stresses in glass originate in the non-equilibrated correlations of the conserved transverse momentum fluctuations. The strong increase of the structural relaxation time upon supercooling causes the rapid growth of a correlation length ($\xi \propto \tau$) characterizing elastic behavior. The emerging viscoelasticity can be captured by a generalized Maxwell model where the memory kernel devoid of hydrodynamic correlations is approximated, while performing the same approximation on the shear-stress correlation function itself misses the far-field elastic correlations in solids. The stress correlations of the generalized Maxwell model exhibit a characteristic anisotropic pattern described by Eshelby for an elastic solid but so far not noticed for supercooled liquids. While we considered isothermal systems for simplicity and included the kinetic stress only in footnotes, we expect the assumption to neglect heat transport not to qualitatively affect the phenomena.

The general results within Zwanzig-Mori theory also apply to, e.g., polymeric liquids[52,53] but might require different approximations for the memory kernels than developed here for supercooled liquids. The phenomenology of the Zwanzig-Mori memory kernels as used in Sec. IV is compatible with the microscopic MCT of structural relaxation in supercooled liquids.[8]

For disordered solids at low temperatures, elasto-plastic models consider localized plastic events which interact via long-ranged elastic fields.[49–51] The present calculation identifies the coupling of momentum and stresses as a mechanism for the development of elastic correlations in the supercooled state. It appears worthwhile to search for further relations between fluid and solidlike mechanical descriptions across the glass transition. Also the connection to fluidity models should be explored.[54,55] In these models of driven or fluidized soft solids, the Maxwell model is generalized by assuming a nonlocal description of the inverse shear viscosity. The present calculations provide the basis for considering the fluid limit of these phenomenological models.

The assumption of a wave vector independent memory kernel was done on grounds of simplicity. It disagrees with some simulation studies,[56–58] in which a strong wavevector dependence of $G_q^{\perp}(t)$ is found, as modeled recently in Ref. 59. We find it important to reconcile these simulation results with the presented approach. As we find long-ranged stress-correlations, a finite-size analysis is required in simulations in order to detect them and to properly determine the viscosity. Newer simulations used finite size scaling to extract correlation lengths for diffusivities and viscosities.[60] Previous studies of the continuum limit of elastic systems indicated that the convergence may be slow.[61]

Our work opens many directions for further studies besides the ones already mentioned. The solid-like stress correlations we find are related to stress correlations in inherent structures of glass-forming simulations[14,18,62] and recover the far-field solutions of a replica approach to soft solids with quenched disorder.[63–65] The detailed relations including the dynamics are yet open.[66] The relation to thermodynamic formalisms which include internal variables could be considered. They give purely local stress strain relations.[67] The ensemble dependences should be studied in the Newtonian system including the conservation law of energy, where detailed investigations using computer simulations and thermodynamic formalism are available.[16,68,69] This could provide the link of the present definition of the shear modulus as a time-persistent correlation to linear response results in equilibrium solids which contain Born- and fluctuation terms.[70] Considering the linear-response calculation which considers a homogeneous driving, recent approaches[71] open the route to the rheology of inhomogeneous fluids in confinement.

Note added in proof. After completion of this work, we became aware of the analysis of the stress correlations in two dimensions using a different approach, viz. fluctuating hydrodynamics,[73] whose results can be recovered from ours by setting $q_z = 0$.

ACKNOWLEDGMENTS

We acknowledge support from the DFG through FOR 1394 projects P3 and P6.

APPENDIX A: ZWANZIG MORI DECOMPOSITION

In order to give a self-contained presentation, this appendix sketches the Zwanzig-Mori decomposition leading to Eq. (14). While the theory and its implications for hydrodynamic fluctuations are well described in textbooks,[1,3] we follow the monograph[8] where the application to non-hydrodynamic correlators is described; alternatively, Frobenius' formula for matrix inversion can be used directly.[29]

The aim is to express the general correlation function $\langle \delta A(t)^* \delta B\rangle$ in terms of correlation functions of slow conserved variables and kernels which contain the memory of the system arising from non-conserved degrees of freedom. The central object is the resolvent $R(s) = (s+i\mathcal{L})^{-1}$ introduced in Sec. III B, which gives the dynamics in Laplace-space. The projectors $\mathcal{P}$ and $\mathcal{Q}$ introduced in Eqs. (12) and (13) project onto the conserved and non-conserved variables. The dynamics devoid of conserved variables is contained in the reduced resolvent $R'(s) = \mathcal{Q}(s + i\mathcal{Q}\mathcal{L}\mathcal{Q})^{-1}\mathcal{Q}$.

Using $\mathcal{P}+\mathcal{Q} = 1$, the complete resolvent can be split into four terms,

$$R(s) = \mathcal{P}R(s)\mathcal{P} + \mathcal{Q}R(s)\mathcal{P} + \mathcal{P}R(s)\mathcal{Q} + \mathcal{Q}R(s)\mathcal{Q}, \tag{A1}$$

which can individually be further simplified using the projectors and the identity $(s + i\mathcal{L})R(s) = 1$. It can be rewritten as

$$(s + i\mathcal{L})\mathcal{P}R(s) + (s + i\mathcal{L})\mathcal{Q}R(s) = 1. \tag{A2}$$

Multiplying this relation by $R'(s)$ from left and $\mathcal{P}$ from right and using $\mathcal{Q}\mathcal{P} = 0$ gives the second term on the right-hand side of Eq. (A1),

$$\mathcal{Q}R(s)\mathcal{P} = -iR'(s)\mathcal{L}\mathcal{P}R(s)\mathcal{P}. \tag{A3}$$

The result $\mathcal{P}R(s)\mathcal{Q} = -i\mathcal{P}R(s)\mathcal{P}\mathcal{L}R'(s)$ can be obtained similarly starting from $R(s)(s + i\mathcal{L}) = 1$. Multiplying Eq. (A2) by $R'(s)$ from left and $\mathcal{Q}$ from right gives the last term on the right-hand side of Eq. (A1),

$$\mathcal{Q}R(s)\mathcal{Q} = R'(s) - iR'(s)\mathcal{L}\mathcal{P}R(s)\mathcal{Q}. \tag{A4}$$

Collecting all terms just obtained, Eq. (A1) can be rewritten

$$R(s) = R'(s) + [1 - i\mathcal{L}R'(s)]\mathcal{P}R(s)\mathcal{P}[1 - i\mathcal{L}R'(s)], \tag{A5}$$

which is used in Eq. (14) of the main text. Finally, in order to find the dynamics of the conserved variables, one multiplies Eq. (A2) by $\mathcal{P}$ from left and right

$$\begin{aligned}(s + i\mathcal{P}\mathcal{L}\mathcal{P})\mathcal{P}R(s)\mathcal{P} &+ i\mathcal{P}\mathcal{L}\mathcal{Q}R(s)\mathcal{P}\\ &= \mathcal{P} = (s + i\mathcal{P}\mathcal{L}\mathcal{P})\mathcal{P}R(s)\mathcal{P} + \mathcal{P}\mathcal{L}R'(s)\mathcal{L}\mathcal{P}R(s)\mathcal{P},\end{aligned} \tag{A6}$$

where in the second line Eq. (A3) was used. This relation leads to the well known Zwanzig-Mori equations of motion for the correlation functions of the conserved variables.[1,3,8] They are summarized in Eq. (25).

APPENDIX B: FORCE AUTO-CORRELATION FUNCTIONS

The results for the force auto-correlation functions given in Eq. (26) can easily be derived from momentum conservation in Zwanzig-Mori formalism. One starts from the expressions for the velocity auto-correlations in Eq. (25), concisely written as

$$\mathrm{K}_q^{(i)}(s) = \frac{k_B T n}{mns + q^2 \mathrm{G}_q^{(i)}(s)}, \tag{B1}$$

where $\mathrm{K}_q^{(i)}(s)$ stands for either longitudinal, $\mathrm{K}_q^{\parallel}(s)$, or transverse, $\mathrm{K}_q^{\perp}(s)$, velocities and $\mathrm{G}_q^{(i)}(s)$ stands for $\tilde{\mathrm{G}}_q^{\parallel}(s)$ or $\mathrm{G}_q^{\perp}(s)$. The momentum conservation law (1) for either longitudinal or transverse fluctuations, $i \in \{\parallel, \perp\}$, holds

$$\partial_t \mathrm{v}^{(i)}(q, t) = \mathrm{F}^{(i)}(q, t)/m \tag{B2}$$

and leads to a differential relation between the velocity and force correlation functions [the latter are defined in Eq. (22)]

$$-\partial_t^2 \mathrm{K}_q^{(i)}(t) = \langle \dot{\mathrm{v}}_q^{(i)*}(t)\dot{\mathrm{v}}_q^{(i)}\rangle = \frac{k_B T q^2}{m^2 n}\, \mathrm{Z}_q^{(i)}(t). \tag{B3}$$

Laplace transformation and the initial conditions $\mathrm{K}_q^{(i)}(t \to 0) = (k_B T)/m + \mathcal{O}(t^2)$ give Eq. (26) after few manipulations. Because of relation (B3) and the boundedness of correlation functions, viz., $|\mathrm{K}_q^{(i)}(t)| \le \mathrm{K}_q^{(i)}(0) = \frac{k_B T}{m}$, the force auto-correlation functions need to vanish for $t \to \infty$.

APPENDIX C: SOLUTION OF THE GENERALIZED HYDRODYNAMIC EQUATIONS

Solving Eq. (48) verifies the equivalence to the generalized Maxwell model and provides insights into the origins of the anisotropy of the stress correlations. After Fourier-transformation, and assuming an external shear deformation, $\bar{\boldsymbol{\kappa}}^{\rm ext}(\mathbf{q}, \omega) = \frac{\dot\gamma(\mathbf{q},\omega)}{2}(\hat{\mathbf{x}}\hat{\mathbf{y}} + \hat{\mathbf{y}}\hat{\mathbf{x}})$, a frequency dependent viscosity $\eta(\omega) = \eta^M/(1 - i\omega\tau) + \Gamma$ can be introduced. Inserting Eq. (48b) into Eq. (48a) and using the incompressibility of the flow gives

$$\left(-imn\omega + \eta q^2\right)\mathbf{v}_p = -i\eta\dot\gamma\left(q_x\hat{\mathbf{y}} + q_y\hat{\mathbf{x}}\right) + i\mathbf{q}p. \tag{C1}$$

This is the diffusion equation for transverse momenta. Again using the incompressibility condition, $\mathbf{q}\cdot\mathbf{v}_p = 0$, gives the solution for the pressure

$$p(\mathbf{q}, \omega) = 2\frac{q_x q_y}{q^2}\eta(\omega)\dot\gamma(\mathbf{q}, \omega). \tag{C2}$$

Inserting Eq. (C1) into Eq. (48b) gives the result for the shear stress

$$\begin{aligned}(-imn\omega + \eta(\omega)q^2)\,&\sigma_{xy}^{\rm gM}(\mathbf{q}, \omega)\\ &= (-imn\omega + \eta(\omega)q_z^2)\,\eta(\omega)\dot\gamma(\mathbf{q}, \omega) + 2q_x q_y \eta(\omega)p(\mathbf{q}, \omega)\\ &= (-imn\omega + \eta(\omega)\,(q_z^2 + 4\frac{q_x^2 q_y^2}{q^2}))\ \eta(\omega)\dot\gamma(\mathbf{q}, \omega),\end{aligned} \tag{C3}$$

where Eq. (C2) was used in the second line. As claimed in Sec. V B, the stress correlation function of the generalized Maxwell model introduced in Sec. IV E follows as Green's function $\mathrm{C}_{xyxy}^{(\sigma)\rm gM}(\mathbf{q}, s = -i\omega) = \sigma_{xy}^{\rm gM}(\mathbf{q}, \omega)/\dot\gamma(\mathbf{q}, \omega)$. If desired, the result from Eq. (C3) can be transformed into real space using the solution of the transient Oseen equation.[72]

Equation (C3) shows that the shear stress obeys the isotropic diffusion equation inherited from the transverse momentum. The source term on the right-hand side contains two terms. The isotropic one, $\propto\omega\dot\gamma$, vanishes in the stationary case. It is the origin of the suppression of $\sigma_{xy}^{\rm gM}(\mathbf{q}, \omega)$ at small frequencies, visible at finite $\mathbf{q}$ along most directions. The incompressibility condition of the flow, however, enters a peculiar angular dependence into the inhomogeneity. Rotating the coordinate systems by 45°, viz., introducing $q_\pm = (q_x \pm q_y)/\sqrt{2}$ in the plane $q_z = 0$, the anisotropic term becomes $4q_x^2 q_y^2 = (q_+^2 - q_-^2)^2$. For directions along the diagonals, $q_+ = 0$ or $q_- = 0$, therefore one obtains

$\sigma_{xy}^{\mathrm{gM}}(\mathbf{q}_\pm,\omega) = \eta(\omega)\dot{\gamma}(\mathbf{q},\omega)$. This is the classical Maxwell relation. Along the diagonals, stress only relaxes via structural processes, which freeze out at the glass transition, $\eta(\tau\to\infty) \to iG_\infty/\omega + \Gamma \to iG_\infty/\omega$ for $\omega\tau_0 \ll 1$ and thus $\sigma_{xy}^{\mathrm{gM}}(\mathbf{q}_\pm,\omega\to 0) = G_\infty\gamma(\omega)$ with $\gamma(\omega)$ being the applied shear strain.

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[31]The correlation functions $\mathbf{Z}(\mathbf{q}, t)$ agree with force correlations only when kinetic contributions are neglected. The final results in Eq. (26) derived in Appendix B are valid including kinetic terms in $\mathbf{G}(\mathbf{q}, t)$.
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