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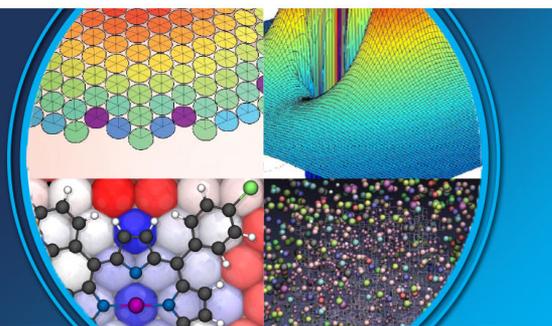
Stresses in non-equilibrium fluids: Exact formulation and coarse-grained theory

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Starting from the stochastic equation for the density operator, we formulate the exact (instantaneous) stress tensor for interacting Brownian particles and show that its average value agrees with expressions derived previously. We analyze the relation between the stress tensor and forces due to external potentials and observe that, out of equilibrium, particle currents give rise to extra forces. Next, we derive the stress tensor for a Landau-Ginzburg theory in generic, non-equilibrium situations, finding an expression analogous to that of the exact microscopic stress tensor, and discuss the computation of out-of-equilibrium (classical) Casimir forces. Subsequently, we give a general form for the stress tensor which is valid for a large variety of energy functionals and which reproduces the two mentioned cases. We then use these relations to study the spatio-temporal correlations of the stress tensor in a Brownian fluid, which we compute to leading order in the interaction potential strength. We observe that, after integration over time, the spatial correlations generally decay as power laws in space. These are expected to be of importance for driven confined systems. We also show that divergence-free parts of the stress tensor do not contribute to the Green-Kubo relation for the viscosity. *Published by AIP Publishing.* https://doi.org/10.1063/1.5019424

I. INTRODUCTION

The physics of out-of-equilibrium fluids is fascinating in its complexity and the great variety of phenomena they exhibit. This is in part due to the many ways a system can be forced out of equilibrium, e.g., by a continuous external driving such as an applied shear,[1,2] or an imposed temperature gradient,[3] or by a (sudden) change of a control parameter as in a temperature quench producing a supercooled liquid (see, e.g., Ref. 4).

In general, describing the static properties of fluids in thermal equilibrium is already a formidable task due to the inherent many-body interactions.[5] In this respect, density functional theory (DFT)[6,7] has been very successful in the study of static density profiles and correlation functions. Concerning the dynamics (or non-equilibrium situations), insights have been obtained in the dilute limit from exactly solvable two-body models,[1,8–10] e.g., from the Boltzmann or the Smoluchowski equation. For multi-body dynamics, approximate treatments include mode coupling theory[11,12] or dynamical density functional theory (DDFT),[13–15] including power functional theory.[16] Non-equilibrium molecular fluids have also been studied extensively by computer simulations.[17–20]

While the above-mentioned approaches start from the microscopic details of the fluid (at the particle level), a very different approach starts from effective field theories (such as Landau-Ginzburg theory[21–23]), where microscopic details are neglected in order to study only the large-scale phenomena, notably in near-critical fluids. Non-equilibrium scenarios also yield insights and challenges in this context: Several computations have been concerned with critical Casimir forces away from equilibrium, i.e., for temperature quenches,[24–26] moving objects,[27–29] or shear in confinement.[30] Non-equilibrium fluctuations arising from conservation laws have also been demonstrated to lead to Casimir forces in various setups.[31–39] Moreover, the pressure and stresses exerted by active systems have attracted growing attention for their unusual properties.[40,41] In all these studies, the computation of forces and stresses in out-of-equilibrium situations is non-trivial, and the form of the applicable stress tensor has been discussed (partly controversially).[26,42,43]

In this manuscript, we derive several expressions for the stress tensor of a liquid. In particular, starting from the microscopic dynamics of interacting Brownian particles, we compute the exact stress tensor for any given (snapshot) density realization. Importantly, the stress tensor's average agrees with the form obtained from the Smoluchowski equation.[44–46] Before averaging, the fluctuating form is naturally well suited

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for use in Green-Kubo fluctuation relations.[47,48] As a second step, we derive a non-equilibrium version of the stress tensor for a Landau-Ginzburg Hamiltonian and identify the various contributions, which we relate to those found in the exact microscopic version. Using the (non-equilibrium) stress tensor to compute the force on an embedded object, the results are in agreement with the existing literature.[26] We then use the insights obtained from the analysis of Brownian particles and Landau-Ginzburg theory and derive a form of the stress tensor which is valid for a general energy functional, in particular reproducing the two aforementioned cases. As an application, we use the expression for the instantaneous stress tensor to compute temporal and spatial stress correlations; this can be done exactly to leading order in the interaction potential V. We show that, after integration over time, the stress-stress correlations decay as power laws in space. These correlations have been of recent interest[49] (see also Ref. [50]) and are expected to influence the flow in confinement[51] or for inhomogeneous flow velocities.[52] Finally, we use the Green-Kubo formula to deduce the viscosity of the liquid from the stress correlations.

The manuscript is organized as follows: Sec. II introduces the system under consideration and defines several observables of interest. Section III gives the exact instantaneous stress tensor for the density operator, while Sec. IV analyzes the stresses in a standard local field theory (with only zeroth and first derivative terms). Section V derives an expression for the stress tensor for a general energy functional. Section VI provides an application: We compute the correlations of the stress tensor.

II. THE SYSTEM AND OBSERVABLES

A. System

To investigate non-equilibrium liquids, we choose the well-studied and experimentally relevant model system of over-damped spherical Brownian particles. This system has the advantage that, even if driven far from equilibrium, the solvent stays equilibrated in any situation and acts as a bath at the given temperature. This way, a well-characterized out-of-equilibrium state is obtained.

Using Brownian dynamics directly implies a canonical or grand-canonical description, where the solvent acts as a bath at the given temperature. We generally consider systems for which canonical and grand-canonical descriptions are equivalent due to a very large (infinite) particle number (for example, a semi-infinite system bound by a planar surface).

The Brownian particles with positions at $\mathbf{x}_\mu$ are subject to a potential $\Psi(\{\mathbf{x}_\mu\})$ that includes pairwise interactions (denoted by V) as well as an external potential (denoted U),

$$\Psi(\{\mathbf{x}_\mu\}) = \sum_{\mu<\nu} V(\mathbf{x}_\mu - \mathbf{x}_\nu) + \sum_\mu U(\mathbf{x}_\mu). \qquad (1)$$

Indices μ and ν run over all particles. The thermal energy scale is denoted by $k_B T \equiv \beta^{-1}$, with Boltzmann's constant k_B and the (solvent-imposed) temperature T. The bare diffusivity (in the absence of interactions) of the Brownian particles is denoted by D. Each particle thus obeys the over-damped Langevin equation

$$\frac{d\mathbf{x}_\mu}{dt} = D\beta\mathbf{F}_\mu + \sqrt{2D}\boldsymbol{\xi}_\mu, \qquad (2)$$

where $\boldsymbol{\xi}_\mu$ is a Gaussian white noise with zero mean and with correlations $\langle \xi_{\mu,i}(t)\xi_{\nu,j}(t') \rangle = \delta_{\mu\nu}\delta_{ij}\delta(t - t')$, and $\mathbf{F}_\mu = -\nabla_{\mathbf{x}_\mu}\Psi$ is the force acting on particle μ due to the potential Ψ. (Throughout, i and j label spatial components, while Greek indices label particles.) If $\Psi = 0$, each particle performs isotropic Brownian motion.

Equation (2) is a standard theoretical model, but it neglects hydrodynamic interactions in comparison with the experimental situation of Brownian suspensions so that it is sometimes referred to as the free draining limit.

B. Observables—Mean and fluctuating—In and out of equilibrium

We summarize the important observables for this manuscript in Table I. The basic quantity is the density operator,[5]

$$\rho(\mathbf{x}, t) = \sum_\mu \delta(\mathbf{x} - \mathbf{x}_\mu(t)). \qquad (3)$$

It is the starting point for all considerations that follow. Averaged over the equilibrium distribution, one obtains the mean equilibrium density $\bar\rho$, defined as

$$\bar\rho(\mathbf{x}) \equiv \langle \rho(\mathbf{x}) \rangle^{\text{eq}} = \left\langle \sum_\mu \delta(\mathbf{x} - \mathbf{x}_\mu) \right\rangle^{\text{eq}}. \qquad (4)$$

Here we have introduced the *equilibrium* average $\langle \ldots \rangle^{\text{eq}}$, which, for the over-damped system, is exactly given by

$$\langle \ldots \rangle^{\text{eq}} = \frac{\int d\Gamma \ldots e^{-\beta\Psi(\Gamma)}}{\int d\Gamma e^{-\beta\Psi(\Gamma)}}, \qquad (5)$$

where $\Gamma \equiv \{\mathbf{x}_\mu\}$. As noted above, for large systems the grand canonical average agrees with the canonical one given here. We introduce the density fluctuation field $\phi(\mathbf{x}, t)$, which quantifies the deviation of the density operator from its equilibrium mean

$$\phi(\mathbf{x}, t) = \rho(\mathbf{x}, t) - \bar\rho(\mathbf{x}). \qquad (6)$$

Fluctuations can be characterized by their two-point correlation function

$$C(\mathbf{x}, \mathbf{x}', t, t') = \langle \phi(\mathbf{x}, t)\phi(\mathbf{x}', t') \rangle, \qquad (7)$$

TABLE I. Observables relevant for this manuscript. Note that the mean density, the two-body density, and the pair correlation can be evaluated both in and out of equilibrium.

Symbol	Meaning
$\rho(\mathbf{x}, t)$	Density operator: $\rho(\mathbf{x}, t) = \sum_\mu \delta(\mathbf{x} - \mathbf{x}_\mu(t))$
$\bar\rho(\mathbf{x}) = \langle \rho(\mathbf{x}) \rangle^{\text{eq}}$	Mean density in equilibrium
$\phi(\mathbf{x}, t)$	Fluctuation of density about its equilibrium value, $\phi(\mathbf{x}, t) = \rho(\mathbf{x}, t) - \bar\rho(\mathbf{x})$
$\langle \phi(\mathbf{x}, t)\phi(\mathbf{x}', t') \rangle$	Time-dependent correlations of density fluctuations
$\langle \rho(\mathbf{x}, t) \rangle$	Mean density
$\rho^{(2)}(\mathbf{x}, \mathbf{x}')$	Averaged two-body density: $\rho^{(2)}(\mathbf{x}, \mathbf{x}') = \left\langle \sum_{\mu\neq\nu} \delta(\mathbf{x} - \mathbf{x}_\mu)\delta(\mathbf{x}' - \mathbf{x}_\nu) \right\rangle$
$g(\mathbf{r})$	Pair correlation function in bulk: $g(\mathbf{r}) = \frac{1}{N\langle\rho\rangle} \left\langle \sum_{\nu\neq\mu} \delta(\mathbf{r} - \mathbf{x}_\nu + \mathbf{x}_\mu) \right\rangle$

where we introduced the average $\langle \ldots \rangle$ over noise realizations given a (possibly non-equilibrium) ensemble of initial conditions. The correlation function in Eq. (7) is thus well-defined in or out of equilibrium. In stationary state, C is a function of $t - t'$ only[53] and depends only on the relative coordinate $\mathbf{x} - \mathbf{x}'$ in homogeneous systems. Its spatial Fourier transform, $\tilde{C}$ is the intermediate scattering function.[1]

Another important quantity (related to C) is the two-body density, defined by[5]

$$\rho^{(2)}(\mathbf{x}, \mathbf{x}', t) = \left\langle \sum_{\nu \neq \mu} \delta(\mathbf{x} - \mathbf{x}_\mu(t)) \delta(\mathbf{x}' - \mathbf{x}_\nu(t)) \right\rangle. \quad (8)$$

For bulk systems, $\rho^{(2)}$ depends only on one coordinate $\mathbf{r}$ (see Ref. 5) and can be expressed via the pair correlation function g,

$$\begin{aligned}
g(\mathbf{r}) &= \frac{1}{\langle \rho \rangle^2} \rho^{(2)}(\mathbf{x} + \mathbf{r}, \mathbf{x}) \\
&= \frac{1}{N\langle \rho \rangle} \left\langle \sum_{\mu \neq \nu} \delta(\mathbf{r} + \mathbf{x}_\mu - \mathbf{x}_\nu) \right\rangle. \quad (9)
\end{aligned}$$

Again, the above average for g can be evaluated in or out of equilibrium. One example for a non-equilibrium pair correlation function is found in systems under shear, where g is distorted compared with that of the reference equilibrium case.[1]

III. EXACT MICROSCOPIC STRESS TENSOR

In this section, we derive and discuss the exact microscopic stress tensor (including its fluctuations) for the system of Brownian particles.

A. Microscopic theory

The stress tensor is related to forces in the system. These forces can be read off directly on the equation of motion, which for the density operator ρ is given by[54]

$$\frac{\partial \rho}{\partial t}(\mathbf{x}, t) = \nabla \cdot \left[D\rho(\mathbf{x}, t) \nabla \frac{\delta \beta \mathcal{E}}{\delta \rho(\mathbf{x}, t)} + \sqrt{2D\rho(\mathbf{x}, t)} \boldsymbol{\eta}(\mathbf{x}, t) \right]. \quad (10)$$

Equation (10), interpreted with the Itō convention, is an exact reformulation of Eq. (2). The term $\boldsymbol{\eta}$ is a vectorial Gaussian white noise field with zero mean and correlations

$$\left\langle \eta_i(\mathbf{x}, t) \eta_j(\mathbf{x}', t') \right\rangle = \delta_{ij} \delta(\mathbf{x} - \mathbf{x}') \delta(t - t'). \quad (11)$$

$\mathcal{E}$ is the (free) energy functional, which contains an ideal gas part, a contribution from interactions via the inter-particle potential V, and the external potential U,

$$\begin{aligned}
\mathcal{E}[\rho(\mathbf{x})] = {}& k_B T \int d\mathbf{x} \rho(\mathbf{x}) \ln(\rho(\mathbf{x})) \\
&+ \frac{1}{2} \int d\mathbf{x} d\mathbf{x}' \rho(\mathbf{x}) V(\mathbf{x} - \mathbf{x}') \rho(\mathbf{x}') \\
&+ \int d\mathbf{x} \rho(\mathbf{x}) U(\mathbf{x}). \quad (12)
\end{aligned}$$

The reader should note that $\mathcal{E}$ is not the free energy functional of DFT.[6] Indeed, one important difference of this work with respect to DDFT[13–15] is the presence of the noise in Eq. (10). The stress tensor for DDFT was discussed in Ref. 46.

Equation (10) may now be rewritten for identification of the stress tensor $\boldsymbol{\sigma}$: The divergence of the stress tensor appears directly,[45,46]

$$\begin{aligned}
\frac{\partial \rho}{\partial t}(\mathbf{x}, t) = {}& -\beta D \nabla \cdot [\nabla \cdot \boldsymbol{\sigma}(\mathbf{x}, t) - \rho(\mathbf{x}, t) \nabla U(\mathbf{x})] \\
&+ \nabla \cdot \left[\sqrt{2D\rho(\mathbf{x}, t)} \boldsymbol{\eta}(\mathbf{x}, t) \right]. \quad (13)
\end{aligned}$$

Equation (13) is a force balance between external and interparticle forces, where the latter are expressed via the stress tensor. The divergence of the stress tensor is thus identified by comparing Eqs. (13) and (10). Using Eq. (12), we obtain

$$\nabla \cdot \boldsymbol{\sigma}(\mathbf{x}) = -k_B T \nabla \rho(\mathbf{x}) - \rho(\mathbf{x}) \nabla \int d\mathbf{x}' V(\mathbf{x} - \mathbf{x}') \rho(\mathbf{x}'). \quad (14)$$

The divergence of the stress tensor at position $\mathbf{x}$ thus has a local entropic or osmotic contribution involving the density at $\mathbf{x}$, and an interaction term which involves the potential V. We emphasize that Eq. (14) gives the instantaneous stress tensor, which is valid for any given (snapshot) configuration of particles.

The noise-averaged form of Eq. (14) can be rewritten using the two-body density of Eq. (8), which can also be expressed via

$$\langle \rho(\mathbf{x}) \rho(\mathbf{x}') \rangle = \langle \rho(\mathbf{x}) \rangle \delta(\mathbf{x} - \mathbf{x}') + \rho^{(2)}(\mathbf{x}, \mathbf{x}'). \quad (15)$$

Noting that the first term on the rhs of Eq. (15) does not contribute to Eq. (14) (reflecting the fact that a particle cannot exert a force on itself), we obtain the familiar form for the divergence of $\boldsymbol{\sigma}$,[45]

$$\begin{aligned}
\langle \nabla \cdot \boldsymbol{\sigma}(\mathbf{x}) \rangle = {}& -k_B T \nabla \langle \rho(\mathbf{x}) \rangle \\
&- \int d\mathbf{x}' [\nabla V(\mathbf{x} - \mathbf{x}')] \rho^{(2)}(\mathbf{x}, \mathbf{x}'). \quad (16)
\end{aligned}$$

This expression may, for example, be found starting from the Smoluchowski equation.[46] The expression for the stress tensor itself, both instantaneous and averaged, can be obtained from Eqs. (14) and (16), respectively. Indeed, we show in Appendix A that for a spherical potential $V(\mathbf{r}) = V(r)$ with $r = |\mathbf{r}|$, the following expression of $\boldsymbol{\sigma}$ leads to the correct force balance,

$$\begin{aligned}
\boldsymbol{\sigma}(\mathbf{x}) = {}& -k_B T \rho(\mathbf{x}) \mathbf{I} + \frac{1}{2} \int_0^1 d\lambda \int d\mathbf{r} \\
&\times \frac{\mathbf{rr}}{r} V'(r) \rho(\mathbf{x} + (1 - \lambda)\mathbf{r}) \rho(\mathbf{x} - \lambda\mathbf{r}). \quad (17)
\end{aligned}$$

Averaging Eq. (17), we find for the mean stress tensor

$$\begin{aligned}
\langle \boldsymbol{\sigma}(\mathbf{x}) \rangle = {}& -k_B T \langle \rho(\mathbf{x}) \rangle \mathbf{I} + \frac{1}{2} \int_0^1 d\lambda \int d\mathbf{r} \\
&\times \frac{\mathbf{rr}}{r} V'(r) \rho^{(2)}(\mathbf{x} + (1 - \lambda)\mathbf{r}, \mathbf{x} - \lambda\mathbf{r}), \quad (18)
\end{aligned}$$

which is the celebrated Irving-Kirkwood formula[44] for the stress tensor; Eq. (17) extends it to individual microscopic configurations. Note that adding a divergence-free term to the stress tensor does not change the force balance in Eq. (13) so that different expressions of the stress tensor are acceptable. However, the expression of Eq. (18) can be argued to possess the most physical symmetries.[55]

B. Stress tensor for the field ϕ

For further use, we also give the form of the stress tensor in terms of the fluctuating field ϕ, such that $\rho = \bar{\rho} + \phi$. From Eq. (17), this directly gives

$$
\sigma(\mathbf{x}) = -k_B T \left[\bar{\rho}(\mathbf{x}) + \phi(\mathbf{x}) \right] \mathbf{I} + \frac{1}{2} \int_0^1 d\lambda \int d\mathbf{r}
$$
$$
\times \frac{\mathbf{rr}}{r} V'(r) \left[\bar{\rho} + \phi \right] (\mathbf{x} + (1 - \lambda)\mathbf{r}) \left[\bar{\rho} + \phi \right] (\mathbf{x} - \lambda\mathbf{r}).
\tag{19}
$$

If the correlations of the field ϕ are known (e.g., assuming Gaussian fluctuations[56]), Eq. (19) can then be used to compute the correlations of the stress tensor and the viscosity via Green-Kubo relations.

Equation (19) can be simplified further to a form which displays clearly the off-diagonal components, and which will be useful for computing shear viscosity. Up to a divergence-free term, one gets for bulk systems, where $\bar{\rho}$ is not a function of $\mathbf{x}$, that [this may be directly inferred from Eq. (14)]

$$
\sigma(\mathbf{x}) = - \left(k_B T \phi(\mathbf{x}) + \bar{\rho} \int d\mathbf{x}' \ V(\mathbf{x} - \mathbf{x}')\phi(\mathbf{x}') \right) \mathbf{I}
$$
$$
+ \frac{1}{2} \int_0^1 d\lambda \int d\mathbf{r} \frac{\mathbf{rr}}{r} V'(r)\phi(\mathbf{x} + (1 - \lambda)\mathbf{r})\phi(\mathbf{x} - \lambda\mathbf{r}).
\tag{20}
$$

The above equation shows that only the terms quadratic in ϕ contribute to the off-diagonal components of the stress tensor in bulk (and thus, as we will see later, to the viscosity). While Eq. (20) is useful for computation of off-diagonal elements, care should be taken for the diagonal term in this representation, because the integral over the potential may become arbitrarily large [Eq. (20) omits diagonal terms due to $\bar{\rho}$, which are necessary to regularize the pair correlation for large V].

C. The different terms in the force balance

The terms in Eq. (13) are interpreted physically as force densities. There is the force (density) acting on the external potential U (for example, the force acting on a wall which bounds the fluid),

$$
\mathbf{f}^{(U)}(\mathbf{x}) = \rho(\mathbf{x})\nabla U(\mathbf{x}).
\tag{21}
$$

In equilibrium, there is no net particle current in the system so that the external force balances on average the divergence of σ

$$
\langle \mathbf{f}^{(U)}(\mathbf{x}) \rangle^{\text{eq}} = \langle \nabla \cdot \sigma(\mathbf{x}) \rangle^{\text{eq}}.
\tag{22}
$$

Equation (22) reflects the well-known fact that, in equilibrium, the stress tensor is directly related to the force acting on walls or embedded objects. Out of equilibrium, the mismatch of $\mathbf{f}^{(U)}$ and $\nabla \cdot \sigma$ gives rise to particle currents.

Taking the time derivative of the density operator in Eq. (3), one obtains

$$
\frac{\partial \rho}{\partial t}(\mathbf{x}, t) = -\nabla \cdot \left[\sum_\mu \mathbf{v}_\mu(t)\delta(\mathbf{x} - \mathbf{x}_\mu(t)) \right],
\tag{23}
$$

where $\mathbf{v}_\mu = d\mathbf{x}_\mu/dt$. The instantaneous current is thus identified to be

$$
\mathbf{j}(\mathbf{x}, t) = \sum_\mu \mathbf{v}_\mu(t)\delta(\mathbf{x} - \mathbf{x}_\mu(t)).
\tag{24}
$$

Comparing Eqs. (23) and (13), we obtain for the mean current

$$
\frac{k_B T}{D} \langle \mathbf{j} \rangle \equiv \langle \mathbf{f}^{(\mathbf{j})} \rangle = \langle \nabla \cdot \sigma \rangle - \langle \mathbf{f}^{(U)} \rangle.
\tag{25}
$$

Equation (25) shows that $\nabla \cdot \sigma$ and $\mathbf{f}^{(U)}$ balance the frictional force $\mathbf{f}^{(\mathbf{j})}$ of that current.[1] Equation (25) has yet another important consequence: Out of equilibrium, the stress tensor σ may not generally be used to compute forces on walls or embedded objects because one needs to account for the contribution of the currents.

IV. STRESS TENSOR IN LANDAU-GINZBURG THEORY

In this section, we consider the stress tensor for field theories obeying the Landau-Ginzburg Hamiltonian. In this phenomenological description, no fundamental relations for forces exist, and, indeed, the form of the stress tensor in this context has been a subject of recent debate.[26,42,43] The form for the stress derived in this section is in close analogy to the microscopic stress tensor of Eq. (17), and the found equation of motion is analogous to Eq. (25). This derivation, in direct comparison to the exact one, thus sheds light on the aforementioned debate and demonstrates how non-equilibrium forces in Landau-Ginzburg theory can be found unambiguously. It also demonstrates the main differences between computations of forces in or out of equilibrium.

Phenomenological field theories are particularly well suited to investigate large-scale generic phenomena. One may, for example, study the universal aspects of systems near critical points[21–23] or the properties of long-ranged correlations which are present due to out-of-equilibrium initial conditions as, e.g., in Ref. 38. In such scenarios, one seeks expressions independent of the microscopic details (such as the interaction potential V). We thus investigate in this section the possibility of expressing the stress tensor directly at the coarse-grained level of the field theory, based purely on the Landau-Ginzburg Hamiltonian. Let us consider the Hamiltonian for a scalar field Φ in d dimensions,

$$
H[\Phi] = \int d\mathbf{x} \left[\frac{\kappa}{2}(\nabla\Phi)^2 + \mathcal{U}(\mathbf{x}) \right] \equiv \int d\mathbf{x} \ \mathcal{H}(\mathbf{x}).
\tag{26}
$$

Although higher orders in $\nabla\Phi$ can be included based on symmetry arguments,[23] we do not consider this case here. $\mathcal{U}$ can be a general polynomial of Φ, but the simplest example of the above field theory is the Gaussian case where $\mathcal{U}(\mathbf{x}) = m(\mathbf{x})\Phi(\mathbf{x})^2/2$. Here, $m(\mathbf{x})$ can be a function of position so that it may include contributions from external potentials. For bulk, with m constant in space, the correlation length is then set by $\sqrt{\kappa/m}$.[23]

As mentioned, at the level of effective field theories such as Eq. (26), the definition of mechanical quantities is not obvious. Indeed, one needs to define the nature of the field Φ and specify whether it corresponds to a matter field (for instance, a particle density, a spin density, or local charge density) or a potential field (for instance, the local electrostatic potential or chemical potential). If the field Φ is in an arbitrary non-equilibrium configuration, in the mechanical sense, then there are locally unbalanced body forces in the system. We proceed by applying a fictitious external field so that the system is in local mechanical equilibrium. The force exerted by

this fictitious field on a volume therefore cancels out exactly the local body forces generated by the internal interactions in the system. Concretely, we apply an external field h which shifts the overall energy (Hamiltonian) of Φ to

$$H_h[\Phi] = H[\Phi] + \int d\mathbf{x}\, h(\mathbf{x})\Phi(\mathbf{x}). \qquad (27)$$

Next, for a given configuration of Φ, the external field is chosen to ensure local mechanical equilibrium of the given configuration when the field is applied, i.e., $\frac{\delta H_h}{\delta \Phi(\mathbf{x})} = 0$. The required field h is thus

$$h(\mathbf{x}) = -\frac{\delta H[\Phi]}{\delta \Phi(\mathbf{x})}, \qquad (28)$$

from which we can extract the force density $\mathbf{f}_h$ acting on the sub-volume V due to the imposed field, which follows from its fundamental definition from the Hamiltonian H_h (see e.g., Ref. 26),

$$\mathbf{f}_h(\mathbf{x}) = -\Phi(\mathbf{x})\nabla h(\mathbf{x}) = \Phi(\mathbf{x})\nabla \frac{\delta H[\Phi]}{\delta \Phi(\mathbf{x})}. \qquad (29)$$

If Φ is a density field, the force density may be viewed intuitively as the product of the density and the gradient of the chemical potential associated with $H[\Phi]$.

Because the force due to h balances the force density that is not due to h, we obtain for the (body) force density $\mathbf{f}^{(\mathbf{j})}$ when $h = 0$:

$$\mathbf{f}^{(\mathbf{j})}(\mathbf{x}) = -\Phi(\mathbf{x})\nabla \frac{\delta H[\Phi]}{\delta \Phi(\mathbf{x})}. \qquad (30)$$

We added the superscript $\mathbf{j}$ in order to emphasize the similar nature of the forces in Eqs. (30) and (25), which will become more apparent below.

We note that Eq. (30) is exactly the force density in Eq. (10), when one makes the identification $\Phi \equiv \rho$ and $H = \mathcal{E}$. It is important to note that if the field theory is written down in terms of the fluctuations of the field Φ about its average value $\overline{\Phi}$, as $\Phi = \overline{\Phi} + \phi$, and the Hamiltonian for the fluctuations is $H_f[\phi]$, the local body force is given by

$$\mathbf{f}^{(\mathbf{j})}(\mathbf{x}) = -\left[\overline{\Phi}(\mathbf{x}) + \phi(\mathbf{x})\right]\nabla \frac{\delta H_f}{\delta \phi(\mathbf{x})}. \qquad (31)$$

In the limit of small fluctuations the above can be used to derive an approximate model B dynamics for interacting particle systems,[57–60] where only linear terms in ϕ are kept, yielding

$$\mathbf{f}^{(\mathbf{j})}(\mathbf{x}) = -\overline{\Phi}(\mathbf{x})\nabla\left[\int d\mathbf{x}'\, \frac{\delta^2 H_f}{\delta\phi(\mathbf{x})\delta\phi(\mathbf{x}')}\bigg|_{\phi=0}\phi(\mathbf{x}')\right]. \qquad (32)$$

Next, we identify the force on the external potential, as in Eq. (21). Here, the force density $\mathbf{f}^{(U)}$ is identified by the change of $\mathcal{H}$ under a small displacement of the potential or, equivalently, a small displacement of the object giving rise to the potential U.[26] Defining the vector $\mathbf{X}$ from the origin to a randomly chosen point on the object, we have

$$\mathbf{f}^{(U)} = -\nabla_X \mathcal{H}. \qquad (33)$$

Manipulating the partial and functional derivatives as detailed in Appendix B, we obtain the relation between the force density in Eqs. (30) and (33),

$$\mathbf{f}^{(\mathbf{j})} = \nabla \cdot \mathbf{T} - \mathbf{f}^{(U)}, \qquad (34)$$

where we have introduced the stress tensor $\mathbf{T}$,

$$T_{ij}(\mathbf{x}) = \delta_{ij}\left(\mathcal{H}(\mathbf{x}) - \Phi(\mathbf{x})\frac{\delta H}{\delta\Phi(\mathbf{x})}\right) - \frac{\partial\mathcal{H}}{\partial\nabla_j\Phi(\mathbf{x})}\nabla_i\Phi(\mathbf{x}). \qquad (35)$$

Equation (35) is the main result of this section. (In this section, we use $\mathbf{T}$ instead of $\boldsymbol{\sigma}$, following the usual field theory notation.) The structure of Eq. (34) is identical to that in Eq. (25) so that the field theory stress tensor from Eq. (35) is on the same footing as the microscopic one. As mentioned below Eq. (25), the computation of forces on external objects, $\mathbf{f}^{(U)}$, is non-trivial out of equilibrium because of the term involving the current in Eq. (34). This will be relevant, for example, when computing Casimir forces between walls or objects. As a crosscheck, we show in Appendix B that Eq. (34) gives the same value for $\mathbf{f}^{(U)}$ as the expression previously derived in Eq. (18) of Ref. 26. A particular case worth mentioning is that of no-flux boundary conditions, where the surface normal component of $\mathbf{f}^{(\mathbf{j})}$ is forced to vanish. For the geometry of two parallel plates, the force (or pressure) acting on the plates can then be computed by evaluating the stress tensor in Eq. (35) at the given surface. This was used for computation of non-equilibrium Casimir forces in Ref. 38 and tested quantitatively with simulations of interacting Brownian particles in Ref. 39.

In global equilibrium, one can show[26] that $\mathbf{f}^{(\mathbf{j})}$ vanishes on average [see Eq. (B6)], as was the case in the microscopic theory in Eq. (25). Our formulation (and the form of $\mathbf{T}$) then agrees with the commonly accepted equilibrium definition of the stress tensor $\mathbf{T}^{\mathrm{eq}}$, i.e.,

$$\langle \nabla \cdot \mathbf{T}\rangle^{\mathrm{eq}} = \langle \nabla \cdot \mathbf{T}^{\mathrm{eq}}\rangle^{\mathrm{eq}} = \langle \mathbf{f}^{(U)}\rangle^{\mathrm{eq}} \qquad (36)$$

and $\mathbf{T}^{\mathrm{eq}}$ expressed as [using Eq. (B6)][61]

$$T_{ij}^{\mathrm{eq}} = \delta_{ij}\mathcal{H} - \frac{\partial\mathcal{H}}{\partial\nabla_j\Phi}\nabla_i\Phi. \qquad (37)$$

V. GENERAL FORMULA FOR THE STRESS TENSOR

Inspired by the similarities of the formulations in Secs. III and IV, we now proceed to derive a form for the stress tensor for a more general case, which encompasses energy functionals such as Eqs. (12) and (26). To this end, we introduce a Hamiltonian for a field Φ [which, for the case of Eq. (12), takes the role of the density ρ],

$$H[\Phi] = \frac{1}{2}\int \Phi(\mathbf{x})V(\mathbf{x} - \mathbf{x}')\Phi(\mathbf{x}')d\mathbf{x}d\mathbf{x}' + \int \mathcal{U}(\Phi(\mathbf{x}), \mathbf{x})d\mathbf{x}. \qquad (38)$$

Here, the first term incorporates non-local interactions, while the second part describes local interactions, as well as external potentials. $\mathcal{U}(\Phi, \mathbf{x})$ can be any function of $\Phi(\mathbf{x})$ and $\mathbf{x}$ for the following derivation to be valid. To make connections to the previous sections, we note that the local part for Brownian particles of [Eq. (12)] is recovered with

$$\mathcal{U}(\Phi, \mathbf{x}) = k_B T\Phi \log(\Phi) + U\Phi, \qquad (39)$$

while the non-local part of Eq. (38) already takes the form of Eq. (12). On the other hand, the non-local part of the Landau-Ginzburg case [Eq. (26)] is recovered for

$$V(\mathbf{x}) = -\kappa\nabla^2\delta(\mathbf{x}), \qquad (40)$$

which, used in Eq. (38), yields $\frac{\kappa}{2}(\nabla\Phi)^2$. (Note that by "non-local" we mean any contributions giving rise to correlations of the field Φ with a non-zero correlation length in equilibrium.) This shows that the cases of Secs. III and IV are contained in Eq. (38).

In order to derive the stress tensor from Eq. (38), we again rely on the body force density of Eq. (30), which is equally valid for the Hamiltonian of Eq. (38). This yields

$$\mathbf{f}^{(\mathbf{j})}(\mathbf{x}) = -\Phi(\mathbf{x})\nabla\frac{\delta H[\Phi]}{\delta\Phi(\mathbf{x})},$$
$$= -\Phi(\mathbf{x})\nabla\int d\mathbf{x}'V(\mathbf{x}-\mathbf{x}')\Phi(\mathbf{x}') - \Phi(\mathbf{x})\nabla\frac{\partial\mathcal{U}}{\partial\Phi}. \quad (41)$$

We now use Eq. (B1) for $\mathcal{U}$,

$$\nabla_i\mathcal{U}(\Phi(\mathbf{x}),\mathbf{x}) = (\nabla_i\Phi)\frac{\partial\mathcal{U}}{\partial\Phi} - \frac{\partial\mathcal{U}}{\partial X_i} = (\nabla_i\Phi)\frac{\partial\mathcal{U}}{\partial\Phi} + f_i^{(U)}, \quad (42)$$

where, as before, $-\frac{\partial\mathcal{U}}{\partial X_i}$ is the force density in direction i acting on the object described by the coordinate $\mathbf{X}$. We now find the divergence of the stress tensor,

$$\nabla\cdot\boldsymbol{\sigma}(\mathbf{x}) = \mathbf{f}^{(\mathbf{j})}(\mathbf{x}) + \mathbf{f}^{(U)}(\mathbf{x}) \quad (43)$$
$$= -\Phi(\mathbf{x})\nabla\int d\mathbf{x}'V(\mathbf{x}-\mathbf{x}')\Phi(\mathbf{x}')$$
$$+\nabla\left[\mathcal{U} - \Phi(\mathbf{x})\frac{\partial\mathcal{U}}{\partial\Phi(\mathbf{x})}\right], \quad (44)$$

where we could equivalently have used the symbol $\mathbf{T}$ for the stress tensor, as in Sec. IV. Equation (44) shows that the local part of H yields a diagonal contribution to the stress tensor, while its non-local part further yields off-diagonal components. This also demonstrates that stresses are only propagated in the presence of a correlation length in the system. In order to invert the divergence of the non-local part, we use the steps of Appendix A. We arrive at

$$\sigma_{ij}(\mathbf{x}) = \delta_{ij}\left[\mathcal{U}(\Phi(\mathbf{x}),\mathbf{x}) - \Phi(\mathbf{x})\frac{\partial\mathcal{U}}{\partial\Phi(\mathbf{x})}\right]$$
$$+ \frac{1}{2}\int d\mathbf{r}\frac{r_i r_j}{r}V'(r)\int_0^1 d\lambda\Phi(\mathbf{x}-\lambda\mathbf{r})\Phi(\mathbf{x}+[1-\lambda]\mathbf{r}). \quad (45)$$

This is the main result of this section. To repeat, this form of the stress tensor can be used for energy functionals which contain a quadratic non-local part, and an arbitrary local part, as given in Eq. (38). As mentioned before, this result reproduces, in particular, the stress tensor for the exact description of Brownian particles, Eq. (12), as well as for the phenomenological Landau-Ginzburg theory. The two theories are thus solidly linked, and the claimed validity of Eq. (35) is underpinned. This is hopefully enlightening regarding the debate of stresses in phenomenological theories.

In order to demonstrate the above-mentioned recovery of Eqs. (17) and (35), we start with the Brownian case. Using Eq. (39) in Eq. (45) yields

$$\mathcal{U}(\Phi(\mathbf{x}),\mathbf{x}) - \Phi(\mathbf{x})\frac{\partial\mathcal{U}}{\partial\Phi(\mathbf{x})} = -k_B T\Phi(\mathbf{x}), \quad (46)$$

which is the correct result—see Eq. (17).

To reproduce Eq. (35), we use $V(\mathbf{x}) = -\kappa\nabla^2\delta(\mathbf{x})$ in Eq. (45). After some computation steps, we find

$$\frac{1}{2}\int d\mathbf{r}\frac{r_i r_j}{r}V'(r)\int_0^1 d\lambda\Phi(\mathbf{x}-\lambda\mathbf{r})\Phi(\mathbf{x}+[1-\lambda]\mathbf{r})$$
$$= \kappa\left[\frac{\delta_{ij}}{6}\left[2\Phi\nabla^2\Phi - (\nabla\Phi)^2\right] + \frac{2}{3}\Phi\nabla_i\nabla_j\Phi - \frac{1}{3}\nabla_i\Phi\nabla_j\Phi\right]. \quad (47)$$

On the other hand, evaluating Eq. (35) for $\mathcal{H} = \frac{\kappa}{2}(\nabla\Phi)^2$, one obtains

$$\kappa\left[\delta_{ij}\left(\frac{1}{2}[\nabla\Phi(\mathbf{x})]^2 + \Phi(\mathbf{x})\nabla^2\Phi(\mathbf{x})\right) - \nabla_i\Phi(\mathbf{x})\nabla_j\Phi(\mathbf{x})\right]. \quad (48)$$

While Eqs. (47) and (48) are not equal, their difference

$$\frac{2\kappa}{3}\left[\nabla_i(\Phi\nabla_j\Phi) - \delta_{ij}\nabla_k(\Phi\nabla_k\Phi)\right] \quad (49)$$

is divergence-free. This demonstrates that Eq. (45) indeed reproduces Eqs. (17) and (35). The observed difference in divergence-free terms is acceptable and has no physical consequences as regards stresses and forces. Indeed, these differences are introduced when inverting the divergence of Eq. (44), which is a not unique process.

VI. APPLICATION: STRESS CORRELATIONS OF BROWNIAN SUSPENSIONS

In this section, we make use of the expressions derived for the microscopic stress tensor Eq. (19) to compute the two-point correlations of the stress at different positions and times for a bulk equilibrium system. We provide the limit of high temperatures, which corresponds to the leading-order term in the external potential V. We also compare with the results obtained from the Gaussian field theory, using Eq. (35) to define the stress tensor.

Note that this two-point correlation function can be used, via Green-Kubo relations,[62] to compute the viscosity (in linear response to a flow field). Because of this, and because it incorporates the fluctuations around equilibrium, the correlation function is a quantity whose computation requires a form for the stress tensor which is valid out of equilibrium. [For example, the form of Eq. (37) should not be used.]

A. Diagonal components—Pressure fluctuations

We start with the diagonal part of the stress tensor in Eq. (19), denoting $\delta\boldsymbol{\sigma} = \boldsymbol{\sigma} - \langle\boldsymbol{\sigma}\rangle^{\text{eq}}$. The leading term at small V (or high T) results from the ideal contribution in Eq. (19). It reads (without summing repeated indices)

$$\lim_{\beta V\to 0}\langle\delta\sigma_{ii}(\mathbf{x},t)\delta\sigma_{kk}(\mathbf{0},0)\rangle^{\text{eq}} = (k_B T)^2\langle\phi(\mathbf{x},t)\phi(\mathbf{0},0)\rangle^{\text{eq}}. \quad (50)$$

In order to determine the leading order in V, the correlation function in Eq. (50) should be evaluated for $V = 0$, which corresponds to the ideal gas. The correlations for the ideal gas are denoted $\langle\cdot\rangle^{\text{id}}$ and are computed in Appendix C. The two-point correlation reads

$$\langle\Phi(\mathbf{x},t)\Phi(\mathbf{0},0)\rangle^{\text{id}} = \frac{\bar{\rho}}{(4\pi Dt)^{3/2}}e^{-\frac{|\mathbf{x}|^2}{4Dt}}. \quad (51)$$

We obtain

$$\lim_{\beta V \to 0} \langle \delta\sigma_{ii}(\mathbf{x}, t)\delta\sigma_{kk}(\mathbf{0}, 0)\rangle^{\text{eq}} = \frac{(k_B T)^2 \bar{\rho}}{(4\pi Dt)^{3/2}} e^{-\frac{|\mathbf{x}|^2}{4Dt}}. \quad (52)$$

We observe from Eq. (52) that the temporal correlations are due to a diffusion process. The long-ranged character of the correlations in space becomes more apparent after integrating Eq. (52) over time

$$\int_0^\infty dt \lim_{\beta V \to 0} \langle \delta\sigma_{ii}(\mathbf{x}, t)\delta\sigma_{kk}(\mathbf{0}, 0)\rangle^{\text{eq}} = \frac{(k_B T)^2 \bar{\rho}}{4\pi D|\mathbf{x}|}. \quad (53)$$

This quantity decays slowly, as the inverse of the distance. Because the time integral of such equilibrium correlations appears typically in linear response formulae, we expect the long-ranged form of Eq. (53) to be relevant for non-equilibrium perturbations.

As a final note, the equal-time fluctuations of pressure from Eq. (52) are

$$\int d\mathbf{x} \lim_{\beta V \to 0} \langle \delta\sigma_{ii}(\mathbf{x}, 0)\delta\sigma_{kk}(\mathbf{0}, 0)\rangle^{\text{eq}} = (k_B T)^2 \bar{\rho}. \quad (54)$$

These are the pressure fluctuations of an ideal gas, which have a partly controversial history, starting with Gibbs.[63]

We note that the stress tensor of Landau-Ginzburg theory, Eq. (35), together with a Gaussian Hamiltonian, Eq. (26), yields a qualitatively different result for spatial pressure correlations. The reason is that Eq. (35) yields a stress tensor which is purely quadratic in the fluctuating field Φ, resulting from the symmetry assumptions underlying the field theoretic Hamiltonian in Eq. (26).[23] In contrast to that, it is the term $\sim \rho \ln \rho$ in Eq. (12) which yields the correct ideal gas contribution.

B. Diagonal–off-diagonal correlations

As visible in Eq. (20), the off-diagonal component of the stress involves the potential V. The leading term of the correlation between diagonal and off-diagonal components of $\boldsymbol{\sigma}$ is thus linear in V. This term reads (for $i \neq j$)

$$\lim_{\beta V \to 0} \langle \delta\sigma_{ij}(\mathbf{x}, t)\delta\sigma_{kk}(\mathbf{0}, 0)\rangle^{\text{eq}} = k_B T \int_0^1 d\lambda \int d\mathbf{r}$$
$$\times \frac{r_i r_j}{r} V'(r) \langle \phi(\mathbf{x} + (1-\lambda)\mathbf{r}), t)\phi(\mathbf{x} - \lambda\mathbf{r}, t)\phi(\mathbf{0}, 0)\rangle^{\text{id}}, \quad (55)$$

where, again, the correlation of ϕ is evaluated for the ideal gas. Since the density of the ideal gas has Poissonian statistics,[64] the three-point correlator in Eq. (55) is *per se* non-zero. Using Eq. (C7), we find the correlation in terms of Fourier transforms,

$$\langle \tilde{\phi}(\mathbf{k}, t)\tilde{\phi}(\mathbf{k}', t)\tilde{\phi}(\mathbf{q}, 0)\rangle^{\text{id}} = (2\pi)^3 \bar{\rho}\delta(\mathbf{k} + \mathbf{k}' + \mathbf{q})e^{-Dq^2 t}, \quad (56)$$

where, in this paper, Fourier transforms are defined by

$$\tilde{f}(\mathbf{k}) = \int d\mathbf{x} f(\mathbf{x})e^{-i\mathbf{k}\cdot\mathbf{x}}. \quad (57)$$

While there is indeed a non-zero three-point correlation in Eq. (56), its contribution to the correlation of stresses in Eq. (55) is zero (Appendix C). The reason is that Eq. (56) results from a single Brownian particle and its self-correlations

(as is apparent from the linearity in $\bar{\rho}$), and this particle cannot exert a force upon itself. Some relations useful to derive this statement mathematically are given in Appendix C. We thus note that there is no correlation between diagonal and off-diagonal stress components at the given order in V.

C. Off-diagonal components—Shear stress fluctuations

Here we start by computing the exact leading term in V in the microscopic theory. We then also derive the shear stress fluctuations in the Gaussian Landau-Ginzburg theory.

1. Exact leading order in V

To leading order, the off-diagonal component of the stress correlator is proportional to V^2. For ease of notation, we abbreviate,

$$\lim_{\beta V \to 0} \langle \delta\sigma_{ij}(\mathbf{x}, t)\delta\sigma_{kl}(\mathbf{0}, 0)\rangle = \Sigma_{ijkl}(\mathbf{x}, t). \quad (58)$$

For $i \neq j$ and $k \neq l$, this expression reads

$$\Sigma_{ijkl}(\mathbf{x}, t) = \frac{1}{4} \int d\mathbf{r}d\mathbf{r}' \int_0^1 d\lambda d\lambda' \frac{r_i r_j}{r} V'(r) \frac{r'_k r'_l}{r'} V'(r')$$
$$\times \langle \phi(\mathbf{x} + \mathbf{r} - \lambda\mathbf{r}, t)\phi(\mathbf{x} - \lambda\mathbf{r}, t)$$
$$\times \phi(\mathbf{r}' - \lambda'\mathbf{r}', 0)\phi(-\lambda'\mathbf{r}', 0)\rangle^{\text{id}}. \quad (59)$$

In Fourier space, the correlation reads (again, see Appendix C)

$$\langle \tilde{\phi}(\mathbf{k}, t)\tilde{\phi}(\mathbf{k}', t)\tilde{\phi}(\mathbf{q}, 0)\tilde{\phi}(\mathbf{q}', 0)\rangle^{\text{id}}$$
$$= \bar{\rho}(2\pi)^3\delta(\mathbf{k} + \mathbf{k}' + \mathbf{q} + \mathbf{q}')\mathcal{C}(\mathbf{k} + \mathbf{k}', t)$$
$$+ (2\pi)^6 \bar{\rho}^2 [\delta(\mathbf{k} + \mathbf{k}')\delta(\mathbf{q} + \mathbf{q}')$$
$$+ \delta(\mathbf{k} + \mathbf{q})\delta(\mathbf{k}' + \mathbf{q}')\mathcal{C}(\mathbf{k}, t)\mathcal{C}(\mathbf{k}', t)$$
$$+ \delta(\mathbf{k} + \mathbf{q}')\delta(\mathbf{k}' + \mathbf{q})\mathcal{C}(\mathbf{k}, t)\mathcal{C}(\mathbf{k}', t)], \quad (60)$$

with $\mathcal{C}(\mathbf{k}, t) = e^{-Dk^2 t}$. The four-point correlation function in Eq. (60) contains a term linear in $\bar{\rho}$, which, as in Eq. (56), shows the non-Gaussian behavior of ideal particles. Again, this term does not contribute to the stress correlator. The last three terms in Eq. (60) are equivalent to a Gaussian decoupling, where all pairs of functions ϕ appear as in Wick's theorem. The first of these three is time independent and does not contribute to Eq. (59) by symmetry. We then find for $i \neq j$ and $k \neq l$

$$\Sigma_{ijkl}(\mathbf{x}, t) = \int \frac{d\mathbf{k}}{(2\pi)^3} e^{i\mathbf{k}\cdot\mathbf{x}} \int_0^1 d\lambda d\lambda' \frac{\bar{\rho}^2}{2} \int \frac{d\mathbf{p}}{(2\pi)^3}$$
$$\times \tilde{\mathcal{A}}_{ij}(\lambda\mathbf{k} - \mathbf{p})\tilde{\mathcal{A}}_{kl}(-\lambda'\mathbf{k} + \mathbf{p})e^{-D[\mathbf{p}^2 + (\mathbf{k} - \mathbf{p})^2]t}, \quad (61)$$

where for $i \neq j$

$$\tilde{\mathcal{A}}_{ij}(\mathbf{k}) = \frac{k_i k_j}{k} \tilde{V}'(k). \quad (62)$$

We continue by analyzing Eq. (61) in real space, which is found easily from the Gaussian decoupling of the four-point function in Eq. (59). We thus have, using Eq. (51) (again $i \neq j$),

$$\Sigma_{ijij}(\mathbf{x}, t) = \frac{1}{2} \int d\mathbf{r}d\mathbf{r}' \int_0^1 d\lambda d\lambda' \frac{r_i r_j}{r} V'(r) \frac{r'_i r'_j}{r'} V'(r')$$
$$\times \frac{\bar{\rho}^2}{(4\pi Dt)^3} e^{\frac{-|\mathbf{x} + \mathbf{r} - \lambda\mathbf{r} - \mathbf{r}' + \lambda'\mathbf{r}'|^2 - |\mathbf{x} - \lambda\mathbf{r} + \lambda'\mathbf{r}'|^2}{4Dt}}. \quad (63)$$

We now address the large distance behavior of the stress correlations. For a short-ranged potential, the values of $|\mathbf{r}|$ and $|\mathbf{r}'|$ that contribute to the above integral are constrained to that range. For values of $|\mathbf{x}|$ much larger than the interaction range, we may expand the exponential in Eq. (63) for $|\mathbf{r}| \ll |\mathbf{x}|$ and $|\mathbf{r}'| \ll |\mathbf{x}|$. Because of the symmetry of the integrands in Eq. (63), we seek the leading terms of order $r_i r_j r_i' r_j'$ of the exponential in Eq. (63). The integrals over λ and λ' can then be performed to obtain

$$\Sigma_{ijij}(\mathbf{x}, t) = \mathcal{G}[V]\bar{\rho}^2 \frac{10D^2t^2 - Dt(x_i^2 + x_j^2) + x_i^2 x_j^2}{9216(Dt)^7\pi^3} e^{-\frac{|\mathbf{x}|^2}{2Dt}}, \quad (64)$$

where we have defined the functional

$$\mathcal{G}[V] = \frac{8\pi^2}{225} \left(\int_0^\infty dr\, r^5 V'(r) \right)^2. \quad (65)$$

Here, too, we perform the time integral, which is the important quantity in Green-Kubo relations, and obtain

$$\int_0^\infty dt\, \Sigma_{ijij}(\mathbf{x}, t) = \mathcal{G}[V]\bar{\rho}^2 \frac{5 + 40\frac{x_i^2 x_j^2}{|\mathbf{x}|^4} - 4\frac{x_i^2 + x_j^2}{|\mathbf{x}|^2}}{48D\pi^3 |\mathbf{x}|^8}. \quad (66)$$

Shear stress fluctuations in Eq. (66) and pressure fluctuations in Eq. (53) thus decay as power laws in space. These observations should have implications for the rheology of suspensions in confined systems.[30,50,51]

The long-ranged property of the shear stress correlations has been noted in Ref. 49 via a Zwanzig-Mori approach. In particular, a Fourier inversion of Eq. (6) from Ref. 49 with $s = 0$ yields the analogue of $\int_0^\infty dt\, \Sigma_{ijij}(\mathbf{x}, t)$, giving contributions $\sim |\mathbf{x}|^{-3}$. These show a different power than those in Eq. (66), $\sim |\mathbf{x}|^{-8}$, which could be due to the absence of momentum conservation in our formulation. This must be investigated in the future. While the framework of DDFT has been extended beyond Brownian dynamics (e.g., including inertia[15,65]), the approach of this work may be further developed in analogy to "model H."[21]

2. From Landau-Ginzburg theory

Next we consider the off-diagonal stress correlations from Landau-Ginzburg theory, using the stress tensor **T** in Eq. (35). This, together with a Gaussian decoupling of the four-point correlation (again, $i \neq j$), gives

$$\langle T_{ij}(\mathbf{x}, t) T_{ij}(\mathbf{0}, 0) \rangle \approx \kappa \Big([\nabla_i \nabla_j C(\mathbf{x}, t)]^2$$
$$+ [\nabla_i \nabla_i C(\mathbf{x}, t)][\nabla_j \nabla_j C(\mathbf{x}, t)] \Big). \quad (67)$$

We are interested in the case where $|\mathbf{x}|$ is much larger than the correlation length. In this limit, the correlation function reads in model B dynamics[23,56] (see Appendix D for the leading order in correlation length)

$$C(\mathbf{x}, t) = \frac{k_B T}{m} \frac{1}{(4\pi \tilde{D}t)^{3/2}} e^{-\frac{|\mathbf{x}|^2}{4\tilde{D}t}}, \quad (68)$$

where m is the coefficient appearing in the Gaussian Hamiltonian in Eq. (26) of the field theory. Explicitly, m may also be expressed in terms of the small wave-vector limit of the direct correlation function $c^{(2)}$[5,56] (see Appendix D),

$$m = k_B T \left(\frac{1}{\bar{\rho}} - c_0^{(2)} \right), \quad (69)$$

which may also be related to the isothermal compressibility.[5] We have also introduced $\tilde{D} \equiv D/S_0$, with $S_0 = (1 - \bar{\rho}c_0^{(2)})^{-1} = k_B T / \bar{\rho}m$ being the small wave-vector limit of the structure factor; $\tilde{D}$ is an approximation for the diffusion coefficient,[1] which emerges when taking the small wave-vector limit of Eq. (D4). Equation (67) yields

$$\langle T_{ij}(\mathbf{x}, t) T_{ij}(\mathbf{0}, 0) \rangle$$
$$= \kappa^2 \frac{(k_B T)^2}{m^2} \frac{2\tilde{D}^2 t^2 - \tilde{D}t(x_i^2 + x_j^2) + x_i^2 x_j^2}{512(\tilde{D}t)^7\pi^3} e^{-\frac{|\mathbf{x}|^2}{2\tilde{D}t}}. \quad (70)$$

Apart from prefactors, this expression is qualitatively equal to Eq. (63): It can be shown that the dependencies on **x** and t of Eqs. (70) and (63) differ only by terms which arise from divergence-free parts of the stress tensor.

D. Viscosity

We finish with the evaluation of the shear viscosity η to leading order in the interaction potential. Using the Green-Kubo relation,[62] the viscosity is given in terms of the stress correlator ($i \neq j$),

$$\eta = \frac{\beta}{\mathcal{V}} \int_0^\infty dt \int d\mathbf{x}\, d\mathbf{x}' \langle \sigma_{ij}(\mathbf{x}, t) \sigma_{ij}(\mathbf{x}', 0) \rangle^{\text{eq}}, \quad (71)$$

where $\mathcal{V}$ is the volume of the system. Performing one spatial integral, one obtains

$$\eta = \beta \int d\mathbf{x} \int_0^\infty dt \langle \sigma_{ij}(\mathbf{x}, t) \sigma_{ij}(\mathbf{0}, 0) \rangle^{\text{eq}}. \quad (72)$$

Importantly, one should not integrate the result of Eq. (66) over **x** to obtain the result of Eq. (72), because Eq. (66) is only valid for large **x**. Instead, the integration over **x** is straightforward from Eq. (61), since it selects the mode $\mathbf{k} = 0$ in the integral, leading to

$$\eta = \frac{\beta \bar{\rho}^2}{4D} \int \frac{d\mathbf{p}}{(2\pi)^3} \frac{p_i^2 p_j^2}{p^4} \tilde{V}'(p)^2$$
$$= \frac{\beta \bar{\rho}^2}{120\pi^2 D} \int_0^\infty dp\, p^2 \tilde{V}'(p)^2. \quad (73)$$

This is the exact result for the viscosity of Brownian suspensions to leading order in the potential V, which is naturally quadratic. This expression is also in agreement with perturbation computations, where the perturbed pair distribution is found under shear. While such results have been obtained for hard spheres,[9] we have not been able to find the result of Eq. (73) in the literature for a direct check.

As pointed out, the stress tensor is not uniquely defined, but only up to divergence-free terms. The viscosity in Eq. (73) is, however, a measurable material property, which should be computable unambiguously. We therefore demonstrate that divergence-free terms do not contribute to Eq. (72). First we note that for any vector (or tensor) field $\mathbf{A}(\mathbf{x})$ which decays quicker than $1/|\mathbf{x}|^3$ for large $|\mathbf{x}|$, we have

$$\int d\mathbf{x}\, A_i(\mathbf{x}) = \int d\mathbf{x}(\nabla_j x_i) A_j(\mathbf{x}) = -\int d\mathbf{x}\, x_i \nabla_j A_j(\mathbf{x}). \quad (74)$$

We have used that $\nabla_j x_i = \delta_{ij}$. In the last step, partial integration was performed using the decay properties of $\mathbf{A}(\mathbf{x})$ at large $|\mathbf{x}|$

so that boundary terms vanish. This yields the relation

$$\int d\mathbf{x} \mathbf{A}(\mathbf{x}) = -\int d\mathbf{x} \mathbf{x} \nabla \cdot \mathbf{A}. \tag{75}$$

Proceeding by applying this to the auto-correlator of stress in Eq. (72), we denote $\mathbf{A}(\mathbf{x}) = \int_0^\infty dt \langle \delta\boldsymbol{\sigma}(\mathbf{x}, t)\delta\sigma_{ij}(\mathbf{0}, 0)\rangle^{\text{eq}}$. If this expression decays quickly enough, any divergence-free parts do not contribute to the viscosity in Eq. (72). Using the form of $\boldsymbol{\sigma}$ from Eq. (20) with $i \neq j$, we see that, to the considered order, the stress correlation decays quickly enough, and that therefore the viscosity in Eq. (73) is unambiguous.

VII. SUMMARY

The exact stochastic equation for the evolution of the density operator[54] allows one to identify the (stochastic) form of the stress tensor directly. The mean of this quantity naturally agrees with the one found from the Smoluchowski equation. It is interesting to note that non-equilibrium states with a finite particle current give rise to an additional force contribution so that the stress tensor has to be applied with care when computing forces on boundaries or objects. The non-equilibrium body force in a general field theory, where the field Φ is associated with a density operator, can be derived using force balance arguments. The result of this agrees with that obtained from the evolution of the density operator for the field theory given by Eq. (12) with the identification $\Phi = \rho$. This allows a systematic derivation of the field theoretic stress tensor, valid out of equilibrium, and the associated procedure for computing non-equilibrium forces on immersed objects/potentials.

We further computed the spatial and temporal correlations of the stress tensor exactly to leading order in interaction potential V. These correlations (or more generally forces) are more long ranged than the equilibrium correlation length, a fact which we attribute to the conservation of particles in the system considered here. In contrast to Ref. 49, our system conserves particle density, but not momentum. Off-diagonal stress correlations from the field theory were shown to agree qualitatively with those from the microscopic stress tensor.

Future work can thus involve computing these correlations for the case of particle density and momentum conservation. The impact of such correlations on flow in confinement poses further interesting questions.[30,51]

APPENDIX A: DERIVATION OF THE STRESS TENSOR FOR ANY REALIZATION OF THE DENSITY FIELD [EQ. (17)]

Here we show that the divergence of Eq. (17) equals Eq. (14) under minimal assumptions and without any thermodynamic averaging. An alternative derivation can be found in Ref. 66. Making the trivial decomposition into an ideal part and the interacting term, we can write

$$\boldsymbol{\sigma} = \boldsymbol{\sigma}^i + \boldsymbol{\sigma}^{ni}, \tag{A1}$$

where $\boldsymbol{\sigma}^i$ is the ideal gas contribution,

$$\boldsymbol{\sigma}^i(\mathbf{x}) = -k_B T \rho(\mathbf{x})\mathbf{I}. \tag{A2}$$

The remaining non-ideal term due to inter-particle interactions can be written as

$$\nabla \cdot \boldsymbol{\sigma}^{ni}(\mathbf{x}) = -\rho(\mathbf{x})\int d\mathbf{r} \nabla V(\mathbf{r})\rho(\mathbf{x} - \mathbf{r})$$
$$= -\frac{1}{2}\rho(\mathbf{x})\int d\mathbf{r}\,[\nabla V(\mathbf{r})\rho(\mathbf{x} - \mathbf{r})$$
$$+ \nabla V(-\mathbf{r})\rho(\mathbf{x} + \mathbf{r})], \tag{A3}$$

where we used a translation of the integration variable, and in the second line we have symmetrized the integral over the variables $\mathbf{r}$ and $-\mathbf{r}$. At this point it is important to choose interaction potentials obeying the reflection symmetry $\nabla V(\mathbf{r}) = -\nabla V(-\mathbf{r})$ [this is clearly satisfied by isotropic potentials with $V(\mathbf{r}) = V(r)$], for which one may write

$$\nabla V(\mathbf{r}) = \frac{\mathbf{r}}{r}V'(r). \tag{A4}$$

This leads to

$$\nabla \cdot \boldsymbol{\sigma}^{ni}(\mathbf{x}) = -\frac{1}{2}\int d\mathbf{r} \nabla V(\mathbf{r})\,[\rho(\mathbf{x})\rho(\mathbf{x} - \mathbf{r}) - \rho(\mathbf{x})\rho(\mathbf{x} + \mathbf{r})]. \tag{A5}$$

Denoting

$$f(\mathbf{x}, \mathbf{r}, \lambda) = \rho(\mathbf{x} - \lambda\mathbf{r})\rho(\mathbf{x} + (1 - \lambda)\mathbf{r}), \tag{A6}$$

we can write

$$\nabla \cdot \boldsymbol{\sigma}^{ni}(\mathbf{x}) = -\frac{1}{2}\int d\mathbf{r} \nabla V(\mathbf{r})\,[f(\mathbf{x}, \mathbf{r}, 1) - f(\mathbf{x}, \mathbf{r}, 0)] \tag{A7}$$

$$= -\frac{1}{2}\int d\mathbf{r} \nabla V(\mathbf{r})\left[\int_0^1 d\lambda \frac{\partial f(\mathbf{x}, \mathbf{r}, \lambda)}{\partial \lambda}\right]. \tag{A8}$$

It is easy to see that

$$\frac{\partial f(\mathbf{x}, \mathbf{r}, \lambda)}{\partial \lambda} = -\mathbf{r} \cdot \nabla_{\mathbf{x}}\,[\rho(\mathbf{x} - \lambda\mathbf{r})\rho(\mathbf{x} + (1 - \lambda)\mathbf{r})], \tag{A9}$$

where the gradient should be taken with respect to $\mathbf{x}$. Putting this together for isotropic potentials then yields

$$\nabla \cdot \boldsymbol{\sigma}^{ni}(\mathbf{x}) = \nabla \cdot \left[\frac{1}{2}\int d\mathbf{r} \frac{\mathbf{r}\mathbf{r}}{r}V'(r)\right.$$
$$\left. \times \int_0^1 d\lambda \rho(\mathbf{x} - \lambda\mathbf{r})\rho(\mathbf{x} + (1 - \lambda)\mathbf{r})\right]. \tag{A10}$$

The equality of the divergence of Eqs. (17) and (14) then follows.

APPENDIX B: RELATIONS USED FOR THE DERIVATION OF THE STRESS TENSOR IN THE FIELD THEORY

The following relation is useful for derivation of Eq. (34) (noting that the Hamiltonian depends on the field and its gradients):

$$\nabla_i \mathcal{H} = (\nabla_i \Phi)\frac{\partial \mathcal{H}}{\partial \Phi} + (\nabla_i \nabla_j \Phi)\frac{\partial \mathcal{H}}{\partial \nabla_j \Phi} - \frac{\partial \mathcal{H}}{\partial X_i}. \tag{B1}$$

We have added a term in the Hamiltonian, which depends explicitly on a position $\mathbf{X}$, the position of the object giving rise to the external potential U. Another explicit form is

$$\frac{\delta H}{\delta \Phi(\mathbf{x})} = \frac{\partial \mathcal{H}}{\partial \Phi} - \nabla_j \frac{\partial \mathcal{H}}{\partial \nabla_j \Phi}. \tag{B2}$$

Putting this together yields for the force in Eq. (30),

$$f_i^{(j)} = -\nabla_i \left[\Phi(\mathbf{x}) \frac{\delta H}{\delta \Phi(\mathbf{x})} \right] + \nabla_i \mathcal{H} + \frac{\partial \mathcal{H}}{\partial X_i} - \nabla_j \left[(\nabla_i \Phi) \frac{\partial \mathcal{H}}{\partial \nabla_j \Phi} \right]. \tag{B3}$$

We can also identify $-\frac{\partial \mathcal{H}}{\partial X_i}$ with the force density in direction i acting on the object by the coordinate $\mathbf{X}$. This gives Eqs. (34) and (35) in the main text.

In order to demonstrate the agreement between Eq. (34) and Ref. 26, we use

$$\nabla_i \left(\Phi(\mathbf{x}) \frac{\delta H}{\delta \Phi(\mathbf{x})} \right) = \Phi(\mathbf{x}) \nabla_i \frac{\delta H}{\delta \Phi(\mathbf{x})} + \frac{\delta H}{\delta \Phi(\mathbf{x})} \nabla_i \Phi(\mathbf{x}) \tag{B4}$$

to arrive at

$$f_i^{(U)} = \nabla_j \left[\delta_{ij} \mathcal{H} - \nabla_i \Phi \frac{\partial \mathcal{H}}{\partial \nabla_j \Phi} \right] - \frac{\delta H}{\delta \Phi(\mathbf{x})} \nabla_i \Phi(\mathbf{x}), \tag{B5}$$

which is identical to Eq. (18) of Ref. 26.

In global equilibrium, one can prove[26] that the body force vanishes exactly and, furthermore, that

$$\nabla_i \left\langle \Phi(\mathbf{x}) \frac{\delta H}{\delta \Phi(\mathbf{x})} \right\rangle^{\mathrm{eq}} = 0 = \left\langle \Phi(\mathbf{x}) \nabla_i \frac{\delta H}{\delta \Phi(\mathbf{x})} \right\rangle^{\mathrm{eq}}. \tag{B6}$$

APPENDIX C: CORRELATIONS OF AN IDEAL GAS

For the computation of the stress tensor correlations to leading order in potential V, we require the time-dependent correlation functions of the ideal gas, which can be computed exactly.[64] We write the Fourier transform of the density field for N ideal particles as

$$\tilde{\rho}(\mathbf{k}, t) = \sum_{\mu=1}^{N} \exp\left(-i\mathbf{k} \cdot [\mathbf{x}_{\mu 0} + \mathbf{X}_\mu(t)] \right), \tag{C1}$$

where $\mathbf{x}_{\mu 0}$ denotes the position of particle μ at time $t = 0$ and $\mathbf{X}_\mu(t)$ is its subsequent displacement at time t. In this approximation, all the $\mathbf{X}_\mu(t)$ are independent Brownian motions with diffusion constant D. Two averages are now taken. Firstly, the average over the initial coordinate $\mathbf{x}_{i0}$ takes the form

$$\langle \cdot \rangle = \frac{1}{\mathcal{V}} \int d\mathbf{x} \cdot, \tag{C2}$$

where $\mathcal{V}$ is the volume of the system. The second average is over the Brownian motions $\mathbf{X}_\mu(t)$. The first two correlation functions are

$$\langle \tilde{\rho}(\mathbf{k}, t) \rangle^{\mathrm{id}} = \overline{\rho}(2\pi)^3 \delta(\mathbf{k}) \tag{C3}$$

and

$$\langle \tilde{\rho}(\mathbf{k}, t)\tilde{\rho}(\mathbf{q}, 0) \rangle^{\mathrm{id}} = (2\pi)^3 \overline{\rho}\delta(\mathbf{k} + \mathbf{q})e^{-Dq^2 t} + (2\pi)^6 \overline{\rho}^2 \delta(\mathbf{k})\delta(\mathbf{q}). \tag{C4}$$

From these correlations, we can get the correlations of the fluctuations. First, we note that the decomposition $\rho(\mathbf{x}, t) = \overline{\rho} + \phi(\mathbf{x}, t)$ reads in Fourier space $\tilde{\rho}(\mathbf{k}, t) = (2\pi)^3 \overline{\rho}\delta(\mathbf{k}) + \tilde{\phi}(\mathbf{k}, t)$. We deduce that

$$\langle \tilde{\phi}(\mathbf{k}, t) \rangle^{\mathrm{id}} = 0, \tag{C5}$$

$$\langle \tilde{\phi}(\mathbf{k}, t)\tilde{\phi}(\mathbf{q}, 0) \rangle^{\mathrm{id}} = (2\pi)^3 \overline{\rho}\delta(\mathbf{k} + \mathbf{q})e^{-Dq^2 t}. \tag{C6}$$

Equation (C6) is used to get Eq. (51) from Eq. (50) in the main text.

The three-point function is found similarly. After a few computation steps, we get

$$\begin{aligned}
&\langle \tilde{\rho}(\mathbf{k}, t)\tilde{\rho}(\mathbf{k}', t)\tilde{\rho}(\mathbf{q}, 0) \rangle^{\mathrm{id}} \\
&= (2\pi)^3 \overline{\rho}\delta(\mathbf{k} + \mathbf{k}' + \mathbf{q}) \exp(-Dq^2 t) \\
&\quad + (2\pi)^6 \overline{\rho}^2 \left[\delta(\mathbf{k} + \mathbf{k}')\delta(\mathbf{q}) \right. \\
&\quad \left. + \delta(\mathbf{k} + \mathbf{q})\delta(\mathbf{k}') + \delta(\mathbf{k}' + \mathbf{q})\delta(\mathbf{k}) \right] \\
&\quad \times \exp(-Dq^2 t) + (2\pi)^9 \overline{\rho}^3 \delta(\mathbf{k})\delta(\mathbf{k}')\delta(\mathbf{q}).
\end{aligned} \tag{C7}$$

The three-point correlation of the fluctuations reduces to Eq. (56) in the main text.

A useful relation is

$$\tilde{A}_{ij}(\mathbf{k}) = -\delta_{ij}\tilde{V}(k) - \frac{k_i k_j}{k}\tilde{V}'(k) \tag{C8}$$

with inverse Fourier transform

$$A_{ij}(\mathbf{r}) = \frac{r_i r_j}{r}V'(r). \tag{C9}$$

We also note that $\tilde{A}_{ij}(\mathbf{k}) = \tilde{A}_{ij}(-\mathbf{k})$. The following relation reflects the fact that a particle cannot exert forces on itself:

$$\int \frac{d\mathbf{p}}{(2\pi)^3} \tilde{A}_{ij}(\lambda \mathbf{k} - \mathbf{p}) = A_{ij}(0) = 0. \tag{C10}$$

With these relations, one may demonstrate that Eq. (56) gives no contribution to Eq. (55).

The four-point correlation function needed for evaluation of Eq. (59) is found to be

$$\begin{aligned}
\langle \tilde{\rho}(\mathbf{k}, t)\tilde{\rho}(\mathbf{k}', t)\tilde{\rho}(\mathbf{q}, 0)\tilde{\rho}(\mathbf{q}', 0) \rangle^{\mathrm{id}} &= \left[\overline{\rho}(2\pi)^3 \delta(\mathbf{k} + \mathbf{k}' + \mathbf{q} + \mathbf{q}')\mathcal{C}(\mathbf{k} + \mathbf{k}', t) + (2\pi)^6 \overline{\rho}^2 \left[\delta(\mathbf{k} + \mathbf{k}')\delta(\mathbf{q} + \mathbf{q}') \right. \right. \\
&\quad \left. + \delta(\mathbf{k} + \mathbf{q})\delta(\mathbf{k}' + \mathbf{q}')\mathcal{C}(\mathbf{k}, t)\mathcal{C}(\mathbf{k}', t) + \delta(\mathbf{k} + \mathbf{q}')\delta(\mathbf{k}' + \mathbf{q})\mathcal{C}(\mathbf{k}, t)\mathcal{C}(\mathbf{k}', t) \right] \\
&\quad + (2\pi)^6 \overline{\rho}^2 \left[\delta(\mathbf{k} + \mathbf{k}' + \mathbf{q})\delta(\mathbf{q}')\mathcal{C}(\mathbf{k} + \mathbf{k}', t) + \delta(\mathbf{k} + \mathbf{k}' + \mathbf{q}')\delta(\mathbf{q})\mathcal{C}(\mathbf{k} + \mathbf{k}', t) \right. \\
&\quad \left. + \delta(\mathbf{k} + \mathbf{q} + \mathbf{q}')\delta(\mathbf{k}')\mathcal{C}(\mathbf{k}, t) + \delta(\mathbf{k}' + \mathbf{q}' + \mathbf{q})\delta(\mathbf{k})\mathcal{C}(\mathbf{k}', t) \right] + (2\pi)^9 \overline{\rho}^3 \left[\delta(\mathbf{k} + \mathbf{k}')\delta(\mathbf{q})\delta(\mathbf{q}') \right. \\
&\quad + \delta(\mathbf{k} + \mathbf{q})\delta(\mathbf{k}')\delta(\mathbf{q}')\mathcal{C}(\mathbf{k}, t) + \delta(\mathbf{k} + \mathbf{q}')\delta(\mathbf{k}')\delta(\mathbf{q})\mathcal{C}(\mathbf{k}, t) + \delta(\mathbf{k}' + \mathbf{q})\delta(\mathbf{k})\delta(\mathbf{q}')\mathcal{C}(\mathbf{k}', t) \\
&\quad \left. + \delta(\mathbf{k}' + \mathbf{q}')\delta(\mathbf{k})\delta(\mathbf{q})\mathcal{C}(\mathbf{k}', t) + \delta(\mathbf{q} + \mathbf{q}')\delta(\mathbf{k}')\delta(\mathbf{k}) \right] + (2\pi)^{12} \overline{\rho}^4 \delta(\mathbf{k})\delta(\mathbf{k}')\delta(\mathbf{q})\delta(\mathbf{q}') \right],
\end{aligned}$$

where we introduced $\mathcal{C}(\mathbf{k},t) = e^{-Dk^2t}$. With this, Eq. (60) in the main text is found, and Eq. (61) (for $i \neq j$ and $k \neq l$) via

$$\langle \tilde{\sigma}_{ij}(\mathbf{k},t)\tilde{\sigma}_{kl}(\mathbf{q},0)\rangle = \frac{1}{4}\int_0^1 d\lambda d\lambda' \int \frac{d\mathbf{p}d\mathbf{r}}{(2\pi)^6}\tilde{A}_{ij}(\lambda\mathbf{k}-\mathbf{p})\tilde{A}_{kl}(\lambda'\mathbf{q}-\mathbf{r})\langle\tilde{\phi}(\mathbf{p},t)\tilde{\phi}(\mathbf{k}-\mathbf{p},t)\tilde{\phi}(\mathbf{r},0)\tilde{\phi}(\mathbf{q}-\mathbf{r},0)\rangle. \qquad (C11)$$

APPENDIX D: EFFECTIVE HAMILTONIAN OF REF. 56

The effective Gaussian Hamiltonian for the system of Brownian particles proposed in Ref. 56 (see Ref. 67 for a partly related approach) is

$$H = \frac{1}{2}\int d\mathbf{x}d\mathbf{y}\phi(\mathbf{x})\Delta(\mathbf{x},\mathbf{y})\phi(\mathbf{y}). \qquad (D1)$$

This form contains a quadratic potential which plays the role of an effective interaction potential between densities,

$$\Delta(\mathbf{x},\mathbf{y}) \equiv k_B T\left(\frac{1}{\bar{\rho}(\mathbf{x})}\delta(\mathbf{x}-\mathbf{y}) - c^{(2)}(\mathbf{x},\mathbf{y})\right). \qquad (D2)$$

As discussed in Ref. 56, the Hamiltonian in Eq. (D1) yields the correct result for the first two moments of the fluctuating field ϕ in equilibrium, i.e., $\langle\phi\rangle^{\mathrm{eq}} = 0$ and

$$\langle\phi(\mathbf{x})\phi(\mathbf{y})\rangle^{\mathrm{eq}} = \left(\frac{1}{\bar{\rho}(\mathbf{x})}\delta(\mathbf{x}-\mathbf{y}) - c^{(2)}(\mathbf{x},\mathbf{y})\right)^{-1}. \qquad (D3)$$

The right-hand side of Eq. (D3) is to be understood in the sense of inverse operators. The non-trivial part in the Hamiltonian in Eq. (D1) is the direct correlation function $c^{(2)}$, which is an important and well-studied object in the theory of liquids.[5] The corresponding Model B equation of motion reads[56]

$$\frac{\partial\phi}{\partial t} = \frac{D}{k_B T}\nabla\cdot\left(\langle\rho\rangle\nabla\frac{\delta H}{\delta\phi}\right) + \nabla\cdot\left(\sqrt{2D\langle\rho\rangle}\boldsymbol{\eta}\right),$$
$$= R\Delta\phi + \nabla\cdot\left(\sqrt{2D\langle\rho\rangle}\boldsymbol{\eta}\right), \qquad (D4)$$

with noise correlations given in Eq. (11). With this, Eq. (68) of the main text is found for small wave-vectors.

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