Long-range stress correlations in viscoelastic and glass-forming fluids

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Abstract

A simple and rigorous approach to obtain stress correlations in viscoelastic liquids (including supercooled liquid and equilibrium amorphous systems) is proposed. The long-range dynamical correlations of local shear stress are calculated and analyzed in 2-dimensional space. It is established how the long-range character of the stress correlations gradually emerges as the relevant dynamical correlation length ℓ grows in time. The correlation range ℓ is defined by momentum propagation due to acoustic waves and vorticity diffusion which are the basic mechanisms for transmission of shear stress perturbations. We obtain the general expression defining the time- and distance- dependent stress correlation tensor in terms of material functions (generalized relaxation moduli). The effect of liquid compressibility is quantitatively analyzed; it is shown to be important at large distances and/or short times. The revealed long-range stress correlation effect is shown to be dynamical in nature and disconnected with static structural correlations in liquids (correlation length ξ_s). Our approach is based on the assumption that ξ_s is small enough as reflected in weak wave-number dependencies of the generalized relaxation moduli. We provide a simple physical picture connecting the elucidated long-range fluctuation effect with anisotropic correlations of the (transient) inherent stress field, and discuss its implications.

1. Introduction

Liquids are characterized by an essentially irregular (amorphous) structure with *short-range* spatial correlations. Yet, liquid systems often show very long-time relaxation processes. The most important examples of such complex fluids are macromolecular (polymer) melts or solutions [1,2] and supercooled liquids or colloidal systems near the glass transition [3–5]. The flow dynamics of such systems is a subject of rheology. One important rheological characteristic is the response function $G(t)$ defining the relaxation of shear stress $\sigma(t)$ after a small step of the shear strain, $\dot{\gamma}(t) = \gamma_0 \delta(t)$, $\gamma_0 \ll 1$ (here $\dot{\gamma}$ is the shear rate [2]):

$$\sigma(t) = G(t)\gamma_0$$

The memory function $G(t)$, also known as the shear relaxation modulus, has been determined in rheometric experiments for many complex fluids. It decreases with time from the initial value $G(0)$ (instantaneous affine shear modulus) to G_∞, the long-time plateau shear modulus at the 'macroscopic' laboratory time-scale t_{lab}, $G_\infty \equiv G(t_{lab})$. Note that typically $G_\infty = 0$ for liquids well above the glass transition temperature T_g, while $G_\infty > 0$ for elastic solids (like crosslinked polymer systems or crystals) and for supercooled liquids below T_g. [1]. The same function $G(t)$ also characterizes the time-dependent correlations of the stress fluctuations, $\tilde{\sigma}(t) = \sigma(t) - \bar{\sigma}$, where $\bar{\sigma}$ is the mean stress (averaged over the time-scale t_{lab}): [6]

$$C(t) \equiv \langle \tilde{\sigma}(0)\tilde{\sigma}(t) \rangle = \frac{T}{V} G(t) \tag{1}$$

where T is the temperature in energy units (absolute temperature times the Boltzmann constant) and V is the volume of the system. The fluctuation relation (1) is valid for systems that are allowed to deform as a whole faster than t_{lab} ($\bar{\sigma}$ becomes negligible as a result, $\bar{\sigma} \to 0$), while in the case of fixed boundaries (the standard canonical ensemble) it must be replaced by [7–11]

$$\langle \tilde{\sigma}(0)\tilde{\sigma}(t) \rangle \simeq \frac{T}{V} [G(t) - G_\infty] \tag{2}$$

Eqs. (1), (2) involve the shear stress averaged over the whole volume. [2]

Let us turn to the space-resolved correlations of the local stress $\sigma(\underline{r}, t)$, $C(\underline{r}, t) = \langle \tilde{\sigma}(\underline{r}', t')\tilde{\sigma}(\underline{r}' + \underline{r}, t' + t) \rangle$. This generalized stress correlation function is also related to rheological characteristics of the fluid (see the next section). The distance-dependent stress correlations have been considered in several recent simulation studies [12–14] as well as theoretically [15]. These studies suggest that the stress

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[1] Note also that G_∞ is generally not equal to the equilibrium shear modulus G_e introduced in eq. (18)

[2] Note that G_∞ is nearly equal to the modulus μ considered in refs. [10,11]. It is also worth noting that, by contrast, the glassy modulus μ can significantly exceed G_e since a deformed glass (with prescribed boundary conditions) would not relax to the equlibrium state on the laboratory time-scale.

correlation function is both non-local and anisotropic. Let us briefly comment on the theoretical results obtained in Ref. [15]:

(i) In the short-time regime, $t \ll \tau$ (where τ is the characteristic time associated with relaxation of $G(t)$ which was assumed to be exponential) the theory [15] predicts *infinitely long-range* stress correlations (at any short time $t > \tau_m$, where τ_m is the microscopic time limit of the theory): $C(\underline{r},t) \propto t^2/r^{d+2}$ for $r > c_T t$, where c_T is the transverse shear velocity (see eq. (44) below and the text around it). This result seems surprising in view of the implicitly made assumption that all structural correlations in the liquid are short-range (including the stress correlations at $t = 0$). Formally, this assumption is adopted in ref. [15] by disregarding the wave-vector dependence of the memory function $G(q,t)$.

(ii) Expanding on the previous point, we note that the structural locality (short-range correlations at $t = 0$) and a finite speed of any physical signal mean that stress correlations must be of *finite* range at any t.

(iii) We believe that the issues mentioned above cannot be resolved within the incompressible liquid model adopted in ref. [15], and therefore chose to analyze the effects of finite compressibility in the present paper.

Therefore, we consider here the stress correlations in *compressible* viscoelastic fluids. Instead of the Zwanzig-Mori projection formalism [6] used in ref. [15] we develop a physical approach based solely on the concepts of response functions and random stress (rather than on the mode-coupling theory, see section 3). It also allows to specify the basic assumptions in a clear way and physically demonstrate their validity. The theory is based on the general relationships between the stress correlation function and the memory response functions coming from the fluctuation-dissipation theorem (FDT) [26] considered in the next section. The main results concerning the space- and time-resolved stress correlation function and its long-range behavior are presented in the 4th section followed by a summary and discussion of the new findings in the last section.

2. The FDT relations and material functions

The stress correlation function is defined in the general case as

$$C_{\alpha\beta\alpha'\beta'}(\underline{r},t) = \langle \tilde{\sigma}_{\alpha\beta}(\underline{r}+\underline{r}',t+t')\tilde{\sigma}_{\alpha'\beta'}(\underline{r}',t')\rangle \tag{3}$$

where $\langle ... \rangle$ means averaging over an equilibrium ensemble, $\tilde{\sigma}_{\alpha\beta}(\underline{r},t) = \sigma_{\alpha\beta}(\underline{r},t) - \langle \sigma_{\alpha\beta}\rangle$ is the tensor of local stress increments, α, β, ... are Cartesian components, $\langle \sigma_{\alpha\beta}\rangle = \langle \sigma_{\alpha\beta}(\underline{r},t)\rangle$ is the stress tensor averaged over the equilibrium ensemble (the system is assumed to be translation-invariant). The function C has obvious symmetries: it is invariant with respect to exchanges $\alpha\beta \to \beta\alpha$, $\alpha'\beta' \to \beta'\alpha'$, and

$$C_{\alpha\beta\alpha'\beta'}(\underline{r},t) = C_{\alpha'\beta'\alpha\beta}(-\underline{r},-t)$$

The time reversibility and uniformity demand that

$$C_{\alpha\beta\alpha'\beta'}(\underline{r},t) = C_{\alpha\beta\alpha'\beta'}(\underline{r},-t) = C_{\alpha'\beta'\alpha\beta}(-\underline{r},t) \tag{4}$$

In addition, all even-dimensional systems and all achiral systems obey

$$C_{\alpha\beta\alpha'\beta'}(\underline{r},t) = C_{\alpha\beta\alpha'\beta'}(-\underline{r},t) \tag{5}$$

hence

$$C_{\alpha\beta\alpha'\beta'}(\underline{r},t) = C_{\alpha'\beta'\alpha\beta}(\underline{r},t)$$

The general fluctuation-dissipation theorem (FDT) relates the function C with the linear response of stress to a small instant deformation of the system at $t = -0$ like

$$\underline{r} \to \underline{r} + \underline{u}(\underline{r})$$

where $\underline{u}(\underline{r})$ is an infinitesimal particle-displacement field (the particle momenta $\underline{p}$ are also changed to render the whole transformation canonical in the Hamiltonian phase-space of the system: $p_\alpha \to p_\alpha - u_{\beta,\alpha}p_\beta$, where $u_{\beta,\alpha} = \frac{\partial u_\beta}{\partial r_\alpha}$). Then, according to the FDT the mean stress increment $\langle\widetilde{\sigma}_{\alpha\beta}\rangle$ induced by the infinitesimal strain field

$$\gamma_{\alpha\beta}(\underline{r}) = u_{\alpha,\beta} + u_{\beta,\alpha}$$

can be written as [6]:

$$\langle\sigma_{\alpha\beta}(\underline{r},t)\rangle = \frac{1}{2T}\int \mathrm{d}^d r' C_{\alpha\beta\alpha'\beta'}(\underline{r}-\underline{r}',t)\gamma_{\alpha'\beta'}(\underline{r}') \tag{6}$$

where d is the space dimension ('tilde' over σ is omitted here and below, and summation over repeated indices is implied). Note that $\langle\sigma_{\alpha\beta}\rangle$ means an out-of-equilibrium average stress increment due to an applied field here and below. Summation over α' and β' is essential in eq. (6); such summation was omitted in the analogous equation 9 of ref. [15] which therefore does not describe the general case. [3] Doing Fourier transformation of the last equation we get

$$\left\langle\sigma_{\alpha\beta}(\underline{q},t)\right\rangle = \frac{1}{2T}C_{\alpha\beta\alpha'\beta'}(\underline{q},t)\gamma_{\alpha'\beta'}(\underline{q}) \tag{7}$$

where $C_{\alpha\beta\alpha'\beta'}(\underline{q},t) = \frac{1}{V}\left\langle\sigma_{\alpha\beta}(\underline{q},t)\,\sigma^*_{\alpha'\beta'}(\underline{q},0)\right\rangle$ and V is the total volume of the system. Note that Fourier transforms of C and γ are indicated by the wave-vector argument $\underline{q}$, while the function names are not changed for notation simplicity. Note also that eqs. (4), (5) imply that $C_{\alpha\beta\alpha'\beta'}(\underline{q},t)$ is real and does not depend on the sign of t:

$$C_{\alpha\beta\alpha'\beta'}(\underline{q},t) = C_{\alpha\beta\alpha'\beta'}(\underline{q},-t) = C^*_{\alpha\beta\alpha'\beta'}(\underline{q},t)$$

Let us try to obtain the stress correlation function C using the FDT relation (7) and based on the known relaxation moduli. To this end we first find the stress response to the deformation of the system using an independent approach outlined in ref. [27]. In the linear response approximation the mean stress must be a linear function of the flow velocity field. For a Newtonian fluid the local stress is just proportional to the local rate-of-strain $\dot{\gamma}_{\alpha\beta}(\underline{r}) = v_{\alpha,\beta} + v_{\beta,\alpha}$ (here $v_\alpha = v_\alpha(\underline{r},t)$ is the

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[3] Note that κ^{ext}_{xy} in eq. 9 of ref. 15 is the symmetric part of the external velocity gradient, which is generally different from the total rate-of-strain including also a part generated by internal forces (see the discussion in the 4th paragraph of section 5.

flow velocity), while in the general case of a complex fluid with memory effects the relation is

$$\left\langle \sigma_{\alpha\beta}(\underline{q},t)\right\rangle = \frac{1}{2}\int_{-\infty}^{t} E_{\alpha\beta\alpha'\beta'}(\underline{q},t-t')\dot{\gamma}_{\alpha'\beta'}(\underline{q},t')\mathrm{d}t' \tag{8}$$

Note that E is symmetric with respect to α', β' permutations. [4] Using the Laplace transformation the latter equation can be written as (in what follows we assume no flow at $t<0$):

$$\left\langle \sigma_{\alpha\beta}(\underline{q},s)\right\rangle = E_{\alpha\beta\alpha'\beta'}(\underline{q},s)\dot{\gamma}_{\alpha'\beta'}(\underline{q},s)/2 \tag{9}$$

where $\left\langle \sigma_{\alpha\beta}(\underline{q},s)\right\rangle = \int_0^\infty \left\langle \sigma_{\alpha\beta}(\underline{q},t)\right\rangle e^{-st}\mathrm{d}t$, etc. (A special case of the above relation was employed before [27].) Taking into account the space isotropy and that

$$\dot{\gamma}_{\alpha\beta}(\underline{q},s) = \mathrm{i}\left(q_\alpha v_\beta(\underline{q},s) + q_\beta v_\alpha(\underline{q},s)\right) \tag{10}$$

the stress response to the flow can be written as

$$\left\langle \sigma_{\alpha\beta}(\underline{q},s)\right\rangle = G(q,s)\dot{\gamma}_{\alpha\beta}(\underline{q},s) + \dot{\epsilon}\left[M(q,s)\delta_{\alpha\beta} + (K-2G-M)\,\frac{q_\alpha q_\beta}{q^2}\right] \tag{11}$$

where $\dot{\epsilon} = \dot{\epsilon}(\underline{q},s) = \frac{1}{2}\mathrm{Tr}\dot{\gamma}$, and $G(q,s)$, $K=K(q,s)$, $M=M(q,s)$ are Laplace transforms of the following 3 material functions (the generalized time-dependent relaxation moduli): the shear modulus $G(q,t)$, the longitudinal modulus $K(q,t)$ (not to be confused with the bulk modulus) and the mixed modulus $M(q,t)$ which all depend on the magnitude $\left|\underline{q}\right|$ of the wave-vector (but do not depend on its direction). The physical meaning of these material functions is clear: $G(q,t)$ defines the shear stress response to a shear strain, while $K(q,t)$ and $M(q,t)$ define, respectively, the longitudinal and transverse stresses generated by a longitudinal strain (by transverse stress we mean the normal stress in a direction perpendicular to $\underline{q}$). More formally, these definitions can be presented using the natural coordinate frame with the first axis ($\underline{e}_1$) parallel to $\underline{q}$. The response to an instant deformation $\underline{v}(t) = \underline{u}\delta(t)$ with $\underline{u} = (u_1, u_2, 0)$ then reads (here $\delta(t)$ is the Dirac's delta)

$$\left\langle \sigma_{11}(\underline{q},t)\right\rangle = K(q,t)\epsilon, \quad \left\langle \sigma_{22}(\underline{q},t)\right\rangle = M(q,t)\epsilon, \quad \left\langle \sigma_{12}(\underline{q},t)\right\rangle = G(q,t)\gamma \tag{12}$$

where $\epsilon = \mathrm{i}qu_1$, $\gamma = \mathrm{i}qu_2$ (all other components of the induced stress, except σ_{11}, σ_{22} and $\sigma_{21}=\sigma_{12}$, are equal to zero). [5] Equations (12) are valid if an external time-dependent force is applied to the fluid in order to keep it still (no flow, $\underline{v}=0$) at $t>0$. Otherwise, if no external force is applied, the fluid motion is defined by the momentum equation

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[4] Note also that eq. (8) can be used to predict the stress field if the flow field is known (is *imposed* or *prescribed*).

[5] The moduli G, K, M are related to the components of the tensor $E_{\alpha\beta\alpha'\beta'}$: $G = E_{1212}$, $K = E_{1111}$, and $M = E_{2211}$.

$$\frac{\partial}{\partial t} J_\alpha = \sigma_{\alpha\beta,\beta}, \quad t > 0 \tag{13}$$

where $J_\alpha = \rho v_\alpha$ is the momentum flux and ρ is the fluid mass per unit volume. Within the linear approximation we can treat ρ as a constant (thermodynamic density of the fluid) and rewrite the above equation in terms of the ensemble-averaged quantities:

$$\rho \frac{\partial}{\partial t} \langle v_\alpha \rangle = \mathrm{i} q_\beta \langle \sigma_{\alpha\beta} \rangle , \quad t > 0 \tag{14}$$

Solving eqs. (14), (11) for the stress $\left\langle \sigma_{\alpha\beta}(\underline{q}, t) \right\rangle$ and comparing the result with eq. (7) we get (using again a natural coordinate frame related to a given $\underline{q} \neq 0$)

$$C_T(q,s) \equiv C_{1212}(\underline{q},s) = T \frac{\rho s G(q,s)}{\rho s + q^2 G(q,s)} \tag{15}$$

$$C_{||}(q,s) \equiv C_{1111}(\underline{q},s) = T \frac{\rho s K(q,s)}{\rho s + q^2 K(q,s)} \tag{16}$$

$$C_{\perp}(q,s) \equiv C_{2211}(\underline{q},s) = T \frac{\rho s M(q,s)}{\rho s + q^2 K(q,s)} \tag{17}$$

where

$$C_{\alpha\beta\alpha'\beta'}(\underline{q},s) = \int_0^\infty C_{\alpha\beta\alpha'\beta'}(\underline{q},t) e^{-st} \mathrm{d}t$$

Eq. (15) is well-established [31,73]; its FDT-based derivation is given in ref. [27]. The second relation (16) is mentioned in ref. [28].

It is worth stressing again that the above results are valid for $q \neq 0$. The situation for $q = 0$ is more subtle [7,28]: in this case the fluctuations of the mean stress averaged over the whole system are involved; their dynamics and statistics depend on the boundary conditions. If the total volume and shape of the system are not allowed to fluctuate (which is often the most convenient option for computer simulations), then $C_{||,T}(0,t)$ may not coincide with the $\lim_{q\to 0} C_{||,T}(q,t)$ [28]. As a result, the FDT relations for $q = 0$ in the general case become [7–10,28]

$$C_{||}(0,t) = T\left[K(t) - K_e\right], \quad C_T(0,t) = T\left[G(t) - G_e\right] \tag{18}$$

where $G(t) = G(q=0,t)$, $K(t) = K(q=0,t)$, and K_e, G_e are the equilibrium longitudinal and shear moduli (for infinitesimal strain), respectively ($G_e > 0$ for cross-linked polymer systems or crystalline solids, but $G_e = 0$ for uncrosslinked systems like liquids and amorphous systems considered herein) [6], [7]. As for the

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[6] We do not consider here nonergodic amorphous systems below the putative ideal glass transition temperature T_K which may exhibit $G_e > 0$ [64–66]. In our view it is highly likely that the finite rigidity in this regime is due to (hidden) long-range static structural correlations.

[7] To avoid fictitious problems (arriving at $G_e = 0$ even for crystalline solids [67]) we define the moduli taking first the limit of infinitesimal strain, $\gamma \to 0$, and then the thermodynamic limit, the number of particles $N \to \infty$.

3rd correlation function $C_\perp(0,t)$, it is not independent for $q = 0$: $C_\perp(0,t) = C_\parallel(0,t) - 2C_T(0,t)$ (since $M(0,t) = K(0,t) - 2G(0,t)$). [8]

The FDT-based relations (15) - (17) provide 3 independent components of the stress correlation tensor $C_{\alpha\beta\alpha'\beta'}$. However, the whole tensor function C remains yet unknown as it generally involves 4 or 5 independent components in total (including C_{2222} for 2-dimensional systems, and, in addition, C_{2233} in 3 dimensions). These extra components can not be derived from any FDT relation, nor can they be directly expressed in terms of a material function like those considered above. (Note that the number of independent material functions (= 3) is defined mathematically by the most general linear relationship (as given in eq. (11)) between the tensor σ and the vector $\underline{v}$, compatible with the system isotropy and involving the second vector $\underline{q}$.) Fortunately, however, all the stress-correlation components can be calculated in the hydrodynamic regime as demonstrated in the next section.

3. The hydrodynamic fluctuations

In what follows we focus on the dynamics at long length-scales and therefore small wave-vectors q, $qa_m \ll 1$, where a_m is the molecular size. The details of the short-scale structure (at length-scale $\sim a_m$) and the short-time processes (like molecular collisions with time-scale τ_m) are disregarded (wiped-out) within the adopted hydrodynamic approach. Of course, thermal fluctuations drive the fluid motion also at large scales. Such slow motions are explicitly taken into account in the model considered below.

According to this approach the local stress field can be generally represented as a sum of 2 contributions: [9]

$$\sigma_{\alpha\beta}(\underline{q},t) = \sigma^{\mathcal{D}}_{\alpha\beta}(\underline{q},t) + \sigma^{\mathrm{n}}_{\alpha\beta}(\underline{q},t) \tag{19}$$

where the first term $\sigma^{\mathcal{D}}$ is the 'regular' flow-generated stress defined by the current strain and strain rate or, more generally, by the whole flow (deformation) history; $\sigma^{\mathcal{D}}$ depends on the flow in the linear-response fashion reflected in eq. (8). The linear response is valid since the flow is weak at long length-scales, $qa_m \ll 1$. The second term σ^{n} is the random stress due to structural (packing) irregularities and thermal fluctuations of particle velocities (thermal noise) which is independent of the macroscopic flow for the same reason: weak flow does not affect much the local fluid structure defining the noise σ^{n}.

To obtain the *noise* correlation function

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[8] It is important that the stress correlation function $C_{\alpha\beta\alpha'\beta'}(\underline{q},t)$ is discontinuous at $q = 0$. For example, $\lim_{q\to 0} C_\parallel(q,t) \neq C_\parallel(q=0,t)$ in the general case. [28] Moreover, $C_{\alpha\beta\alpha'\beta'}(q=0,t)$ is also known to be notoriously ensemble-dependent. [69–71,28] By contrast, the correlation function $C_{\alpha\beta\alpha'\beta'}(\underline{q},t)$ for a *finite* q is independent of the statistical ensemble in the thermodynamic limit, $N \to \infty$. [28] That is why in what follows we focus on the $q \neq 0$ regime for the stress correlation function.

[9] The treatment given here is close in spirit to the classical fluctuation theories [29,30].

$$C^{\rm n}_{\alpha\beta\alpha'\beta'}\left(\underline{q},t\right)=\frac{1}{V}\left\langle\sigma^{\rm n}_{\alpha\beta}(\underline{q},t+t')\sigma^{\rm n*}_{\alpha'\beta'}(\underline{q},t')\right\rangle \tag{20}$$

we recall that $\boldsymbol{\sigma}^{\rm n}$ reflects *thermal fluctuations* of the fluid structure. Such structural correlations are always short-range in a fluid, their range ξ_s is comparable to a_m. [10] Therefore, a q-dependence of $C^{\rm n}$ must be weak for $q\xi_s \ll 1$. Hence, $C^{\rm n}$ can be approximated by setting $q=0$. Conveniently, for $q=0$ the fixed boundary conditions also eliminate the flow (i.e., $\boldsymbol{\sigma}^{\mathcal{D}}=0$), so the random stress $\boldsymbol{\sigma}^{\rm n}$ coincides with the total stress in this case. The correlation function $C^{\rm n}$ can be then obtained using eq. (18):

$$\frac{1}{T}C^{\rm n}_{\alpha\beta\alpha'\beta'}\left(\underline{q},t\right)\simeq\frac{1}{T}C^{\rm n}_{\alpha\beta\alpha'\beta'}\left(0,t\right)=$$

$$=\left[G(t)-G_e\right]\left(\delta_{\alpha\alpha'}\delta_{\beta\beta'}+\delta_{\alpha\beta'}\delta_{\alpha'\beta}\right)+\left[M(t)-M_e\right]\delta_{\alpha\beta}\delta_{\alpha'\beta'} \tag{21}$$

where $M(t)=K(t)-2G(t)$, $M_e=K_e-2G_e$, and it is taken into account that at $q=0$ the stress correlation tensor must be isotropic. For liquids therefore

$$C^{\rm n}_{\alpha\beta\alpha'\beta'}\left(\underline{q},t\right)\to 0 \ \text{ at } \ t\to\infty$$

as it should be since the mean random stress must vanish due to its fluctuation nature in the liquid state ($\lim_{t_{max}\to\infty}\frac{1}{t_{max}}\int_0^{t_{max}}\boldsymbol{\sigma}^{\rm n}(t)\mathrm{d}t=0$).

The correlation function C of the total stress can then be obtained by solving the general momentum equation (13) with the total stress defined in eq. (19), where $\boldsymbol{\sigma}^{\mathcal{D}}$ can be calculated using eqs. (8), (11), while the $\boldsymbol{\sigma}^{\rm n}$ contribution can be considered as an independent driving force whose statistics are defined in eqs. (20), (21). [11]

It is important that the relaxation moduli involved in eq. (11) are analytical functions of q since the deformation-generated stress is defined by the *local* structure of the system (local correlations of neighboring interacting particles). Hence the q-dependence of the material functions must be weak for length-scales exceeding the structural correlation length ξ_s comparable with the molecular size (and interaction

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[10] Fragile glass-forming liquids are characterized by a super-Arrhenius increase of the relaxation time on cooling toward $T_{\rm g}$. An actively debated question is whether the associated increase of the activation energy is caused by the growth of an underlying static glass correlation length $\xi_{\rm s}$ [40,16–18]. Since static pair correlations (as measured by the static structure factor) do not change much with decreasing temperature, a length scale extracted from these correlations is unlikely to be a suitable candidate for $\xi_{\rm s}$. Recent suggestions involve point-to-set correlations which increase more strongly, but typically do not exceed a few $a_{\rm m}$ over the range of temperatures accessible to computer simulations [17]. However, even for temperatures approaching the laboratory $T_{\rm g}$, $\xi_{\rm s}$ would not need to exceed more than $\sim 10a_{\rm m}$ in order to explain the super-Arrhenius behavior of the relaxation time [17,18]. So, the growth of $\xi_{\rm s}$ in glass-forming liquids is expected to be rather limited [40,49,46,48].

[11] The independence of $\boldsymbol{\sigma}^{\rm n}$ permits to formally consider it as a weak 'external force' applied to the fluid particles and generating their slow motion.

range), a_m. [12] It is also noteworthy that in some systems (like, say, polymeric fluids) the molecules are large, so there is a significant range between the atomic size and a_m where the relaxation modulus $G(q,t)$ and the generalized viscosity $\eta(q)$ (cf. eq. (50)) may significantly depend on q; this dependence has been calculated in ref. [27]. In what follows, however, we consider a more universal regime of longer length-scales, $1/q \gg a_m, \xi_s$, where the material functions can be approximated by the $q = 0$ limit, $G(t) = G(0,t)$, etc. (since the relaxation moduli are continuous at $q = 0$)

$$G(q,t) \simeq G(t), \quad K(q,t) \simeq K(t), \quad M(q,t) \simeq K(t) - 2G(t) \tag{22}$$

The last equation follows merely from the fact that at $q = 0$ the stress (in eq. (11)) must *not* depend on the orientation of $\underline{q}$, so $K - 2G - M = 0$. Thus, only two functions, $G(t)$ and $K(t)$, are involved in the regime of interest. These functions can be directly measured in rheological and acoustic experiments [2,29,31–33].

In what follows we shall restrict the consideration to achiral 2-dimensional (2D) systems which are invariant with respect to reflection of the second axis $\underline{e}_2$ (the unit vector $\underline{e}_2$ is perpendicular to the wave-vector $\underline{q}$). Therefore, for example, C_{1112} must vanish, and the only non-vanishing independent components (apart from the trivial permutations of subscripts) are $C_{1212}(\underline{q},t)$, C_{1111}, C_{2211} and C_{2222}. The Laplace transforms of the first 3 functions calculated as described above coincide with expressions given in eqs. (15) - (17) once the latter are simplified by replacing $G(q,s)$ with $G(s)$, $K(q,s)$ with $K(s)$, and $M(q,s)$ with $M(s) = K(s) - 2G(s)$. The last function, C_{2222}, which cannot be deduced from a FDT relation, takes the form:

$$C_{2222}(q,s) \equiv C_2(q,s) \simeq T\left[K(s) - \frac{q^2 M(s)^2}{\rho s + q^2 K(s)}\right], \quad 0 < q \ll 1/\xi_s \tag{23}$$

(We first obtain the Fourier transform of $C_{2222}(q,t)$ with respect to *time*, and then the Laplace transform of this function.) It is obvious that while $C_{2222}(t)$ generally differs from $C_{1111}(t)$, they tend to the same limit ($= TK(t)$) as $q \to 0$ at a fixed time.

Using the results at hand and the space isotropy, the correlation tensor function can be written in the following general form (valid for any coordinate frame):

$$C_{\alpha\beta\alpha'\beta'}(\underline{q},s) = (C_2 - 2C_T)\,\delta_{\alpha\beta}\delta_{\alpha'\beta'} + (C_\perp - C_2 + 2C_T)\,(q_\alpha q_\beta \delta_{\alpha'\beta'} + q_{\alpha'} q_{\beta'} \delta_{\alpha\beta})/q^2 +$$

$$+ C_T(\delta_{\alpha\alpha'}\delta_{\beta\beta'} + \delta_{\alpha\beta'}\delta_{\beta\alpha'}) + \left(C_\parallel + C_2 - 2C_\perp - 4C_T\right) q_\alpha q_\beta q_{\alpha'} q_{\beta'}/q^4 \tag{24}$$

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[12] In particular, we can neglect the q-dependence of the generalized viscosity $\eta(q) = \int_0^\infty G(q,t)\mathrm{d}t$ (cf. eq. (50)). This dependence was studied in detail by simulations of a model glass-former [72] showing that while $\eta(q)$ is significantly lower than the macroscopic viscosity $\eta = \eta(0)$ for $q\xi_\eta \gtrsim 1$, the q-dependence of the generalized viscosity can be neglected for $q\xi_\eta \ll 1$, where the characteristic viscosity-based length ξ_η grows up to ~ 4 particle diameters ($\sim 4a_m$) in a highly supercooled state. It appears therefore that ξ_η is similar to the static correlation length ξ_s which typically increases up to ~ 5 particle diameters near T_g. [46–49] Accordingly, the condition $q\xi_s \ll 1$ is applied in what follows to specify the region where the material functions are nearly independent of q.

where $C_2 = C_2(q,s)$ is defined in eq. (23), and the functions $C_T = C_T(q,s)$, $C_{||} = C_{||}(q,s)$ and $C_\perp = C_\perp(q,s)$ are defined in eqs. (15) - (17).

In particular, for the correlations of the shear stress (σ_{xy}, where x, y are the fixed-frame coordinates) we get the following general expression

$$C_{xyxy}(\underline{q},s) = C_T + \left(C_{||} + C_2 - 2C_\perp - 4C_T\right) q_x^2 q_y^2/q^4 \simeq$$

$$\simeq \frac{T\rho s G(s)}{\rho s + q^2 G(s)} + 4Tq^2 G(s)^2 \left(\frac{1}{\rho s + q^2 G(s)} - \frac{1}{\rho s + q^2 K(s)}\right) \frac{q_x^2 q_y^2}{q^4} \qquad (25)$$

which is valid for $q\xi_s \ll 1$. The above equation, valid for compressible 2D systems, generalizes equation 2 of ref. [15]. The latter equation formally agrees with eq. (25) if we suppress the term involving $K(s)$ there (thus assuming the incompressibility limit, $K \to \infty$) and take into account that $q^2 = q_x^2 + q_y^2$ in 2D. The correlation function defined in eq. (25) is analyzed in different time-distance regimes in the next section.

4. Stress correlations in real space/time

The spatial and temporal correlations of local shear stress in infinite 2D systems are analyzed in this section based on eq. (25). Formally the problem to obtain the correlation function $C(\underline{r},t)$ boils down to calculating the inverse Fourier and Laplace transforms of $C(q,s)$ (we omit the subscripts $xyxy$ here and below for simplicity). The latter function as defined in eq. (25) consist of two terms: $C = C^{is} + C^s$. The first term C^{is} is *isotropic*; the second term C^s is anisotropic and *singular* at $q = 0$, $s = 0$ reflecting its long-range and long-time behavior (in the asymptotic sense as clarified below).

We first turn to the isotropic part

$$C^{is}(q,s) = T\frac{\rho s G(s)}{\rho s + q^2 G(s)} \qquad (26)$$

The only unknown function involved here is $G(s) = \int_0^\infty G(t)e^{-st}\mathrm{d}t$, where $G(t)$ is the shear relaxation modulus. Below we assume that $G(t)$ develops a plateau in the time-range between τ_{min} and τ_{pl} (in the case of supercooled fluids τ_{min} is the characteristic time of fast vibrational relaxation); by contrast, at longer times exceeding some $\tau_{max} > \tau_{pl}$ the relaxation modulus is assumed to decay significantly, either exponentially or, at least, faster than $1/t$. Such behavior is typical of entangled polymer systems and of supercooled liquids near T_g (note that τ_{max} can become practically infinite below T_g). We thus focus on the two main regimes of stress relaxation: (i) the plateau regime $\tau_{min} \ll t \ll \tau_{pl}$, where $G(t) \simeq G_{pl}$ is nearly constant, and (ii) the long-time regime, $t \gg \tau_{max}$, where $G(t)$ is small ($G(t) \ll G_{pl}$). In the first regime $G(s) \simeq G_{pl}/s$, hence

$$C^{is}(q,s) \simeq T\frac{G_{pl}s}{s^2 + q^2 c_T^2}, \quad s \gg 1/\tau_{pl} \qquad (27)$$

where $c_T = (G_{pl}/\rho)^{1/2}$ is the transverse (shear) sound velocity. The correlation function in real space-time reads (for $t \ll \tau_{pl}$; here and below we have in mind but

do not mention explicitly another condition $t \gg \tau_{min}$ involving the plateau onset time τ_{min}):

$$C^{is}(\underline{r},t) \simeq TG_{pl} \int \cos(qc_T t)\, e^{\mathrm{i}\underline{q}\cdot\underline{r}} \mathrm{d}^2 q/(2\pi)^2 \tag{28}$$

For $t = 0$ the r.h.s. of the above equation formally gives $TG_{pl}\delta(\underline{r})$ pointing to local stress correlations. Doing the integral for $t > 0$ we get

$$C^{is}(\underline{r},t) \simeq T\frac{\rho}{2\pi t^2}\varphi\left(\frac{r}{c_T t}\right), \quad t \ll \tau_{pl} \tag{29}$$

where

$$\varphi(x) = -\left(1-x^2\right)_+^{-3/2} \tag{30}$$

Here the subscript '+' means truncation of the negative part: $(y)_+^\nu = y^\nu$ if $y > 0$, $(y)_+^\nu = 0$ if $y < 0$; y is a real expression and the exponent ν is a parameter. Thus C^{is} is negative (anticorrelation of shear stress) for $r < l_1 = c_T t$, and it vanishes at larger distances: $C^{is} = 0$ at $r > l_1$. The 'isotropic' stress correlations therefore decay in time as t^{-2}, while their range $l_1 = c_T t$ is defined by the transverse sound.

Eq. (26) implies that

$$\int C^{is}(r,t)\mathrm{d}^2 r = TG(t)$$

for any t, hence it must be nearly equal to TG_{pl} in the short-time regime we consider. On the other hand, using eq. (29) we find

$$\int C^{is}(r,t)\mathrm{d}^2 r \simeq TG_{pl} I$$

where

$$I = \int \varphi(x)\mathrm{d}^2 x/(2\pi) \tag{31}$$

Therefore, it must be expected that $I = 1$, which may seem to contradict to the above definition of $\varphi(x)$ implying that it is either negative or zero. Moreover, the integral I seems to be divergent. Fortunately, both this contradiction and the divergence are actually fictitious. In fact, the formally diverging integral, eq. (31), can be defined in terms of the analytical continuation replacing the exponent (-3/2) in eq. (30) by a parameter. Alternatively the whole function φ can be defined as

$$\varphi(x) = -\Re \lim_{\epsilon \to 0} \left(1 + \mathrm{i}\epsilon - x^2\right)^{-3/2} \tag{32}$$

with the idea that the limit must be taken after integration. The latter equation agrees with eq. (30) and can be used to calculate any integral involving $\varphi(x)$ by first taking the integral, and then setting $\epsilon \to 0$. Both ways give $I = 1$ as it should be.

Let us turn to the long-time regime, $t \gg \tau_{max}$, roughly corresponding to $s \ll 1/\tau_{max}$. Here $G(s) = \int_0^\infty G(t)\exp(-st)\mathrm{d}t$ is nearly independent of s: $G(s) \simeq \eta$, where

$$\eta = \int_0^\infty G(t)\mathrm{d}t$$

is the shear viscosity. So

$$C^{is}(q,s)/T \simeq \frac{s\eta}{s+q^2\eta/\rho} = \eta\left(1 - \frac{q^2 D}{s+q^2 D}\right) \tag{33}$$

where $D = \eta/\rho$ is the vorticity (transverse momentum) diffusion constant (also known as kinematic viscosity of the fluid [2]). The inverse Fourier-Laplace transforms of $C^{is}(q,s)$ then give

$$C^{is}(r,t) \simeq T\frac{\rho}{4\pi t^2}\varphi_2\left(\frac{r}{2l_2(t)}\right), \quad t \gg \tau_{max} \tag{34}$$

where

$$l_2(t) = \sqrt{Dt}, \quad \varphi_2(x) = \left(x^2-1\right)\exp\left(-x^2\right)$$

Thus, in both time-regimes (short-time and long-time) $C^{is}(r,t)$ behaves qualitatively in the same way: $C^{is} \sim T\rho/t^2$ for $r \lesssim \ell(t)$, while the isotropic stress correlations are strongly suppressed, $C^{is} \to 0$, for $r \gg \ell(t)$, where the propagation length $\ell(t)$ can be generally defined as [27]

$$\ell(t) \sim \sqrt{\eta(t)t/\rho} \tag{35}$$

Here

$$\eta(t) = \int_0^t G(t)\mathrm{d}t \sim G(s = 1/t)$$

is the effective time-dependent viscosity [27]. In the two time-regimes considered above this length is

$$\ell(t) \sim \begin{cases} l_1 = c_T t\ , & t \lesssim \tau_{pl} \\ l_2 = \sqrt{\eta t/\rho}\ , & t \gtrsim \tau_{max} \end{cases} \tag{36}$$

Therefore, the propagation length $\ell(t)$ is elasto-inertial (acoustic) in nature in the short-time regime, and visco-inertial (diffusive momentum spreading) at long times.

Let us now turn to the singular part of stress correlations

$$C^s(\underline{q},s) = 4T\frac{q_x^2 q_y^2}{q^2}G(s)^2\left(\frac{1}{\rho s + q^2 G(s)} - \frac{1}{\rho s + q^2 K(s)}\right) \tag{37}$$

At short times, $t \ll \tau_{pl}$, the relaxation moduli can be approximated as (the condition $\tau_{min} \ll t$ is also assumed here and below)

$$G(s) \simeq G_{pl}/s, \quad K(s) \simeq K_{pl}/s$$

Doing then the inverse Laplace transformation of eq. (37), we get

$$C^s(\underline{q},t) \simeq 4T\frac{G_{pl}^2}{\rho}\frac{q_x^2 q_y^2}{q^4}\left[\frac{1-\cos(qc_T t)}{c_T^2} - \frac{1-\cos(qc_L t)}{c_L^2}\right], \quad t \ll \tau_{pl} \tag{38}$$

where $c_L = \sqrt{K_{pl}/\rho}$ is the longitudinal sound velocity. Transforming it to the real space correlation function yields

$$C^s(\underline{r},t) \simeq \frac{TG_{pl}^2}{2\pi\rho t^2}\left[c_T^{-4}\psi\left(\frac{r}{c_T t},\theta\right) - c_L^{-4}\psi\left(\frac{r}{c_L t},\theta\right)\right] \tag{39}$$

where θ is the angle between $\underline{r}$ and the x-axis, and the new nondimensional function ψ is

$$\psi(x,\theta) = \sin^2(2\theta)\left[4\delta(x^2) + \left(1-x^2\right)_+^{-3/2}\right] + \frac{4\cos(4\theta)}{x^4}\left[-3+\phi(x)\right] \tag{40}$$

Here x is the reduced radius, $\delta(y)$ is the symmetric (even) Dirac's delta-function: $\int_0^\infty \delta(y)\mathrm{d}y = 1/2$, and

$$\phi(x) = \left(3-2x^2\right)\left(1-x^2\right)_+^{-1/2} \tag{41}$$

(the meaning of the '+' symbol is explained below eq. (30)).

The full stress correlation function $C = C^{is} + C^s$ for $\tau_{min} \ll t \ll \tau_{pl}$ is defined in eqs. (39), (29). It is useful to write it in a more explicit way:

$$C(\underline{r},t)/T \simeq \frac{G_{pl}}{2}\left(1-\frac{c_T^2}{c_L^2}\right)\delta(\underline{r}) -$$

$$-\frac{\rho}{2\pi t^2}\left[\cos^2(2\theta)\left(1-\frac{r^2}{(c_T t)^2}\right)_+^{-3/2} + \sin^2(2\theta)\left(\frac{c_T}{c_L}\right)^4\left(1-\frac{r^2}{(c_L t)^2}\right)_+^{-3/2}\right] +$$

$$+\frac{2}{\pi}G_{pl}\frac{(c_T t)^2}{r^4}\cos(4\theta)\left[\phi\left(\frac{r}{c_T t}\right) - \phi\left(\frac{r}{c_L t}\right)\right] \tag{42}$$

Thus, there are 3 contributions to the correlation function here:

(i) The first $\delta(\underline{r})$-term reflects the *local* structural correlations; it is nearly constant for $\tau_{min} \ll t \ll \tau_{pl}$ due to little change of the memory function $G(t)$ in this regime.

(ii) The second term is due to acoustic waves; it decays rather fast as $1/t^2$, and its range is characterized by two finite length scales, $\ell \simeq c_T t$ and $\ell' \simeq c_L t$ which both increase in time. Note that this correlation contribution strongly decreases at $r \approx \ell$: it is typically much lower for $r > \ell$ than for $r < \ell$ since normally $(c_T/c_L)^4 \ll 1$. This term is anisotropic: it depends on the $\underline{r}$-direction given by θ and includes both isotropic and quadrupolar ($\cos(4\theta)$) parts. Further, being proportional to ρ/t^2 this contribution can be considered as 'inertial' in nature. Noteworthily, it is negative (corresponding to anti-correlation of the shear stresses) for any θ and distances in the transverse wave zone, $r < c_T t$ (that is, away from the wave fronts where the correlation function changes its sign).

(iii) The last term is of major interest: it is long-range (with correlation lengths $\ell \simeq c_T t$ and $\ell' \simeq c_L t$) and it does not decay in time in the regime $\tau_{min} \ll t \ll \tau_{pl}$ we consider. This term is not relevant for simple liquids well above the glass transition temperature T_g (due to short relaxation time τ_{max}), and it is not present

in crystalline solids [13], but it becomes dominant in viscoelastic liquids for $t \gg r/c_T$. It implies the existence of significant transient (but persistent) stress fluctuations in such liquids (in particular, supercooled liquids close to the glass transition). In the most important regime $t \gg r/c_T$ (note that in supercooled liquids the latter condition is compatible with long $r \gg a_m$ since typically $c_T \tau_{pl} \gg a_m$ in these systems near or below T_g) the dominant contribution to the stress correlation function, eq. (42), can be approximated as

$$C(\underline{r},t) \simeq -\frac{T}{\pi} G_{pl} \cos(4\theta) \left(1 - \frac{G_{pl}}{K_{pl}}\right) r^{-2}, \quad r \ll c_T t, \quad \tau_{min} \ll t \ll \tau_{pl} \tag{43}$$

In this regime the correlation function $C(\underline{r},t)$ is thus nearly independent of time and is long-range (LR) in space showing an algebraic decay as $1/r^2$. It is noteworthy that the magnitude of the long-range correlations depends on both elastic moduli, G_{pl} and K_{pl}. Eq. (43) qualitatively agrees with molecular dynamics simulation results of a 2D binary glass former [12].

It is also interesting that stress correlations for incompressible systems (in the formal limit $K \to \infty$ corresponding to zero compressibility) cannot be obtained by just suppression of all the terms involving c_L in the above equations. In fact, an absolute incompressibility leads to the emergence of the following behavior of $C(\underline{r},t)$ for $r > c_T t$ (defined by the third term, involving the ϕ-function, $\phi(r/(c_L t))$, in the r.h.s. of eq. (42): this term dominates for $r > c_T t$):

$$C(\underline{r},t) \simeq -\frac{6}{\pi} T G_{pl} \frac{(c_T t)^2}{r^4} \cos(4\theta), \quad \tau_{min} \ll t \ll \tau_{pl} \tag{44}$$

This 'incompressible' correlation function is always truly long-range (for any $t > 0$). It shows the $1/r^4$ power law decay for however long r, with an amplitude which is increasing in time (for $t \lesssim \tau_{pl}$). Such infinite-range stress correlations implied by the results of ref. [15] come as a consequence of the incompressible liquid model adopted in ref. [15]. It is important, however, that the correlation term defined in eq. (44) is eventually canceled out when both transverse and longitudinal velocity modes are properly taken into account.

So far we have considered the short-time regime $t \ll \tau_{pl}$. The latter condition is rather restrictive for viscoelastic liquids which normally show a very broad spectrum of relaxation times with a large gap between τ_{min} and τ_{max}. Nevertheless, the detailed analysis of the short-time regime given above is useful to show how the long-range character of stress correlations emerges and develops in time (as illustrated in Figs. 1, 2).

Below we lift the short-time restriction, leaving essentially the only condition $r \ll c_T t$, which is likely to be valid in viscoelastic liquids (generally due to rather high sound velocity and long relaxation times in these systems). The singular terms dominate in eq. (25) in this case, hence

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[13] The reason is that in crystals the correlation function $C(r,t)$ decays rather fast with the characteristic time $\sim r/c_T$ (since the relevant stress fluctuations are due to thermal acoustic waves with wave-vector $k \sim 1/r$ and frequency $\sim k c_T$). For $t \gg r/c_T$ the decay of C due to destructive interference follows the $1/t^2$ power law (in 2D) which is actually given by the second term in eq. (42) in the case of isotropic crystals.

$$C(\underline{q},s) \simeq 4T\frac{q_x^2 q_y^2}{q^4}\tilde{G}(s), \quad \tilde{G}(s) \equiv G(s) - \frac{G(s)^2}{K(s)} \tag{45}$$

The inverse Laplace transform of $\tilde{G}(s)$ gives the effective relaxation modulus $\tilde{G}(t)$ which behaves similarly to the standard shear modulus $G(t)$: in the liquid state $\tilde{G} \to 0$ for $t \gg \tau_{max}$, while in the glassy state it tends to a finite level $\tilde{G}_\infty = G_\infty - \frac{G_\infty^2}{K_\infty}$ for $t \sim t_{lab}$.

Doing also the inverse Fourier transform of eq. (45) we get the stress correlation function in real space-time:

$$C(\underline{r},t) \simeq -\frac{T}{\pi}\cos(4\theta)\tilde{G}(t)r^{-2}, \quad r \ll \ell(t) \tag{46}$$

where the propagation length $\ell(t)$ is defined in eq. (35). This result agrees with eq. (43) generalizing it. Thus, the LR stress correlations gradually decay in time and vanish for $t \gg \tau_{max}$ in the liquid state, or tend to a plateau (proportional to $\tilde{G}_\infty$) for vitrified systems. The latter conclusion qualitatively agrees with the theoretical results [15] obtained for 3D supercooled liquids.

Note that while the results for $C(\underline{q},t)$ obtained in this section are valid for 2D systems, they are also fully applicable in 3 (and higher) dimensions if $\underline{q}$ belongs to the xy plane. Note also that the stress correlations at exactly $t = 0$ (rather than for $t \gg \tau_{min}$) can be deduced directly from the general eq. (25). The general result is

$$C(\underline{r},0) = C^{is}(\underline{r},0) = TG(0)\delta(\underline{r}) \tag{47}$$

pointing to localized static (structural) stress correlations in agreement with our assumption of short ξ_s.

In the next section we discuss the obtained results pointing, in particular, that the revealed long-range character of stress correlations is not related to the static structural correlations which are typically short-range in liquid systems. We also provide a simple physical argument showing that stress correlations in isotropic complex liquids and amorphous systems must be long-range and anisotropic.

5. Discussion and summary

In this paper we analyzed the local stress tensor correlations in complex and supercooled liquids focusing on the time and distance dependencies of the shear stress correlation function $C(\underline{r},t)$. Our approach is based upon the fluctuation-dissipation theorem (FDT) and the concept of random forces due to local structural and momentum thermal fluctuations. It is similar in spirit to the classical theory of hydrodynamic fluctuations [29,30] and can be traced back to the theorems on regression of fluctuations and reciprocal relations in irreversible processes proposed long ago by Onsager [34–37].

The fluids we consider are characterized by slowly fading memory and long structural relaxations including the stress relaxation with the longest time τ_{max}. Such fluids are viscoelastic by definition: for $t \ll \tau_{max}$ they show quasi-elastic behavior (with considerable transient elastic moduli), while for $t \gg \tau_{max}$ they behave as highly viscous media. This property is equally applicable to macromolecular

solutions or melts and to glass-forming simple liquids, the major difference being that polymer systems are normally highly elastic, while glasses are typically brittle. This difference however is not important for the present analysis since we consider weak fluctuations near the equilibrium state rather than non-linear flow effects.

The present theory assumes that ***structural*** correlations (due to irregularities of molecular packing) are local (short-range) in liquid systems we consider. This is true for all equilibrium viscoelastic liquids known so far (including glass-forming systems): the static structural correlation length ξ_s can increase as the system is cooled down close to the vitrification temperature [38,39,43–45], but this increase is typically limited by a few (at most ~ 10) molecular sizes. [40–42,46–49] It is for this reason that we actually neglect the q-dependence of the generalized relaxation moduli G, K and M as such dependence is essentially of structural origin.

Indeed, the $q = 0$ approximation for material functions (cf. eqs. (22)) is equivalent to stating that these functions (relaxation moduli) reflect a localized in space response. The argument concerning their locality (around eqs. (22)) is based on the assumption that the local stress is defined by the local momentum flux (transverse current) and local deformation history of a fluid element (by 'local' we mean short-range within the structural correlation length ξ_s). Now, it is the deformation history (and, therefore, the momentum flux) that is controlled by the definition of the material functions (cf. the constitutive relation (11)) providing the stress response to a prescribed deformation field. The locality of the relaxation moduli then comes merely from the fact that a small deformation of a fluid element 1 (of size somewhat exceeding ξ_s) does not affect much the local stress in a distant fluid element 2 if the latter ***is kept undeformed***. By contrast, the deformation and momentum flux are ***not*** prescribed in eq. (6) (which is equivalent to eq. (7)) apart from the instant initial deformation at $t = 0$: at any later time, $t > 0$, the liquid is being deformed further due to the generated internal stresses. This effect gives rise to fast momentum propagation and, as a result, to significant non-locality of the stress response. So, the distinction of eq. (7) (implying a non-local response) from the constitutive relation, eq. (11) (reflecting the local response) is that the former corresponds to the process in which neither the deformation history, nor the momentum flux are prescribed. To resume, equation (7) allows for internal flow (momentum flux) as a consequence of the external perturbation, whereas the constitutive relation (11) defines the material functions solely in response to the prescribed external deformation (no 'internal' flow on top of it). With no such flow (due to internal forces) the information about a perturbation at one point cannot be transmitted to a distant point, so the response is local.

The effects we consider in this paper correspond to length-scales $\gg \xi_s$. It is demonstrated here that non-simultaneous stress correlations (with finite time-shift t) are long-range (LR) in viscoelastic liquids in agreement with the ideas of ref. [15]. As a major result we show that the stress correlations decay algebraically (rather than exponentially) with distance r, as $1/r^2$ for $r < \ell(t)$ in 2D systems, and that they decay in time only slowly for $t \lesssim \tau_{max}$. Such behavior is of course very different from what is known about the classical simple liquids [29,30]. Moreover, in the glassy state the stress correlation function is predicted to show a finite plateau level in the long-time regime where the liquid is characterized by a nearly constant shear modulus, $G(t) \approx G_\infty$. Similar conclusions have been drawn in ref. [15] based on a different theoretical approach. The relevant correlation length

$\ell(t)$ (defining the region of validity of the $1/r^2$ correlation law) turns out to be the time-dependent momentum propagation length given in eqs. (35), (36). Thus, the correlation length $\ell(t)$ corresponds to sound propagation at short times and to vorticity diffusion at long times. Both processes are fast in highly viscous (viscoelastic or glass-forming) liquids leading to a long length-scale $\ell(t)$ for the characteristic time range coming from the relaxation spectrum of the liquid.

Note that transverse sound waves (with wavelength $\lambda \lesssim \ell(t)$) are not significantly damped in the time regime $\tau_{min} \ll t \ll \tau_{pl}$ corresponding to the rubbery plateau of the shear relaxation modulus $G(t)$: the liquid shows an elastic response in this time range, so the propagation length here is proportional to the sound velocity, $\ell(t) \simeq c_T t$. By contrast, the sound waves get dumped for $t \gtrsim \tau_{pl}$: the liquid response then becomes viscoelastic with the propagation length defined by the generalized time-dependent viscosity $\eta(t)$: $\ell(t) \sim \sqrt{\eta(t)t/\rho}$ (cf. eq. (35)). In this regime ℓ increases with t in a sublinear fashion.

Thus, we show that very long-range and anisotropic correlations of stress must develop in the transient regime $t \lesssim \tau_{max}$ with the characteristic correlation range (for $t \sim \tau_{max}$) being $\ell_{max} \sim (\eta\tau_{max}/\rho)^{1/2}$, where η is the zero-shear viscosity of the liquid. In viscoelastic (glass-forming) liquids η is high (it is roughly proportional to the relaxation time τ_{max}), so the vorticity diffusion is fast giving rise to long ℓ_{max} strongly exceeding the molecular size a_m.

Noteworthily, the stress correlations do not decay exponentially for $r > \ell$: in the distal region $\ell < r < \ell' \approx c_L t$ they still follow a power law, but with a significantly faster decrease with r, $C(\underline{r}, t) \propto r^{-4}$ (cf. Fig. 1). Here ℓ' is the longest correlation length defined by the longitudinal sound velocity c_L. Interestingly, in the plateau time-regime ($\tau_{min} < t < \tau_{pl}$) the correlation function *increases* with time in the distal region $\ell < r < \ell'$: it is proportional there to $t^2 G_{pl}^2 r^{-4}$ (cf. the second part of the last term in eq. (42)).

Thus, the developed theory allows to see how the long-range stress correlations gradually emerge as a function of time-shift (t) starting from purely local simultaneous stress correlations (see eq. (42)). We also predict how the magnitude of the long-range stress correlations gradually decreases in time according to the relaxation law reflecting both shear and longitudinal memory functions, $G(t)$ and $K(t)$ (cf. eqs. (45), (46)).

To further clarify the main results obtained here let us first recall what is known about the stress correlations in ordered (crystalline) solids. The fluctuations of stress there are essentially due to thermally excited acoustic waves. The correlation function of simultaneous fluctuations (with time-shift $t = 0$) is long-range (more precisely, infinite range) and anisotropic as follows from the classical elasticity theory [26]. However, the stress fluctuations in solids decay rather fast with characteristic time $t \sim r/c$ corresponding to frequency of acoustic waves with wave-length $\sim r$ (c is the sound velocity). Thus, the stress fluctuations become uncorrelated at $t \gg r/c$. Similar high-frequency stress fluctuations (σ^f) are also present in viscoelastic (glass-forming) fluids which behave as solid at short times, but this fluctuation field is superimposed with a quasi-permanent (virtually frozen) heterogeneous random stress pattern. The 'frozen' (inherent) stress σ^i does not cause directly any fluid motion: being compatible with mechanical equilibrium it does not generate any internal force, $\sigma^i_{\alpha\beta,\beta} = 0$. However, it is important to emphasize that the inherent (transiently frozen) stress is necessarily present in viscoelastic fluids:

it serves to restore the short-range character of the *total* stress correlations at $t = 0$ (recall that these correlations are structural in nature). Thus, the correlation function of the total stress, $\sigma = \sigma^f + \sigma^i$, is $C(\underline{r}, 0) = C^f(\underline{r}, 0) + C^i(\underline{r}, 0) \simeq 0$ for $r \gg \xi_s$. Hence, the correlation function of the inherent stress, C^i, must be nearly opposite to the function C^f due to acoustic stress fluctuations. Therefore, C^i must also show infinite-range and anisotropic correlations. As the fluctuation part decays rather fast, the stress correlations at longer times ($t \gg r/c$) must be mostly due to the inherent (frozen) stress, hence they must be also long-range and stay for a long time $\sim \tau_{max}$. [14] Such long-range (and nearly frozen) 'elastic' stress fields have be considered in the framework of elasto-plastic models [57–59]. It is also worth mentioning that the anisotropic character of the frozen stress correlations directly follows from the condition of mechanical equilibrium ($\sigma^i_{\alpha\beta,\beta} = 0$) which demands that shear stress must vanish for certain directions of the wave-vector $\underline{q}$. For example, $\sigma^i_{xy} = 0$ if $\underline{q} \neq 0$ is parallel to either x or y axis. The latter example also explains why the second ('singular') term in the stress correlation function (cf. eq. (25)) shows knots for $q_x = 0$ or $q_y = 0$.

It is worth emphasizing again that the predicted long-range spatial and temporal stress correlations are essentially *viscoelastic* in nature. The long-range character of $C(\underline{r}, t)$ is not exactly an inertial effect (in contrast to fast fluctuations due to acoustic waves): in fact, the correlation function is independent of the fluid density ρ for $r \ll \ell(t)$ (in this regime C is defined in eq. (46)). Moreover, C is well-defined in the formal limit $\rho \to 0$ where the infinite range character of stress correlations emerges instantly.

As explained above, the revealed LR stress correlations are dynamical in nature and are not directly related to the static correlation length ξ_s. [15] Let us recall that supercooled liquids are often characterized by a *dynamical* correlation length $\xi_{\rm d}$ which depends—as $\ell(t)$—on the time scale [50–52,40,53–55,18,21,22]. Can we associate the stress correlation range with $\xi_{\rm d}$? The point is that $\xi_{\rm d}$ depends on the nature of variables whose correlations are studied, so many dynamical lengths $\xi_{\rm d}$ can be defined at the same time scale [56,23]. Typically, $\xi_{\rm d}$ is associated with the emergence of dynamical heterogeneities in glass-forming fluids [23] and refers to the size of an increasing number of particles with correlated motion at low T (e.g. the span of one-dimensional string-like motion [21,22], or the length scale associated with dynamic fluctuations as measured by dynamic susceptibilities [50–52,40,53–56], or a length scale characterizing the distance between localized excitations of high mobility [24]). While stress fluctuations certainly affect the cooperative motion of fluid particles, the usually defined $\xi_{\rm d}$ is expected to be much shorter than ℓ, ℓ' [18].

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[14] Some evidence for such distinction between the inherent structure dynamics and the overall liquid dynamics can be found in simulation studies (see Figs. 5, 6 in ref. [68] and Fig. 1 in Supplemental Material of ref. [12]).

[15] However, this does not imply that the LR stress correlations are completely unrelated to the static glass correlation length ξ_s. Since $\ell(t) \sim \sqrt{\eta t/\rho}$ (cf. eq. (35)) at long times, ℓ and ξ_s would be related if ξ_s indeed determines the increase of the viscosity at low T as $\eta/\eta_0 \sim \exp(E_A(T)/T)$ with the putative equation $E_A(T) \propto \xi_s(T)^\psi$ [17] and $\psi \leq d$ (d being the spatial dimension).

It appears that the length $\ell_{max} = \ell(\tau_{max})$ is rather analogous to the dynamical length-scale associated with oscillatory behavior of the transverse current correlation function C^{JJ} used to characterize the spatial extent of solid-like regions in some experimental and theoretical studies. [19,20,18] In the general case this function is defined as

$$C^{JJ}_{\alpha\beta}(\underline{q},t) = \frac{1}{V\rho^2}\left\langle J_\alpha(\underline{q},t)J^*_\beta(\underline{q},0)\right\rangle$$

where $J_\alpha(\underline{q},t)$ is the α-th component of the current (the momentum flux, cf. eq. (13)). Noteworthily, this correlation matrix function is closely related to the stress correlation function. Using eq. (13) we get in the Fourier-Laplace representation:

$$C^{JJ}_{\alpha\alpha'}\left(\underline{q},s\right) = \frac{T}{\rho s}\delta_{\alpha\alpha'} - q_\beta q_{\beta'} C_{\alpha\beta\alpha'\beta'}\left(\underline{q},s\right)/\left(\rho s\right)^2$$

A more special relation of that sort was derived (cf. refs. [73,31,27]) for the *transverse* current correlations (when $\underline{q}$ is perpendicular to the velocity direction):

$$C^{JJ}_T(q,s) \equiv C^{JJ}_{xx}\left(q\underline{e}_y,s\right) = \frac{T}{\rho s + q^2 G(q,s)} \tag{48}$$

On return (by inverse Laplace transform) to the real time t, this correlation function shows oscillations for $t \ll \tau_{max}$ and $q \gg 1/\ell(t)$. A generalization of the above equation for *any* direction of $\underline{q}$ can be derived using eq. (24). The result (where we neglected the q-dependence of the relaxation moduli) is: [73]

$$C^{JJ}_{xx}\left(\underline{q},s\right) \simeq \frac{T}{\rho s}\left\{1 - \frac{G(s)q_y^2}{\rho s + q^2 G(s)} - \frac{K(s)q_x^2}{\rho s + q^2 K(s)}\right\} \tag{49}$$

This function is moderately anisotropic; in the limit $K \to \infty$ the anisotropy is given by the factor q_y^2: $C^{JJ}_{xx} \propto q_y^2/\left[q^2\left(\rho s + q^2 G\right)\right]$. In the real space-time variables the correlation function $C^{JJ}_{xx}\left(\underline{r},t\right)$ decays as $1/\ell(t)^2$ for $r \lesssim \ell(t)$ and it rapidly vanishes for $r \gg \ell(t)$. Therefore, the long-range character of C^{JJ}_{xx} is much weaker than that of the stress correlation function: C^{JJ}_{xx} never shows any frozen long-range effect even in the amorphous solid regime.

Eq. (48), which follows directly from the FDT relation (eq. (15)) for the shear stress correlation function, can be harnessed to define the wave-vector dependent shear viscosity $\eta(q)$ in terms of the transverse current autocorrelation function using the general relation [72,32,33]

$$\eta(q) = \frac{\rho}{q^2}\left[\int_0^\infty \mathrm{d}t C^{JJ}_T(q,t)/C^{JJ}_T(q,t=0)\right]^{-1} = \frac{T}{q^2}\left[\lim_{s\to 0} C^{JJ}_T(q,s)\right]^{-1}$$

By inserting eq. (48) in the equation above we get

$$\eta(q) = \lim_{s\to 0} G(q,s) = G(q,s=0) \tag{50}$$

in agreement with the standard physical definition of the generalized viscosity $\eta(q)$. [2,32,27] It is noteworthy that directly taking $s \to 0$ limit in eq. (15) defining the shear stress correlation function does not help to get $\eta(q)$: it merely shows that

$\lim_{s\to 0} C_T(q,s) = 0$. Hence, the well-known Green-Kubo formula [73,31,32] for the macroscopic shear viscosity in terms of the shear stress autocorrelation function for $q = 0$ is not directly generalizable to the case of nonzero wavevector. This feature is also related to the fact that generally $\lim_{s\to 0}\lim_{q\to 0} C_T(q,s) \neq \lim_{q\to 0}\lim_{s\to 0} C_T(q,s)$ (cf. also footnote 5).

It is remarkable that the r, θ dependencies of the LR part of the stress correlation function, eq. (46), completely agree with the far field stress response to a localized plastic strain event [62,57] which, in turn, was shown to be equivalent to the stress field generated by a pair of 'force dipoles' [57]. The corresponding characteristic quadrupolar (eight-lobe) LR stress pattern was also observed in simulation and experimental studies of the local rearrangements effects in 2D foams [60], 2D glass systems [61] and quasi-2D emulsions [63].

Long-range dynamical correlation effects have been reported in several other recent studies [54,55,12,25]. Computer simulations of ref. [12] reveal LR correlations between plastic events in deeply supercooled 2D liquids, which have been attributed to $1/r^2$ correlations of the local *inherent* stresses a distance r apart, in agreement with our result given in eq. (46). The simulation studies [12] also revealed that the amplitude of power-law stress correlations first grows, then shows a transient plateau and finally decays. That sort of behavior is in harmony with our results (cf. eqs. (42), (46)) showing the early time increase of the correlation amplitude (due to the factor t^2 in the last term of eq. (42)) and then its gradual decrease defined by the $\widetilde{G}(t)$ factor in eq. (46). The LR dynamical correlations between particle displacements and related functions have been demonstrated in refs. [54–56,25]. Remarkably, the studies [54,55] show that the corresponding dynamical correlation length ξ_4 grows linearly in time in glasses, while $\xi_4 \propto \sqrt{\eta t}$ in the long-time regime in liquids, in obvious agreement with the momentum propagation length $\ell(t)$ defining the range of stress correlations considered in the present paper (cf. eq. (36)). Similar results are also found in ref. [25]. It is worth noting that generally the length $\ell(t)$ (cf. eqs. (35), (36)) can be also considered as the range of viscoelastic hydrodynamic interactions in the liquid. [27]

In section 3 we figured out how to obtain the correlation function C^{n} of the random stress (the thermal and structural noise) using a simple physical argument, and how then to derive the correlation function of the total stress, C, based on C^{n}. It is interesting that this approach can be reversed in some sense. Indeed, a few components of the total correlation function C are generally defined in eqs. (15) - (17) in terms of the material relaxation functions. These exact FDT relations can be harnessed to obtain the corresponding components of C^{n}. To this end we use the momentum equation (13), eqs. (8), (11) defining $\boldsymbol{\sigma}^{\mathcal{D}}$ and eq. (19) to find, first, the general relation between the fields of the total stress $\boldsymbol{\sigma}$ and the structural noise $\boldsymbol{\sigma}^n$, then the relation between C^n and C in the frequency (ω) representation for $\omega \neq 0$. Finally, $C^n(q,t)$ is established using the additional condition that C^n must tend to 0 at $t \to \infty$. The resultant relations for the noise correlations are:

$$\frac{1}{T} C^{\mathrm{n}}_{1212}(q,t) = G(q,|t|) - G_e(q)$$

$$\frac{1}{T} C^{\mathrm{n}}_{1111}(q,t) = K(q,|t|) - K_e(q)$$

$$\frac{1}{T}C^{\mathrm{n}}_{2211}(q,t) = M(q,|t|) - M_e(q) \tag{51}$$

where we again use the natural coordinate frame (with axis $\underline{e}_1$ parallel to $\underline{q}$ and $\underline{e}_2$ perpendicular to it). Note that $G_e(q) = 0$ (both at $q = 0$ and $q \neq 0$) for the liquid systems considered herein (including viscoelastic and supercooled liquids). Obviously, given a weak dependence of the generalized moduli G, K, M on q (for $q\xi_s \ll 1$), the exact relations are in perfect agreement with the approximate eq. (21) derived on the physical grounds.

As discussed in section 2, the stress correlation function C defines the stress response after a small instant deformation of the system. It is useful to note that the same function C also defines the stress response to a small external force $f(\underline{r},t)$ applied to all particles in the system ($\underline{r}$ is the particle position). Using the FDT as outlined in ref. [27], we find the linear response in the Fourier-Laplace representation:

$$\Big\langle \sigma_{\alpha\beta}(\underline{q},s) \Big\rangle = \frac{1}{mT}\frac{\mathrm{i}}{s} C_{\alpha\beta\alpha'\beta'}(\underline{q},s) q_{\alpha'} f_{\beta'}(\underline{q},s) \tag{52}$$

In particular, the response to an instant push (momentum increment) at $t = -0$, $f(\underline{r},t) = F(\underline{r})\delta(t+0)$, is

$$\Big\langle \sigma_{\alpha\beta}(\underline{q},s) \Big\rangle = \frac{1}{mT}\frac{\mathrm{i}}{s} C_{\alpha\beta\alpha'\beta'}(\underline{q},s) q_{\alpha'} F_{\beta'}(\underline{q}) \tag{53}$$

The latter equation can be compared with the response to an instant small deformation $\underline{r} \to \underline{r} + \underline{u}(\underline{r})$ at $t = -0$. Using eqs. (7), (10) we get

$$\Big\langle \sigma_{\alpha\beta}(\underline{q},s) \Big\rangle = \frac{\mathrm{i}}{T} C_{\alpha\beta\alpha'\beta'}(\underline{q},s) q_{\alpha'} u_{\beta'}(\underline{q}) \tag{54}$$

Thus, both response functions are closely related to each other: in real time the response to instant deformation is simply proportional to the time derivative of the response to an instant momentum increment. In other words, both types of linear responses (to the external force field and to the deformation) contain essentially the same information. This means, in particular, that the stress correlation function C cannot be fully restored based solely on the response to a 'force dipole' [57,12] or any other external force field (cf. also the discussion at the end of section 2).

It is worth noting that all the results obtained in this paper are valid for equilibrium systems, either ergodic liquids or equilibrium ensembles of amorphous solids (glasses). In practice the glassy systems are normally out of equilibrium; the effect of their non-ergodicity on stress correlations will be considered separately.

It is stress fluctuations in an *infinite* system that are considered in this paper. Simulation studies concern finite box systems, normally with periodic boundary conditions (PBC). The results obtained below for an infinite space are still applicable in the PBC case with box-size L if $\ell(t) \ll L$. In the general case $C(\underline{r},t)$ with PBC is defined (in 2D) as

$$C(\underline{r},t,L) = L^{-2} \sum_{\underline{q}} C(\underline{q},t) \exp\Big(\mathrm{i}\underline{q}\cdot\underline{r}\Big)$$

where $\underline{q} = (2\pi/L)\,\underline{n}$, $\underline{n}$ is the vector whose components take independently all integer values between $-\infty$ and ∞. In the opposite regime $L \ll \ell(t)$ only one term above

(with $q = 0$) matters for the isotropic part of the stress correlation function, C^{is}, thus giving

$$C^{is}(\underline{r}, t, L) \simeq T\left[G(t) - G_e\right]/L^2, \quad \ell(t) \gg L \tag{55}$$

Therefore, C^{is} turns out to be nearly independent of r in the regime $\ell(t) \gg L$ relevant in most simulation studies due to relatively small box-size L. Note that strictly speaking the above equation provides the correlation function $C^{is}(r,t)$ which is coarse-grained over a time-interval $\Delta t \gg tL/\ell(t)$. This interval is relatively short, $\Delta t/t \ll 1$ at long times; the coarse-graining is necessary due to a singular behavior of $C^{is}(\underline{r}, t)$ at the wave front (cf. eq. (29)). Note also that G_e in eq. (55) is the equilibrium shear modulus (cf. eqs. (18)), and that $G_e = 0$ for the systems we consider (ergodic liquids or glassy systems which are fully equilibrated thermodynamically). Turning to the the singular part of the correlation function, C^s, and the full function $C = C^{is} + C^s$ in the regime $\ell(t) \gg L$, the infinite space results (cf. eqs. (39), (42), (46)) remain applicable provided that $r \ll L$.

The last point concerns the effects of temperature variations. In the present paper we widely used the FDT connecting the equilibrium stress correlation function with a stress response to a mechanical perturbation. Analyzing the response functions we implicitly assumed that the temperature is constant (isothermal process). This assumption is valid for shear deformation modes since in this case the energy and entropy change is only quadratic in deformation amplitude. However, it may not be valid in the case of longitudinal strain (which generally leads to a linear change of energy, entropy and temperature). Of course, the isothermicity condition remains generally true if the heat conductivity is very fast (a high heat diffusion coefficient D_H: $D_H \to \infty$). However, in reality, with finite D_H, this assumption fails at short times t. In particular, the infinitesimal canonical deformation at $t \to 0$ is always adiabatic rather than isothermal. More generally, the results obtained in this paper are valid for

$$D_H t \gg r^2 \tag{56}$$

With $t \sim \tau_{max}$ the latter condition still allows for a quite long-range effect, given that the stress relaxation time is long for viscoelastic fluids while their viscoelastic nature does not affect much the heat transport (D_H). We plan to describe the temperature variation effects in a separate publication.

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FIGURE CAPTIONS

FIG. 1. The reduced correlation function $\left|\widetilde{C}\right|$, $\widetilde{C} \equiv Ct^2/(T\rho)$ vs. $\widetilde{r} = r/(c_T t)$ in log-log scale for $\theta = 0$ (a), $\theta = \pi/4$ (b) and $\theta = \pi/8$ (c). The same dependencies for $\theta = 0$, $\pi/4$ in semi-log scale (d) (see also insets of (a), (b)). $\widetilde{C}$ is calculated for $c_L/c_T = \sqrt{K_{pl}/G_{pl}} \approx 2.45$ corresponding to $K_{pl}/G_{pl} = 6$ using eq. (42) without the $\delta(\underline{r})$-term. The dotted vertical lines show the wave fronts ($r = \ell = c_T t$ and $r = \ell' = c_L t$). Note that C for $\theta = \pi/4$ changes its sign at $\widetilde{r} \approx 0.84$, $\widetilde{r} = 1$ and $\widetilde{r} \approx 2.26$.

FIG. 2. Two-dimensional plots of $\widetilde{C} = Ct^2/(T\rho)$ in polar coordinates $(\widetilde{r}, \theta)$ using eq. (42) with the same c_L/c_T; $\widetilde{x} = \widetilde{r}\cos\theta$, $\widetilde{y} = \widetilde{r}\sin\theta$. (a) $\widetilde{C}$ for short $\widetilde{r} \lesssim 0.02$ showing regions of negative (blue to green) and positive (red to green) correlations; black lines separating these regions correspond to $C = 0$. (b) $\left|\widetilde{C}\right|$ for $\widetilde{r} \lesssim 1$ showing the transverse wave front (red/white dashed circle, $\widetilde{r} = 1$). Black curves (separating the regions $C > 0$ and $C < 0$) include 4 'petals' with the central crossing and 4 arcs (belonging to the circle, $\widetilde{r} = 1$). (c) $\widetilde{C}$ for $\widetilde{r} < 1$ using a color code highlighting the behavior in the region $\widetilde{r} < 0.5$. (d) The whole 2D plot for $\left|\widetilde{C}\right|$ showing 2 wave fronts as red/white circles (for transverse, $\widetilde{r} = 1$, and longitudinal, $\widetilde{r} = \ell'/\ell \approx 2.45$, sound waves). $C > 0$ inside 4 small petals (in the region $\widetilde{r} < 1$) and 4 large petals (for $1 < \widetilde{r} < \ell'/\ell$) shown with black contour lines; $C < 0$ outside the petal regions; $C = 0$ for $\widetilde{r} > \ell'/\ell$.

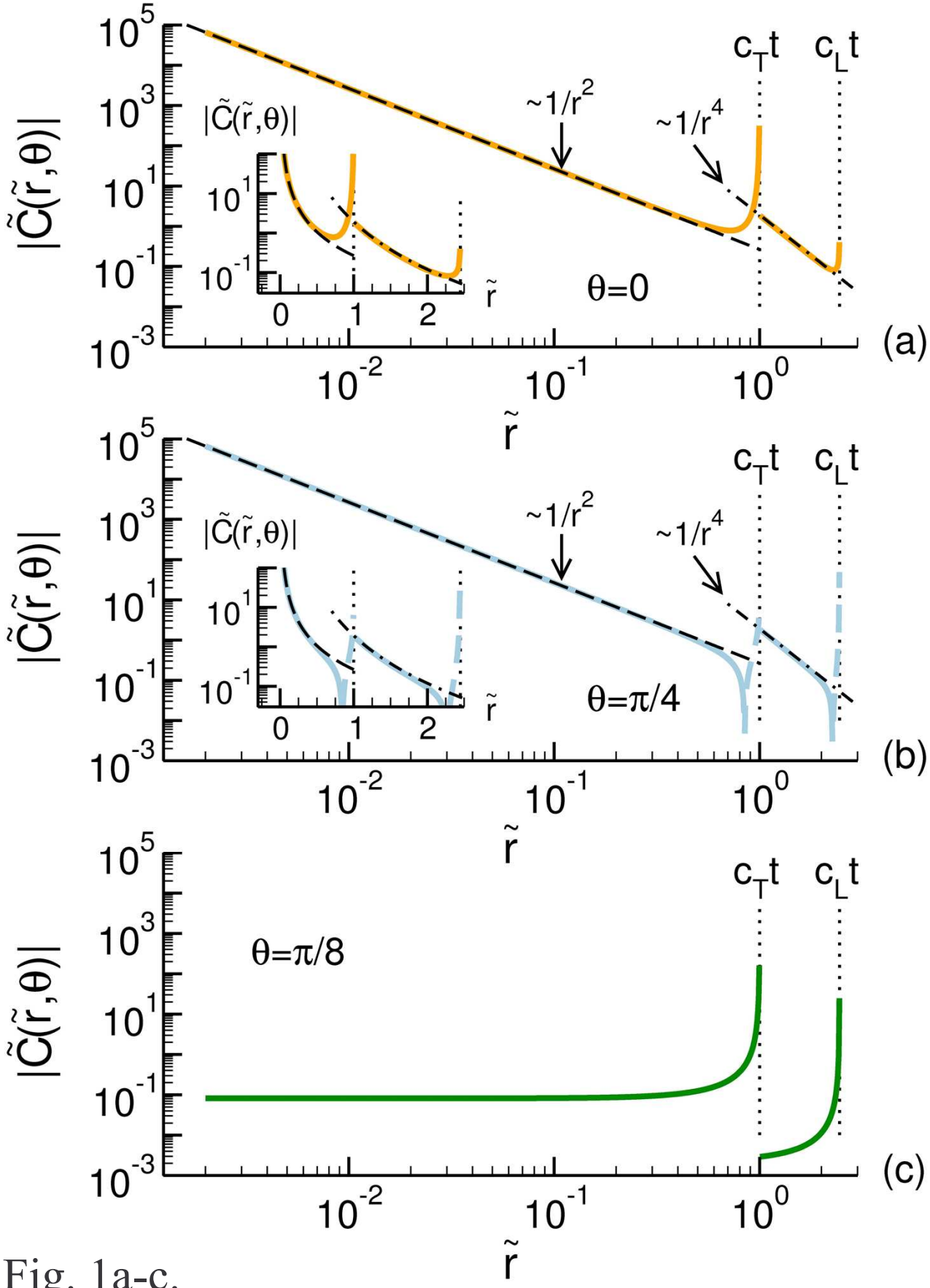
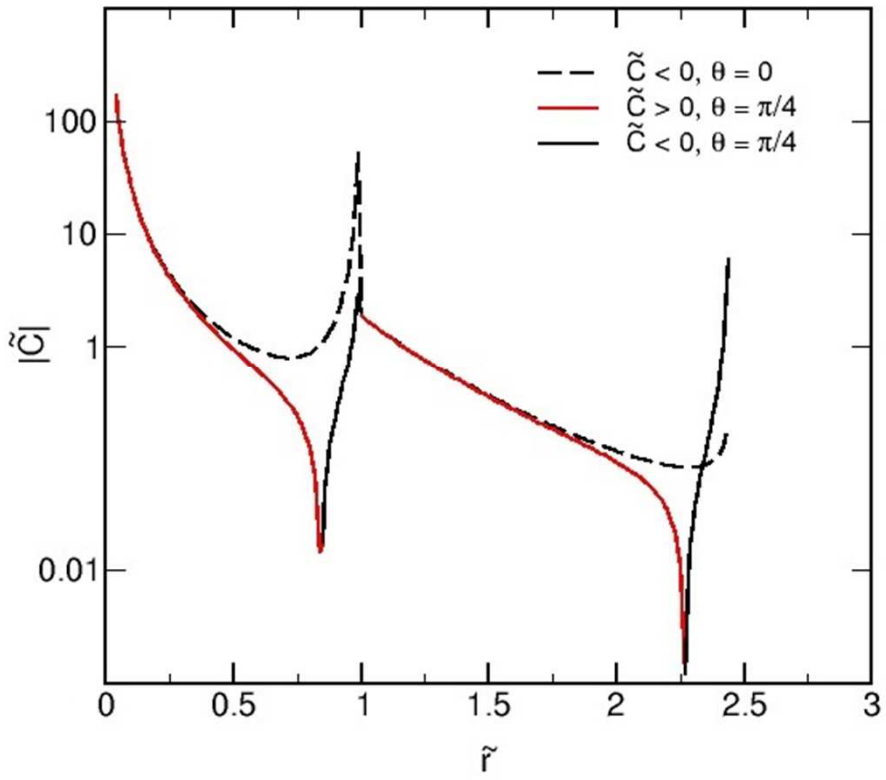
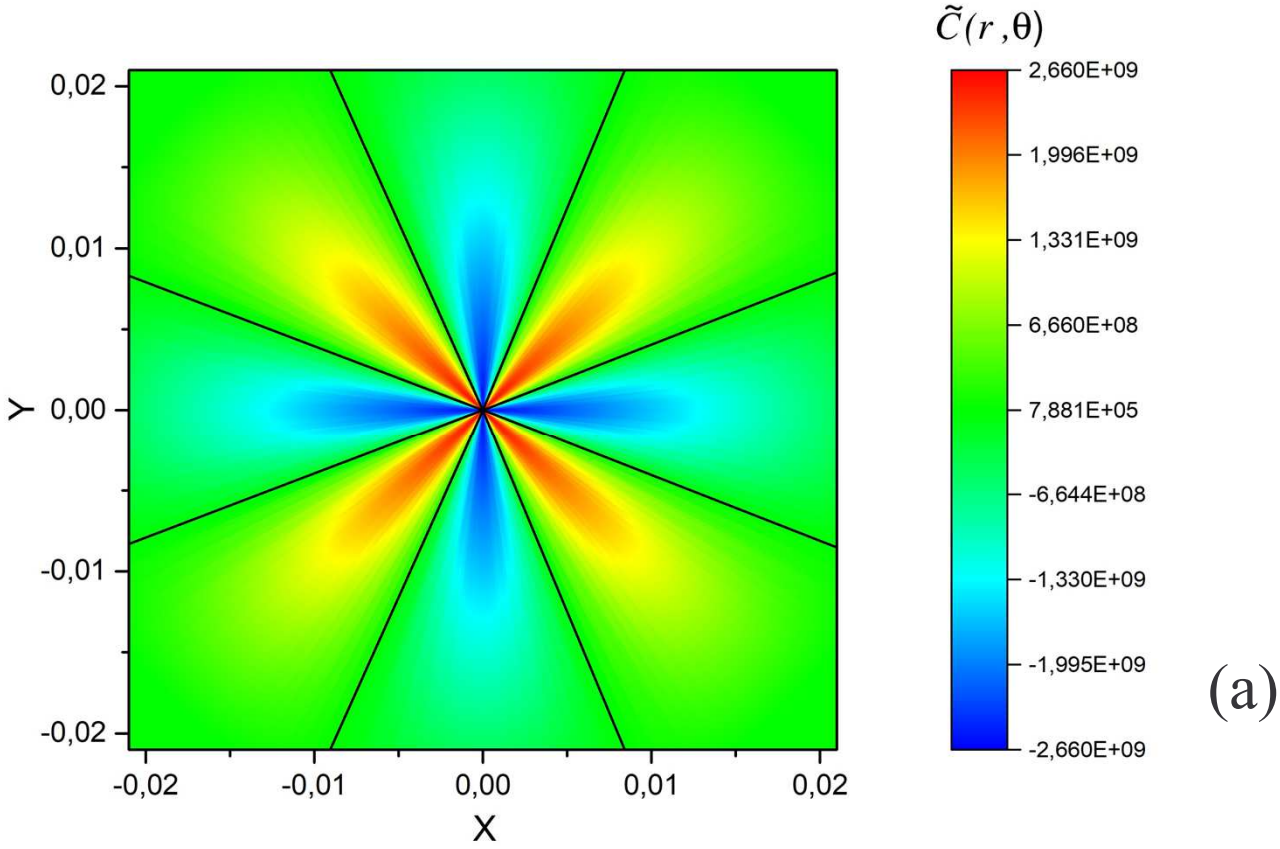

$|\tilde{C}(\tilde{r},\theta)|$
10^5
10^3
10^1
10^{-1}
10^{-3}
$c_T t$
$c_L t$
$\sim 1/r^2$
$\sim 1/r^4$
$|\tilde{C}(\tilde{r},\theta)|$
10^1
10^{-1}
0
1
2
$\tilde{r}$
$\theta=0$
(a)
10^{-2}
10^{-1}
10^0
$\tilde{r}$
$\theta=\pi/4$
(b)
$\theta=\pi/8$
(c)

Fig. 1a-c.

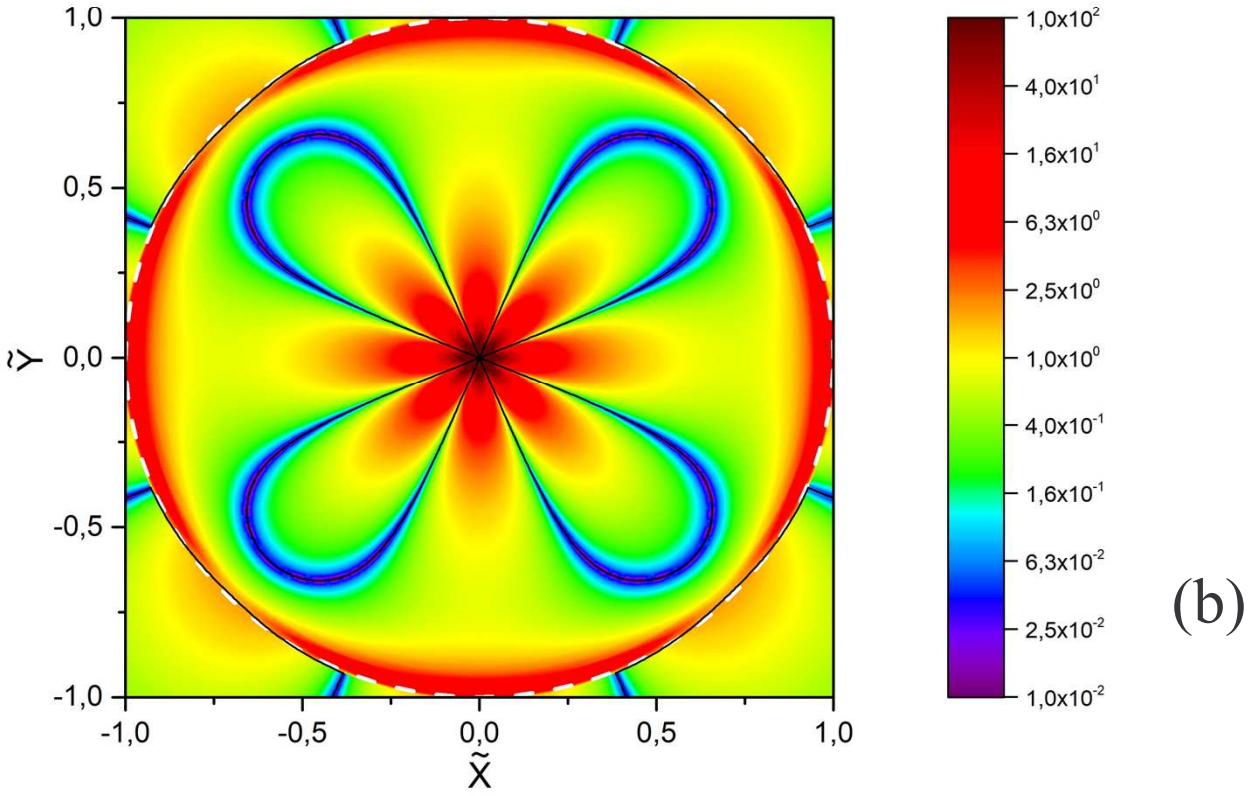

$\tilde{C} < 0, \theta = 0$
$\tilde{C} > 0, \theta = \pi/4$
$\tilde{C} < 0, \theta = \pi/4$
100
10
1
0.01
$|\tilde{C}|$
0
0.5
1
1.5
2
2.5
3
$\tilde{r}$

(d)

Fig. 1d.

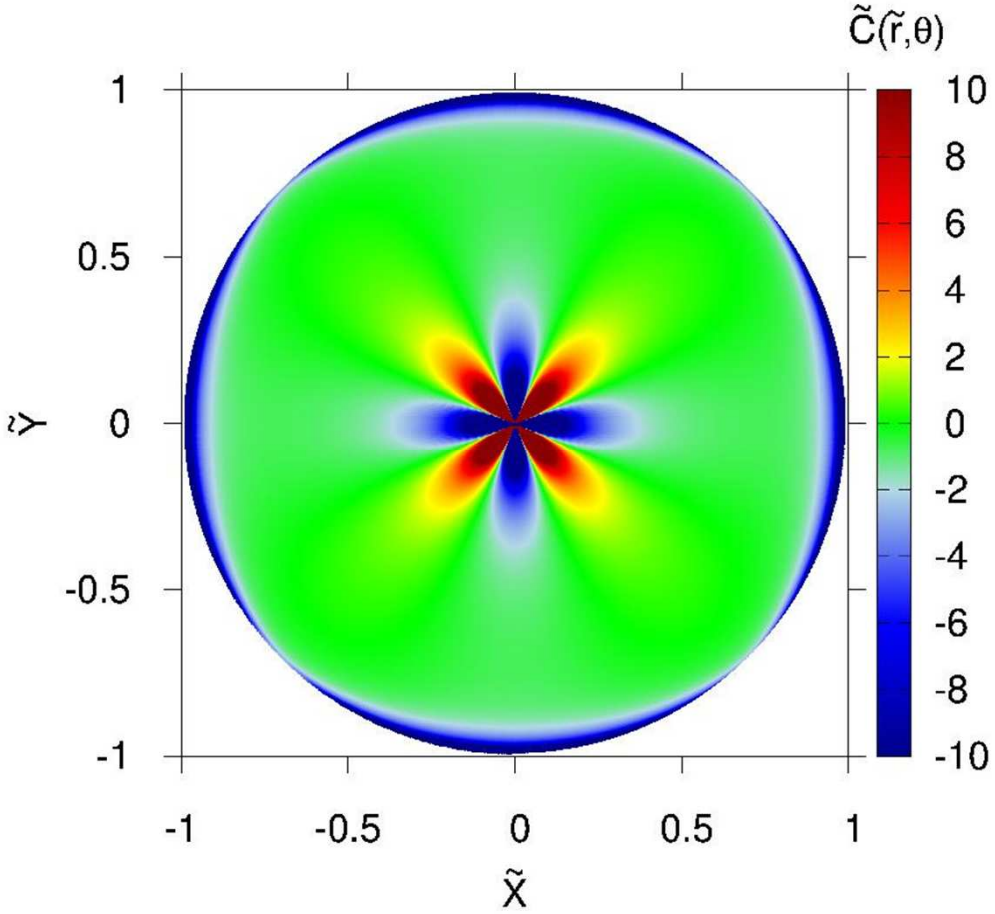

$\tilde{C}(r,\theta)$
2,660E+09
1,996E+09
1,331E+09
6,660E+08
7,881E+05
-6,644E+08
-1,330E+09
-1,995E+09
-2,660E+09
0,02
0,01
0,00
-0,01
-0,02
Y
-0,02
-0,01
0,00
0,01
0,02
X

(a)

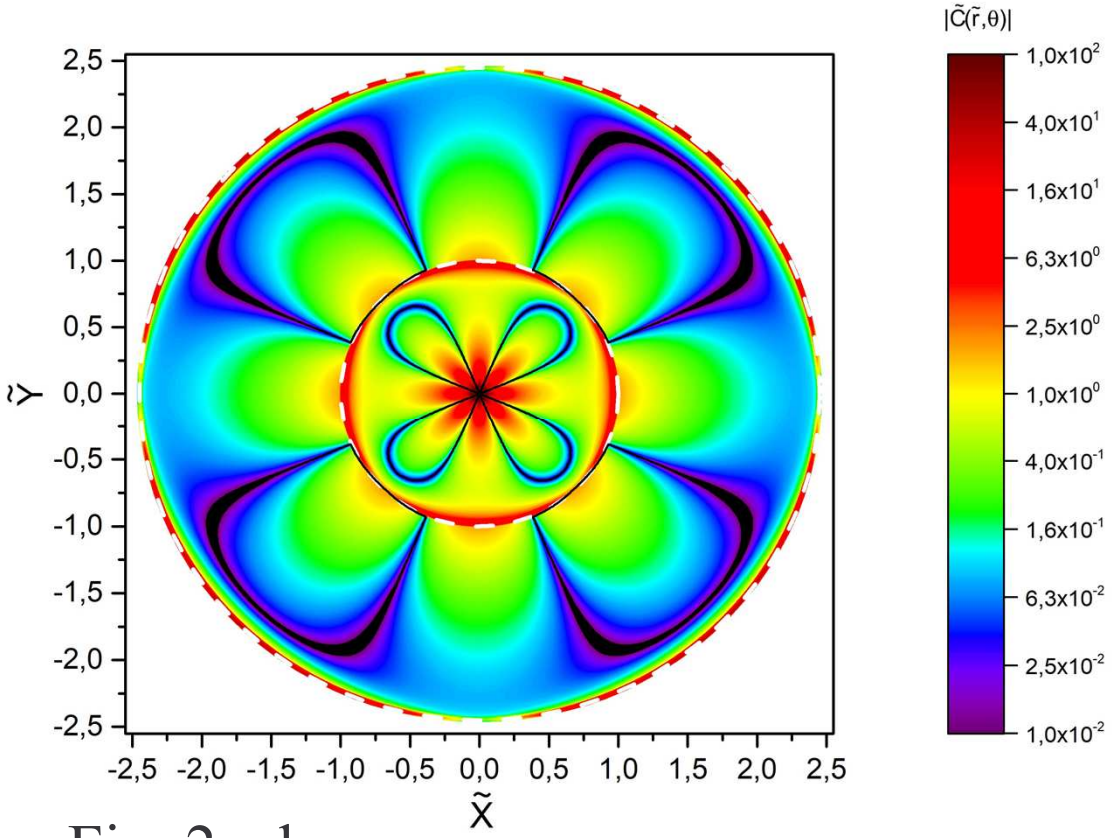

1,0x10^2
4,0x10^1
1,6x10^1
6,3x10^0
2,5x10^0
1,0x10^0
4,0x10^-1
1,6x10^-1
6,3x10^-2
2,5x10^-2
1,0x10^-2
1,0
0,5
0,0
-0,5
-1,0
Ỹ
-1,0
-0,5
0,0
0,5
1,0
X̃

(b)

Fig. 2a,b.


C̃(r̃,θ)
10
8
6
4
2
0
-2
-4
-6
-8
-10
1
0.5
0
-0.5
-1
Ỹ
-1
-0.5
0
0.5
1
X̃

(c)


|C̃(r̃,θ)|
1,0x10²
4,0x10¹
1,6x10¹
6,3x10⁰
2,5x10⁰
1,0x10⁰
4,0x10⁻¹
1,6x10⁻¹
6,3x10⁻²
2,5x10⁻²
1,0x10⁻²
2,5
2,0
1,5
1,0
0,5
0,0
-0,5
-1,0
-1,5
-2,0
-2,5
Ỹ
-2,5 -2,0 -1,5 -1,0 -0,5 0,0 0,5 1,0 1,5 2,0 2,5
X̃

(d)

Fig. 2c,d.