PHYSICAL REVIEW A VOLUME 22, NUMBER 2 AUGUST 1980

Statistical mechanics of stationary states. VI. Hydrodynamic fluctuation theory far from equilibrium

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(Received 19 October 1979)

Fluctuating hydrodynamics is extended to the nonequilibrium regime. Two modifications to the equilibrium theory are made: (i) The dynamics of the fluctuations are linearized about a nonequilibrium steady state (NESS). (ii) The stochastic behavior of the random forces (noise) is equilibriumlike, satisfying a local fluctuation-dissipation theorem. Thus, the noise correlations depend on position through their dependence on the NESS temperature, chemical potential, and velocity. It is shown that this is sufficient to give the results of our earlier microscopic analysis and sheds new light on their interpretation. A microscopic justification of the fluctuating hydrodynamic procedure is offered.

I. INTRODUCTION

The construction of a theory of fluctuating hydrodynamics in equilibrium[1] is fairly straightforward, since the macroscopic equations for the dynamics of the spontaneous fluctuations can be linearized in the deviation from equilibrium. The addition of "fluctuating forces" generates linear stochastic equations, which can be treated unambiguously. When a fluid is maintained out of equilibrium, the equations of motion are nonlinear, and it is not evident what is the self-consistent method of constructing stochastic equations. The purpose of this article is to derive such a fluctuating hydrodynamic theory for nonequilibrium stationary states (NESS), as well as to offer a phenomenological recipe for the calculation of correlation functions which rests only on the knowledge of the macroscopic equations.

The hydrodynamic theory of fluctuations close to equilibrium has been presented very elegantly by Fox and Uhlenbeck.[2] These authors have matched the theory of Onsager and Machlup[3] with the approach of Landau and Liftshitz.[1] A short summary of this theory will be useful in order to expose the objectives of this paper and the important differences that might arise when we consider far-from-equilibrium situations.

The basic equations are the linearized hydrodynamic equations to which random forces are added:

$$\frac{\partial \underline{\tilde{A}}(\vec{r},t)}{\partial t} = \underline{\underline{M}}(\vec{r}\,|\,\vec{r}\,') * \underline{\tilde{A}}(\vec{r}\,',t) + \underline{F}(\vec{r},t) \ . \tag{1.1}$$

In this equation $\underline{\tilde{A}}(\vec{r},t)$ represents the set of densities of conserved variables, which in a simple fluid are the number, energy, and momentum. $\underline{\underline{M}}(\vec{r}\,|\,\vec{r}\,')$ is the matrix representation of the linearized equations, and $\underline{F}$ is the set of "random forces." The symbol $*$ stands for integration over the space variable $\vec{r}\,'$, as well as a dot product, and $\sim$ denotes the deviation from the mean value.

The random forces are assumed to have zero mean value and are δ correlated in time, that is,

$$\langle \underline{F}(\vec{r},t)\, \underline{F}^{\dagger}(\vec{r}\,',t')\rangle = 2\underline{\underline{Q}}(\vec{r},\vec{r}\,')\delta(t-t') \ . \tag{1.2}$$

The matrix $\underline{\underline{Q}}$ can be easily found by solving Eq. (1.1):

$$\underline{\tilde{A}}(\vec{r},t) = e^{\underline{\underline{M}}(\vec{r}\,|\,\vec{r}\,')t} * \underline{\tilde{A}}(\vec{r}\,',0) + \int_0^t d\tau\, e^{\underline{\underline{M}}(\vec{r}\,|\,\vec{r}\,')(t-\tau)} * \underline{F}(\vec{r}\,',\tau) \ . \tag{1.3}$$

Multiplying by $\underline{A}^{\dagger}(\vec{r}_1,t)$, where the superscript $\dagger$ denotes a Hermitian conjugate, taking the limit $t\to\infty$, and using Eq. (1.2) gives

$$\underline{\underline{\Gamma}}(\vec{r},\vec{r}_1) \equiv \lim_{t\to\infty} \langle \underline{\tilde{A}}(\vec{r},t)\underline{\tilde{A}}(\vec{r}_1,t)\rangle = \int_0^{\infty} dz\, e^{\underline{\underline{M}}(\vec{r}\,|\,\vec{r}\,')z} * 2\underline{\underline{Q}}(\vec{r}\,'\,|\,\vec{r}\,'') * e^{\underline{\underline{M}}^{\dagger}(\vec{r}_1\,|\,\vec{r}\,'')z} \ . \tag{1.4}$$

In deriving Eq. (1.4) use has been made of the fact that $\underline{\underline{M}}(\vec{r}\,|\,\vec{r}\,')$ has negative definite eigenvalues and thus $\exp[\underline{\underline{M}}(\vec{r}\,|\,\vec{r}\,')t]\to 0$ when $t\to\infty$. Clearly, $\underline{\underline{\Gamma}}(\vec{r},\vec{r}_1)$ is nothing but the matrix of static correlation functions. Equation (1.4) can be rewritten

in the equivalent form

$$\underline{\underline{M}}(\vec{r}\,|\,\vec{r}\,')*\underline{\underline{\Gamma}}(\vec{r}\,'\vec{r}_1)+\underline{\underline{\Gamma}}(\vec{r}\,|\,\vec{r}\,')*\underline{\underline{M}}^{\dagger}(\vec{r}_1\,|\,\vec{r}\,')=-2\underline{\underline{Q}}(\vec{r}\,|\,\vec{r}_1)\ . \tag{1.5}$$

In *equilibrium*, equilibrium statistical mechanics can be used to compute $\underline{\underline{\Gamma}}$ (especially for long wavelengths). Thus Eq. (1.5) and a knowledge of the linearized macroscopic equations gives $\underline{\underline{Q}}$. The stochastic process, Eq. (1.1), is completely specified by taking the random forces to be Gaussian and Markovian. Equation (1.5) is analogous to the Onsager-Machlup fluctuation-dissipation theorem.[3] Following Lax,[4] we will refer to Eq. (1.5) and similar relations as *generalized Einstein relations*.

The connection to the Landau-Lifshitz theory is accomplished by identifying the random forces with the divergence of a random stress tensor and a random heat flux. Fox and Uhlenbeck have shown that Eq. (1.5) agrees with the correlations of these variables as suggested by Landau and Lifshitz.[1,2]

The picture changes completely when we are interested in nonequilibrium situations.[4] Firstly, we cannot linearize the hydrodynamic equations around equilibrium. Thus the equation for

$$\underline{\hat{a}}(\vec{r},t)\equiv\langle\underline{A}(\vec{r},t)-\langle\underline{A}(\vec{r})\rangle\rangle_{\mathrm{NE}}, \tag{1.6}$$

where the symbol $\langle\ \rangle_{\mathrm{NE}}$ denotes a nonequilibrium ensemble average, is at least quadratic in $\underline{\hat{a}}(\vec{r},t)$:

$$\frac{d\underline{\hat{a}}(\vec{r},t)}{dt}=\underline{\underline{M}}(\vec{r}\,|\,\vec{r}\,')*\underline{\hat{a}}(\vec{r}\,',t)$$
$$+\underline{\underline{N}}(\vec{r}\,|\,\vec{r}\,'\,|\,\vec{r}\,'')*\underline{\hat{a}}(\vec{r}\,',t)\underline{\hat{a}}(\vec{r}\,'',t)+\cdots. \tag{1.7}$$

The addition of random forces to such an equation produces nonlinear stochastic differential equations which can be interpreted in an infinite variety of ways, none of which enjoys preferable physical basis. However, if we are interested in the behavior around nonequilibrium stationary states (NESS), where the fluctuations are expected to be small, then it is reasonable to linearize the stochastic version of Eq. (1.7) about the NESS. Finally, even if an equation analogous to Eq. (1.5) holds, we still are not much better off, since phenomenologically both $\underline{\underline{\Gamma}}$ and $\underline{\underline{Q}}$ are unknown.

The generalization of the fluctuating-hydrodynamics method to far-from-equilibrium situations raises the following questions:

(1) What is the linear stochastic process for a given NESS?

(2) Can we establish a relation between fluctuations and dissipation analogous to Eq. (1.5)?

(3) Can we calculate $\underline{\underline{Q}}$ from another point of view (since $\underline{\underline{\Gamma}}$ is not known)?

(4) Can we then compute $\underline{\underline{\Gamma}}$ by solving the analog of Eq. (1.5)?

Similar questions have been raised previously by other authors. Keizer[5] has suggested that the analog of Eq. (1.5) can be obtained for NESS by substituting for $\underline{\underline{M}}$ the equations linearized around the NESS. The form of $\underline{\underline{Q}}$ is obtained from a phenomenological assumption that rests on the knowledge of the quasimicroscopic process that occurs in the system. Keizer did not address himself, however, to the solution of the analog of Eq. (1.5) in the case of fluids. The solution of such an equation is not trivial, but is interesting as we shall show below.

A discussion of Eq. (1.5) out of equilibrium has also been given by van Vliet[6] using a Hilbert-space formalism resting primarily on a phenomenological Langevin description. [It is interesting to note that in equilibrium, as van Vliet has shown,[6] the two terms on the left-hand side of Eq. (1.5) are equal. This is not generally true out of equilibrium.]

One of the main objectives of this paper is to show that one can derive a linear stochastic process that provides a correct description of the fluctuating properties of a stable NESS. The crux of the method is an expansion in gradients around a homogeneous (not the equilibrium) state and working to linear order in the gradients. In this way the deviation from equilibrium is immaterial. This philosophy is similar to that in the ϵ expansion of Chapman and Enskog for the solution of the Boltzmann equation.[7] Unlike some earlier workers,[8] we do not use a fluctuating Boltzmann equation approach and thus should not be limited to low density.

The linear stochastic process is obtained in $\vec{r}$ space and is completely analogous to Eq. (1.1). However, in $\vec{k}$ space the equation is complicated by the presence of mode-mode coupling terms that mix different k modes. We will show that the most important of these terms are those arising from the nonlinear reversible (or Euler) terms in the hydrodynamic equations. In this sense our findings are similar to the mode-mode coupling that is introduced near equilibrium critical points.[9]

The theory presented below predicts NESS static (and dynamic) correlation functions which can be compared with the results of the statistical-mechanical theory that was developed in this series.[10] In that theory we considered NESS static correlation functions like $\langle\underline{\tilde{A}}(\vec{r})\underline{\tilde{A}}(\vec{r}_1)\rangle_{\mathrm{NE}}$, where the symbol $\langle\ \rangle_{\mathrm{NE}}$ denotes an average over the true nonequilibrium distribution function. We Fourier

transformed this quantity in $\vec{r}-\vec{r}_1$ keeping $\vec{r}$ fixed:

$$\underline{\Gamma}(\vec{k}\,|\,\vec{r}) \equiv \langle \underline{A}_{\vec{k}} \underline{A}_{\vec{k}} \rangle(\vec{r}) = \int d(\vec{r}-\vec{r}_1) e^{i\vec{k}\cdot(\vec{r}-\vec{r}_1)} \langle \underline{\tilde{A}}(\vec{r}) \underline{\tilde{A}}(\vec{r}_1) \rangle_{NE} . \quad (1.8)$$

The result of our statistical-mechanical theory for this quantity is that[10(b),10(c)]

$$\langle \underline{A}_{\vec{k}} \underline{A}_{\vec{k}} \rangle_{NE}(\vec{r}) = \langle \underline{A}_{\vec{k}} \underline{A}_{-\vec{k}} \rangle_L(\vec{r}) - \int_0^\infty dz \langle \underline{A}_{\vec{k}} \underline{A}_{-\vec{k}} \underline{I}_T(-z) \rangle_L(\vec{r}) \cdot \vec{\nabla} \beta \underline{\Phi}_{ss}(\vec{r}) . \quad (1.9)$$

Here the symbol $\langle\ \rangle_L$ stands for an average over a local-equilibrium distribution function. The local distribution function is defined in a steady state (NESS quantities are subscribed by "ss") by

$$f_{L,ss}(X) = \frac{f_{GC}(X) \exp[\beta \underline{\Phi}_{ss}(\vec{r}) * \underline{A}(\vec{r}_1, X]}{\sum_N \int dX f_{GC} \exp[\beta \underline{\Phi}_{ss}(\vec{r}) * \underline{A}(\vec{r}_1, X)]} , \quad (1.10)$$

where X is the phase point, f_{GC} is the grand canonical equilibrium distribution function, and the set $\beta\underline{\Phi}_{ss}$ is chosen such that

$$\langle \underline{A}(\vec{r}) \rangle_L = \langle \underline{A}(\vec{r}) \rangle_{NE} \quad (1.11)$$

for variables $\underline{A}$ that belong to the set of conserved variables. It turns out that $\beta\underline{\Phi}$ is the set of thermodynamic conjugate variables. The result, Eq. (1.8), is valid to first order in the gradients of $\beta\underline{\Phi}$, but to all orders in the deviation from equilibrium.[10(e),11(b)] It was shown that $\vec{\nabla}\beta\underline{\Phi}$ is identical with the gradients of inverse temperature, chemical potential, and velocity fields.[10(d)]

Finally, the variable $\underline{I}_T$ in Eq. (1.9) is the total dissipative flux defined by

$$\dot{\underline{A}}(\vec{r},t) \equiv -\vec{\nabla}\cdot \underline{\vec{J}}(\vec{r},t) , \quad (1.12a)$$

$$\underline{\vec{I}}(\vec{r},t) \equiv \underline{\vec{J}}(\vec{r},t) - \langle \underline{\vec{J}}(\vec{r}) \underline{\tilde{A}}(\vec{r}_1) \rangle_L * \langle \underline{\tilde{A}}(\vec{r}_1) \underline{\tilde{A}}(\vec{r}_2) \rangle_L^{-1} * \underline{\tilde{A}}(\vec{r}_2, t), \quad (1.12b)$$

and

$$\underline{\vec{I}}_T(t) \equiv \int d\vec{r}\, \underline{\vec{I}}(\vec{r},t) . \quad (1.12c)$$

One of the main results of the statistical-mechanical theory is that

$$\langle \underline{A}_{\vec{k}} \underline{A}_{-\vec{k}} \rangle_{NE}(\vec{r}) - \langle \underline{A}_{\vec{k}} \underline{A}_{-\vec{k}} \rangle^{hom}(\vec{r}) = -\int_0^\infty dz\, e^{\underline{M}_{\vec{k}} z} \cdot \langle \hat{\underline{A}}_T \hat{\underline{A}}_T \underline{\vec{I}}_T \rangle^{hom}(\vec{r}) : \vec{\nabla}\beta\underline{\Phi}_{ss}(\vec{r})\, e^{\underline{M}_{\vec{k}}^\dagger z} \quad (1.13)$$

[cf. Eq. (6.5) of Ref. 10(d)]. The new average $\langle\ \rangle^{hom}(\vec{r})$ appearing in Eq. (1.13) is an equilibrium average in a homogeneous system whose (uniform) conjugate variables equal those of the NESS at the point $\vec{r}$. The distribution function for this average is found by replacing

$$\beta\underline{A}(\vec{r}_1, X) * \underline{\Phi}_{ss}(\vec{r}_1)$$

by

$$\beta\underline{A}_T(X) \cdot \underline{\Phi}_{ss}(\vec{r})$$

in Eq. (1.10) For the quantities of interest in this work, we may interchange local equilibrium and "hom" averages without introducing any significant errors [cf. Ref. 10(c)–10(d)].

The most important consequence of Eq. (1.13) is the fact that some of the static correlations in NESS are $O(k^{-2})$ for small wave vectors k. As we have previously discussed,[10] this has interesting implications for the light scattering from NESS. For example, in a simple fluid in which a temperature gradient is present, Eq. (1.13) shows that

$$\langle N_{\vec{k}}\, P^l_{-\vec{k}} \rangle_{NE} = -\frac{V k_B T \rho_0}{2k\Gamma_s} \frac{\vec{k}\cdot\vec{\nabla}\ln T}{k^2} . \quad (1.14)$$

Here, $N_{\vec{k}}$ and $P^l_{\vec{k}}$ represent the number and longitudinal momentum densities, respectively, ρ_0 is the equilibrium number density, Γ_s is the sound attenuation constant and V is the system's volume. Thus the density-momentum correlation is long ranged in space.

As we shall show below, Eq. (1.13) [and hence (1.14)] can be obtained completely from a phenomenological description of fluctuations in NESS. Moreover, such a description is derivable from microscopic considerations.

In Sec. II, a phenomenological derivation of Eq. (1.13) is presented. The discussion rests on three basic assumptions. First, the equations of motion must be linearized about the steady state. Second, the random forces appearing in the fluctuating hydrodynamic equations have the same properties as in local equilibrium. Third, the macroscopic dissipative (Onsager) coefficients are related to the random-force correlations in the usual fashion.[7,11(a),12]

Linearization around the steady state should be valid since the amplitude of the thermal fluctuations in NESS is expected to be small (away from hydrodynamic instabilities or critical points). The second assumption is made reasonable if we recall that the random forces (noise) have very short correlation times. The random-force correlations have a chance to probe only a small region of the system. The last assumption merely

states our belief that the usual Green-Kubo expressions for the macroscopic dissipative coefficients[12] still hold. This is an assumption about the macroscopic equations of motion for the NESS. Said another way, we expect that, independent of any anomalies found in the NESS static correlation functions, the nonequilibrium-statistical-mechanical results for the transport coefficients should be unchanged.

Section III offers a microscopic justification of the main assumptions made in Sec. II. Using the results of the projection operator theory given in Ref. 10(e), we show that a generalized Einstein relation analogous to Eq. (1.5) holds. The derivation clarifies the assumption made above.

In Sec. IV we show how equations similar to Eq. (1.5) can be solved when the NESS has small gradients. A functional expansion to first order in gradients is used. As expected, Eq. (1.13) is again obtained. Section V contains a discussion of the theory and conclusions.

II. PHENOMENOLOGICAL SOLUTION TO THE PROBLEM

In this section we give a phenomenological derivation of the nonequilibrium steady-state contributions to the spontaneous fluctuations in fluid systems. Just as in the case of equilibrium we assume that these fluctuations obey the macroscopic hydrodynamic equations and that they are small enough that the equations may be linearized. Now, however, we must linearize around the steady state. The kernel of the linearized phenomenological hydrodynamic equations takes the form

$$\underline{\underline{M}}_{ss}(\vec{r}\,|\,\vec{r}') = -\vec{\nabla}_r \cdot \{\underline{\underline{\vec{m}}}^{E}_{ss}(\vec{r})\delta(\vec{r}-\vec{r}') - \underline{\underline{\vec{L}}}(\vec{r}) \cdot \vec{\nabla}_r[\underline{\underline{\Gamma}}_0^{-1}(\vec{r})\delta(\vec{r}-\vec{r}')]\}\,, \tag{2.1}$$

where $\underline{\underline{\vec{L}}}(\vec{r})$ is the matrix of Onsager (transport) coefficients,[13]

$$\underline{\underline{\Gamma}}_0^{-1}(\vec{r}) \equiv \left(\frac{\partial \beta \underline{\Phi}(\vec{r})}{\partial \underline{a}(\vec{r})}\right)^{\text{hom}} = [\langle \underline{\hat{A}}_T \underline{\hat{A}}_T \rangle^{\text{hom}}(\vec{r})]^{-1}\,, \tag{2.2}$$

and

$$\underline{\underline{\vec{m}}}^{E}_{ss}(\vec{r}) \equiv \left(\frac{\partial \langle \underline{\vec{J}} \rangle}{\partial \underline{a}}\right)^{\text{hom}}(\vec{r}) = \langle \underline{\vec{J}}_T \underline{\hat{A}}_T \rangle^{\text{hom}}(\vec{r}) \cdot \underline{\underline{\Gamma}}_0^{-1}(\vec{r})\,. \tag{2.3}$$

Here, $\underline{a}(\vec{r})$ is the vector of the local values of the hydrodynamic variables: number, energy, and momentum. $\beta\underline{\Phi}(\vec{r})$, for the purpose of this section, can be thought of as the vector of the local values of the conjugate thermodynamic variables: $\beta(\vec{r})$, $\beta(\vec{r})\mu(\vec{r})$, and $\beta(\vec{r})\vec{v}(\vec{r})$.

Thus we see that $\underline{\underline{\vec{m}}}^{E}_{ss}(\vec{r})$ is associated with the reversible parts of the hydrodynamic fluxes, and $\underline{\underline{\Gamma}}_0^{-1}(\vec{r})$ plays the role of the susceptibility, converting the mechanical densities into their conjugate variables. The correlation function expressions for $\underline{\underline{\Gamma}}_0^{-1}(\vec{r})$ and $\underline{\underline{\vec{m}}}^{E}_{ss}(\vec{r})$ follow from thermodynamic fluctuation theory.[14] The quantities $\underline{\underline{\vec{m}}}^{E}_{ss}$, $\underline{\underline{\vec{L}}}(\vec{r})$, and $\underline{\underline{\Gamma}}_0(\vec{r})$ are the same functions of temperature, chemical potential, and velocity as are found in equilibrium. They are position dependent since the conjugate thermodynamic variables are nonuniform in the NESS. Note that if the system is convecting, then a moving equilibrium system must be considered in computing the various parameters appearing in $\underline{\underline{M}}_{ss}(\vec{r}\,|\,\vec{r}')$.

Having linearized the hydrodynamic equations around the steady state we are now in a position to write the steady-state analogs of Eqs. (1.1)–(1.5) (once we specify the random-force correlations). As was discussed in the Introduction, we make the assumption that the appropriate random forces are equilibriumlike. What is usually done in the fluctuating hydrodynamics is to express the matrix of random-force correlations in terms of the matrix of random-flux[1] correlations. Consistent with the local form for the hydrodynamic equations implied by Eq. (2.1), we write

$$\underline{\underline{Q}}_{ss}(\vec{r},\vec{r}') = \vec{\nabla}_r \cdot \{\underline{\underline{\vec{q}}}(\vec{r}) \cdot \vec{\nabla}_r[\delta(\vec{r}-\vec{r}')]\}\,, \tag{2.4}$$

where

$$2\underline{\underline{\vec{q}}}(\vec{r}) \equiv \int_{-\infty}^{\infty} d\tau \langle \underline{\vec{I}}_T(\tau)\underline{\vec{I}}_T\rangle^{\text{hom}}(\vec{r}) \tag{2.5}$$

is the random-flux correlation matrix.

In equilibrium the random-flux correlations and the Onsager coefficients are related by the generalized Einstein relation or, equivalently, by the Green-Kubo expressions for the transport coefficients.[1,12] We have assumed that these forms hold pointwise in nonequilibrium systems. Thus,

$$\underline{\underline{\vec{L}}}(\vec{r}) + \underline{\underline{\vec{L}}}^{\dagger}(\vec{r}) = 2\underline{\underline{\vec{q}}}(\vec{r})\,. \tag{2.6}$$

The steps leading to Eq. (1.5) now can be applied to the steady-state, linearized stochastic equation, yielding:

$$\underline{\underline{M}}_{ss}(\vec{r}\,|\,\vec{r}_1) * \underline{\underline{\Gamma}}_{ss}(\vec{r}_1,\vec{r}') + \underline{\underline{\Gamma}}_{ss}(\vec{r},\vec{r}_1) * \underline{\underline{M}}^{\dagger}_{ss}(\vec{r}'\,|\,\vec{r}_1) = -2\underline{\underline{Q}}_{ss}(\vec{r},\vec{r}')\,. \tag{2.7}$$

$\underline{\underline{\Gamma}}_{ss}(\vec{r},\vec{r}')$ is the steady-state correlation matrix for the hydrodynamic variables. We wish to find the nonequilibrium contribution to this matrix. It is advantageous to split $\underline{\underline{\Gamma}}$ into the part determined by local (hom) equilibrium and a remainder:

$$\underline{\underline{\Gamma}}_{ss}(\vec{r},\vec{r}') \equiv \langle \underline{\hat{A}}(\vec{r})\underline{\hat{A}}^{\dagger}(\vec{r}')\rangle_{ss} \equiv \underline{\underline{\Gamma}}_0(\vec{r})\delta(\vec{r}-\vec{r}') + \underline{\underline{\Gamma}}_1(\vec{r},\vec{r}')\,. \tag{2.8}$$

Since the equilibrium correlation length is small, we have approximated the short-range spatial decay of the equilibrium static correlation functions by a δ-function decay. This is valid for long-wavelength phenomena and is analogous to the implicit assumption made in writing Eq. (2.4).

Using Eqs. (2.1), (2.4), (2.6), and (2.8) in Eq. (2.7), we obtain

$$\underline{\underline{M}}_{ss}(\vec{r}\,|\,\vec{r}_1) * \underline{\underline{\Gamma}}_1(\vec{r}_1, \vec{r}') + \underline{\underline{\Gamma}}_1(\vec{r}, \vec{r}_1) * \underline{\underline{M}}^{\dagger}_{ss}(\vec{r}'\,|\,\vec{r}_1) = +\vec{\nabla}_r \cdot [\underline{\underline{\vec{m}}}^{E}_{ss}(\vec{r}) \cdot \underline{\underline{\Gamma}}_0(\vec{r})\delta(\vec{r}-\vec{r}')] + \vec{\nabla}_{r'} \cdot [\underline{\underline{\Gamma}}_0(\vec{r}) \cdot \underline{\underline{\vec{m}}}^{E\dagger}_{ss}(\vec{r})\delta(\vec{r}-\vec{r}')]$$
$$- \vec{\nabla}_r \cdot [\underline{\underline{\vec{L}}}(\vec{r}) \cdot \vec{\nabla}_r \delta(\vec{r}-\vec{r}')] - \vec{\nabla}_{r'} \cdot [\underline{\underline{\vec{L}}}^{\dagger}(\vec{r}') \cdot \vec{\nabla}_{r'} \delta(\vec{r}-\vec{r}')]$$
$$+ 2\vec{\nabla}_r \cdot [\underline{\underline{\vec{q}}}(\vec{r}) \cdot \vec{\nabla}_r \delta(\vec{r}-\vec{r}')] \tag{2.9a}$$
$$= \delta(\vec{r}-\vec{r}')\vec{\nabla}_r \cdot [\underline{\underline{\vec{m}}}^{E}_{ss}(r) \cdot \underline{\underline{\Gamma}}_0(\vec{r})]\,. \tag{2.9b}$$

In order to show the last equality, we first note that

$$\langle \vec{\underline{J}}_T \hat{\underline{A}}_T \rangle = \langle \hat{\underline{A}}_T \vec{\underline{J}}_T \rangle \tag{2.10a}$$

(a consequence of the "dot-switching" and translational invariance properties of equilibrium averages), which implies that [cf. Eq. (2.3)]

$$\underline{\underline{\vec{m}}}^{E}_{ss}(\vec{r}) \cdot \underline{\underline{\Gamma}}_0(\vec{r}) = \underline{\underline{\Gamma}}_0(\vec{r}) \cdot \underline{\underline{\vec{m}}}^{E\dagger}_{ss}(\vec{r})\,. \tag{2.10b}$$

The second equality in Eq. (2.9) is now demonstrated by multiplying the right-hand side of Eq. (2.9a) by $\psi_1(\vec{r})\psi_2(\vec{r}')$ [where $\psi_1(\vec{r})$ and $\psi_2(\vec{r}')$ are any two test functions[15]], integrating over all $\vec{r}$ and $\vec{r}'$, integrating by parts, and using Eq. (2.10b). The resulting integral simplifies to

$$\int d\vec{r}\, d\vec{r}' \psi_1(\vec{r})\psi_2(\vec{r}')\delta(\vec{r}-\vec{r}')\vec{\nabla}_r \cdot [\underline{\underline{\vec{m}}}^{E}_{ss}(\vec{r}) \cdot \underline{\underline{\Gamma}}_0(\vec{r})]\,.$$

Since ψ_1 and ψ_2 are arbitrary, this proves Eq. (2.9b). From Eq. (2.3), it can easily be shown that

$$\vec{\nabla}_r \cdot [\underline{\underline{\vec{m}}}^{E}_{ss}(\vec{r}) \cdot \underline{\underline{\Gamma}}_0(\vec{r})] = \langle \vec{\underline{J}}_T \hat{\underline{A}}_T \hat{\underline{A}}_T \rangle^{\mathrm{hom}}(\vec{r}) \cdot \vec{\nabla}\beta\underline{\underline{\Phi}}_{ss}(\vec{r}) \tag{2.11a}$$
$$= \langle \hat{\underline{A}}_T \hat{\underline{A}}_T \vec{\underline{J}}_T \rangle^{\mathrm{hom}}(\vec{r}) \cdot \vec{\nabla}\beta\underline{\underline{\Phi}}_{ss}(\vec{r})\,, \tag{2.11b}$$

where the second equality is shown in the same manner as in Eq. (2.10a). Combining Eqs. (2.10b) and (2.9b), we find that

$$\underline{\underline{M}}_{ss}(\vec{r}\,|\,\vec{r}_1) * \underline{\underline{\Gamma}}_1(\vec{r}_1, \vec{r}') + \underline{\underline{\Gamma}}_1(\vec{r}, \vec{r}_1) * \underline{\underline{M}}^{\dagger}_{ss}(\vec{r}'\,|\,\vec{r}_1)$$
$$= \delta(\vec{r}-\vec{r}')\langle \hat{\underline{A}}_T \hat{\underline{A}}_T \vec{\underline{J}}_T \rangle^{\mathrm{hom}}(\vec{r}) \cdot \vec{\nabla}_r \beta\underline{\underline{\Phi}}(\vec{r})\,. \tag{2.12}$$

Our ultimate objective is to find the behavior of

$$\underline{\underline{\Gamma}}_1(\vec{k}\,|\,\vec{r}) = \int d(\vec{r}-\vec{r}') e^{i\vec{k}\cdot(\vec{r}-\vec{r}')} \underline{\underline{\Gamma}}_1(\vec{r}, \vec{r}')\,,$$

where $\vec{r}$ is held fixed in the integration. We thus change variables in Eq. (2.12) to

$$\vec{\rho} = \vec{r} - \vec{r}'\,,$$
$$\vec{r} = \vec{r}\,,$$

for which the gradients become

$$\vec{\nabla}_r \to \vec{\nabla}_r + \vec{\nabla}_\rho\,,$$
$$\vec{\nabla}_{r'} \to -\vec{\nabla}_\rho\,.$$

Since, in the absence of gradients, Γ_1 must vanish, the leading term in $\underline{\underline{\Gamma}}_1$ is proportional to $\vec{\nabla}\beta\underline{\underline{\Phi}}(\vec{r})$. Assuming that $\vec{\nabla}\beta\underline{\underline{\Phi}}$ is much larger than $\vec{\nabla}^2\beta\underline{\underline{\Phi}}$, $(\vec{\nabla}\beta\underline{\underline{\Phi}})^2$ and higher-order terms, we can write the equation for the leading contribution to Γ_1 by dropping all gradients acting upon the hydrodynamic coefficients m^E, L, and Γ_0. For the same reason we can drop $\vec{\nabla}_r\Gamma_1(\vec{\rho}, \vec{r})$. Finally, we make the assumption that the range of $\Gamma_1(\vec{\rho}, \vec{r})$ is much smaller than the length over which the hydrodynamic coefficients vary so that, for example,

$$\underline{\underline{\vec{m}}}^{E}_{ss}(\vec{r}') = \underline{\underline{\vec{m}}}^{E}_{ss}(\vec{r})\,.$$

What remains in Eq. (2.12) after these approximations is

$$-\{\underline{\underline{\vec{m}}}^{E}_{ss}(\vec{r}) \cdot \vec{\nabla}_\rho - \underline{\underline{\vec{L}}}(\vec{r}) \cdot \underline{\underline{\Gamma}}_0^{-1}(\vec{r}) : \vec{\nabla}_\rho\vec{\nabla}_\rho\} \cdot \underline{\underline{\Gamma}}_1(\vec{\rho}, \vec{r}) + \underline{\underline{\Gamma}}_1(\vec{\rho}, \vec{r}) \cdot \{\underline{\underline{\vec{m}}}^{E\dagger}_{ss}(\vec{r}) \cdot \vec{\nabla}_\rho + [\underline{\underline{\vec{L}}}(\vec{r}) \cdot \underline{\underline{\Gamma}}_0^{-1}(\vec{r})]^{\dagger} : \vec{\nabla}_\rho\vec{\nabla}_\rho\}$$
$$= \langle \hat{\underline{A}}_T \hat{\underline{A}}_T \vec{\underline{J}}_T \rangle^{\mathrm{hom}}(\vec{r}) \cdot \vec{\nabla}\beta\underline{\underline{\Phi}}(\vec{r})\delta(\vec{\rho})\,. \tag{2.13}$$

Fourier transforming the variable ρ we obtain

$$\underline{\underline{M}}^{\mathrm{hom}}_k(\vec{r}) \cdot \underline{\underline{\Gamma}}_1(\vec{k}\,|\,\vec{r}) + \underline{\underline{\Gamma}}_1(\vec{k}\,|\,\vec{r}) \cdot \underline{\underline{M}}^{\dagger\mathrm{hom}}_k(\vec{r}) = \langle \hat{\underline{A}}_T \hat{\underline{A}}_T \vec{\underline{J}}_T \rangle^{\mathrm{hom}}(\vec{r}) \cdot \vec{\nabla}\beta\underline{\underline{\Phi}}(\vec{r})\,, \tag{2.14}$$

where $\underline{\underline{M}}^{\mathrm{hom}}_k(\vec{r})$ is the Fourier-transformed, hydrodynamic matrix linearized around the equilibrium at point $\vec{r}$. The solution to Eq. (2.14) is

$$\underline{\underline{\Gamma}}_1(\vec{k}\,|\,\vec{r}) = -\int_0^\infty d\tau \exp(\underline{\underline{M}}_{\vec{k}}^{\text{hom}}\tau)\cdot\langle \hat{\underline{A}}_T \hat{\underline{A}}_T \hat{\underline{J}}_T\rangle^{\text{hom}}\cdot\vec{\nabla}\beta\underline{\Phi}(\vec{r})\cdot\exp(\underline{\underline{M}}_{\vec{k}}^{\text{hom}\dagger}\tau)\,, \tag{2.15}$$

where all hom quantities are referenced to the point $\vec{r}$.

Equation (2.14) is similar in form to the correction to the static averages found in our microscopic appraoch [cf. Eq. (1.13)]. The difference between $\hat{\underline{J}}_T$ and $\hat{\underline{I}}_T$ is [cf. Eq. (1.12b)] $\vec{\underline{\underline{m}}}_{ss}^{E}(r)\cdot\hat{\underline{A}}$. Written in terms of the conjugate variables, the steady-state conditions are

$$\begin{aligned} 0 = \dot{\underline{a}}_{ss}(\vec{r}) &\approx \vec{\underline{\underline{m}}}_{ss}(\vec{r})\cdot\underline{\underline{\Gamma}}_0(\vec{r})\cdot\vec{\nabla}\beta\underline{\Phi}_{ss}(\vec{r}) \\ &\approx \vec{\underline{\underline{m}}}_{ss}^{E}(\vec{r})\cdot\underline{\underline{\Gamma}}_0(\vec{r})\cdot\vec{\nabla}\beta\underline{\Phi}_{ss}(\vec{r}) \\ &\approx \underline{\underline{\Gamma}}_0(r)\cdot\vec{\underline{\underline{m}}}_{ss}^{E\dagger}(\vec{r})\cdot\vec{\nabla}\beta\underline{\Phi}_{ss}(\vec{r})\,, \end{aligned} \tag{2.16}$$

where $\approx$ means "to lowest order in $\vec{\nabla}\beta\underline{\Phi}_{ss}$." Since $\underline{\underline{\Gamma}}_0(\vec{r})$ is nonsingular, $\vec{\underline{\underline{m}}}_{ss}^{E}(r)\cdot\vec{\nabla}\beta\underline{\Phi}(\vec{r})$ must vanish to lowest order. Thus we have recovered the statistical-mechanical result from hydrodynamic considerations.

It must be stressed that the stochastic properties of the random forces and their connection to the macroscopic dissipative coefficients were not modified in order to obtain our results. In the remainder of this work we justify the procedure given above.

III. A MICROSCOPIC APPROACH

A. Summary of previous results

Before examining the microscopic justification of the method given in the last section, some key results of Ref. 10(e) must be given. In that work we assumed the existence of a set of slow variables $\underline{A}(X(t),\vec{r})$, where X is the phase point. By "slow" we mean that averages containing $\dot{\underline{A}}$ are small. Typically, it is assumed that $\underline{A}$ contains the densities of the conserved variables in a simple fluid. If the set $\underline{A}$ spans the space of slow variables, then we have shown that [cf. Eq. (3.20) of Ref. 10(e)]

$$\dot{\underline{A}}(\vec{r},t) = \dot{\underline{A}}_D(\vec{r},t) + \underline{\underline{M}}_{ss}(\vec{r}\,|\,\vec{r}_1) * \tilde{\underline{A}}(\vec{r}_1,t)\,, \tag{3.1}$$

where to second order in $\dot{\underline{A}}$:

$$\begin{aligned} \underline{\underline{M}}_{ss}(\vec{r}\,|\,\vec{r}') = {} & \langle \dot{\underline{A}}(\vec{r})\tilde{\underline{A}}(\vec{r}_1)\rangle_{L,ss} * \langle \tilde{\underline{A}}(\vec{r}_1)\tilde{\underline{A}}(\vec{r}')\rangle_{L,ss}^{-1} \\ & - \int_0^\infty d\tau [\langle \dot{\underline{A}}_D(\vec{r},\tau)\dot{\underline{A}}_D(\vec{r}_1)\rangle_{L,ss} + \langle \dot{\underline{A}}_D(\vec{r},\tau)\tilde{\underline{A}}(\vec{r}_1)\dot{\underline{A}}_D(\vec{r}_2)\rangle_{L,ss} * \beta\underline{\Phi}_{ss}(\vec{r}_2)] * \langle \tilde{\underline{A}}(\vec{r}_2)\tilde{\underline{A}}(\vec{r}')\rangle_{L,ss}^{-1}\,, \end{aligned} \tag{3.2a}$$

$$\dot{\underline{A}}_D(\vec{r},t) = \dot{\underline{A}}(\vec{r},t) - \langle \dot{\underline{A}}(\vec{r})\rangle_{L,ss} - \langle \dot{\underline{A}}(\vec{r})\tilde{\underline{A}}(\vec{r}_1)\rangle_{L,ss} * \langle \tilde{\underline{A}}(\vec{r}_1)\tilde{\underline{A}}(\vec{r}_2)\rangle_{L,ss}^{-1} * \tilde{\underline{A}}(\vec{r}_2,t)\,, \tag{3.2b}$$

and

$$\tilde{\underline{A}}(\vec{r},t) \equiv \underline{A}(\vec{r},t) - \langle \underline{A}(\vec{r})\rangle_{L,ss}\,. \tag{3.2c}$$

The reader is referred to Sec. III of Ref. 10(e) for a detailed discussion of Eq. (3.1). It suffices to say that Eq. (3.1) describes the evolution of a fluctuation of a slow variable in a NESS. We remark that $\langle \tilde{\underline{A}}(\vec{r})\tilde{\underline{A}}(\vec{r}')\rangle_{L,ss}^{-1}$ is not an algebraic inverse but corresponds to a matrix of inverse functional derivatives. The random force $\dot{\underline{A}}_D$ is orthogonal (to second order in $\dot{A}$) to the slow variables in the sense that

$$\langle \dot{\underline{A}}_D(\vec{r},t)\tilde{\underline{A}}(\vec{r}')\rangle_{L,ss} = 0\,, \quad \langle \dot{\underline{A}}_D(\vec{r},t)\rangle_{L,ss} = 0\,, \tag{3.3}$$

and is therefore a fast variable.

In order to use Eq. (3.1) as a stochastic equation, the NESS correlation properties of $\dot{\underline{A}}_D(\vec{r},t)$ must be known. In order to extract this information, recall [cf. Eq. (2.22) of Ref. 10(e)]that in NESS,

$$\langle B\rangle_{NE} = \langle B\rangle_{L,ss} - \int_0^\infty d\tau \langle B(\tau)\dot{\underline{A}}_D(\vec{r}_1)\rangle_{L,ss} * \beta\underline{\Phi}_{ss}(\vec{r}_1)\,,$$

which is valid to first order in $\dot{\underline{A}}$ for any variable B. Letting

$$B = \dot{\underline{A}}_D(\vec{r},t)\dot{\underline{A}}_D(\vec{r}',t+\sigma)$$

gives

$$\langle \dot{\underline{A}}_D(\vec{r},t)\dot{\underline{A}}_D(\vec{r}',t+\sigma)\rangle_{NE} = \langle \dot{\underline{A}}_D(\vec{r})\dot{\underline{A}}_D(\vec{r}',\sigma)\rangle_{L,ss} + O(\dot{\underline{A}}_D^3)\,. \tag{3.4}$$

Since $\dot{\underline{A}}_D$ is orthogonal to the slow variables in the local-equilibrium sense [cf. Eq. (3.3)], we may conclude that the true correlation between the random forces becomes negligible on the fast time scale. Furthermore, to leading order in $\dot{\underline{A}}$, the averages in Eq. (3.4) are stationary in time. These, as we shall now show, are the only properties needed for deriving a fluctuation-dissipation relation analogous to that given in Eq. (1.5).

As a final remark, we note that the last term on the right-hand side of Eq. (3.2a) was omitted from the phenomenological discussion presented in Sec. II. It arises from the response induced by a fluctuation modifying the Onsager (transport) coefficients. In addition to this reason, the last term on the right-hand side of Eq. (3.2a) appears in order to make the macroscopic equations of motion (linearized about NESS) Galilean invariant. As is shown in the next section, this apparent difference yields a term of higher order in the gradients of $\beta\Phi$.

B. Derivation of a generalized Einstein relation

The formal solution to Eq. (3.1) is

$$\underline{\tilde{A}}(\vec{r},t)=\exp[\underline{\underline{M}}_{ss}(\vec{r}\,|\,\vec{r}')t]*\underline{\tilde{A}}(\vec{r}')+\int_0^t d\tau\,\exp[\underline{\underline{M}}_{ss}(\vec{r}\,|\,\vec{r}')(t-\tau)]*\underline{\dot{A}}_D(\vec{r}',\tau)\,. \tag{3.5}$$

Consider a stable NESS. In this case, the eigenvalues of $\underline{\underline{M}}_{ss}$ have negative real parts. Hence, Eq. (3.5) shows that

$$\lim_{t\to\infty}\langle\underline{A}(\vec{r},t)\underline{\tilde{A}}^{\dagger}(\vec{r}',t)\rangle_{NE}=\lim_{t\to\infty}\int_0^t d\tau\int_0^t ds\,e^{\underline{\underline{M}}_{ss}(\vec{r}\,|\,\vec{r}_1)\tau}*\langle\underline{\dot{A}}_D(\vec{r}_1,t-\tau)\underline{\dot{A}}_D^{\dagger}(\vec{r}_2,t-s)\rangle_{NE}*e^{\underline{\underline{M}}_{ss}^{\dagger}(\vec{r}'\,|\,\vec{r}_2)s}\,, \tag{3.6}$$

where only terms which survive for long times are written. The left-hand side of Eq. (3.6) is the static correlation function $\underline{\underline{\Gamma}}(r,r')$. In addition, using Eq. (3.4) we find that

$$\underline{\underline{\Gamma}}(\vec{r},\vec{r}')=\int_0^\infty d\tau\int_0^\infty ds\,e^{\underline{\underline{M}}_{ss}(\vec{r}\,|\,\vec{r}_1)\tau}*\langle\underline{\dot{A}}_D(\vec{r}_1,s-\tau)\underline{\dot{A}}_D^{\dagger}(\vec{r}_2)\rangle_{L,ss}*e^{\underline{\underline{M}}_{ss}^{\dagger}(\vec{r}'\,|\,\vec{r}_2)s}\,. \tag{3.7}$$

The time correlation function in the integrand of Eq. (3.7) is sharply peaked at $s-\tau=0$, quickly decaying to zero otherwise. This implies that s can be replaced by τ in the second exponential in the integrand. If this is done, we obtain

$$\underline{\underline{\Gamma}}(\vec{r},\vec{r}')=\int_0^\infty d\tau\,e^{\underline{\underline{M}}_{ss}(\vec{r}\,|\,\vec{r}_1)\tau}*\underline{\underline{\gamma}}(\vec{r}_1\,|\,\vec{r}_2)*e^{\underline{\underline{M}}_{ss}^{\dagger}(\vec{r}'\,|\,\vec{r}_2)\tau}\,, \tag{3.8}$$

where

$$\underline{\underline{\gamma}}(\vec{r}_1\,|\,\vec{r}_2)\equiv\int_{-\infty}^\infty ds\,\langle\underline{\dot{A}}_D(\vec{r}_1,s)\underline{\dot{A}}_D^{\dagger}(\vec{r}_2)\rangle_{L,ss}\,. \tag{3.9}$$

An equivalent representation of Eq. (3.8) is

$$\underline{\underline{M}}_{ss}(\vec{r}\,|\,\vec{r}_1)*\underline{\underline{\Gamma}}(\vec{r}_1,\vec{r})+\underline{\underline{\Gamma}}(\vec{r},\vec{r}_1)*\underline{\underline{M}}_{ss}^{\dagger}(\vec{r}'\,|\,\vec{r}_1)=-\underline{\underline{\gamma}}(\vec{r}\,|\,\vec{r}')\,, \tag{3.10}$$

which is precisely of the form given in Eq. (1.5).

The form of the generalized Einstein relation given above differs from the equilibrium form in two respects. First, the macroscopic equations of motion involve $\underline{\underline{M}}_{ss}$, which describes the evolution of a macroscopic system "near" the steady state. That is, the equations of motion are linearized about the NESS. The second, and more important difference, lies in the fact that the correlations of the random forces are given in terms of local-equilibrium averages. As we will show below, this means that they can be expressed in terms of equilibrium like quantities. Hence the only "unknowns" in Eq. (3.10) are the static correlation functions. This is contrary to the situation encountered in equilibrium; there the static correlations can be directly computed, but the random-force correlations cannot.

The fact that the random-force correlations are given in terms of local-equilibrium average is quite physical. Recall that the random forces evolve on the fast time scale, and averages containing them decay quickly in time. On this time scale, a random-force fluctuation does not have a chance to propagate very far. Therefore, the random-force fluctuations probe only their local environment, and this is adequately characterized by a local-equilibrium average.

Equation (3.10) is a statement of the NESS generalized Einstein relation. In general it is nonlocal in space. In order to solve the relation we consider a system where the macroscopic NESS properties vary slowly in space.

IV. SOLUTION OF THE GENERALIZED EINSTEIN RELATION

In a system where the macroscopic properties vary on a long length scale, we expect that the static correlations for short-wavelength phenomena should resemble those in a uniform system whose macroscopic properties coincide with those of the actual system at the point of interest. This is essentially a local-equilibrium hypothesis. Note that the fluctuation length scale must be

small, but need not be microscopic.

Equation (3.10) can be used to find the corrections to the local-equilibrium static correlation function. To show this, we expand Eq. (3.10) in a functional Taylor series about a state whose thermodynamic properties equal those of the NESS at $\vec{r}$, the point of interest. That is, Eq. (3.10) is expanded about a state where $\beta\underline{\Phi}_{ss}(\vec{r}_1)=\beta\underline{\Phi}_{ss}(\vec{r})$ in the local-equilibrium averages, and Φ is treated as a constant vis-a-vis spatial integrations (cf. the hom averages discussed earlier). Writing terms up to linear order in the series, we find

$$\begin{aligned}[\underline{\underline{M}}_{ss}^{hom}(\vec{r}\,|\,\vec{r}_1)*\underline{\underline{\Gamma}}^{hom}(\vec{r}_1-\vec{r}')+\underline{\underline{\Gamma}}^{hom}(\vec{r}-\vec{r}_1)*\underline{\underline{M}}_{ss}^{\dagger\,hom}(\vec{r}'\,|\,\vec{r}_1)+\underline{\underline{S}}^{(0)}(\vec{r}\,|\,\vec{r}')]\\ +\left[\underline{\underline{M}}_{ss}^{hom}(\vec{r}\,|\,\vec{r}_1)*\left(\frac{\delta\underline{\underline{\Gamma}}(\vec{r}_1\,|\,\vec{r}')}{\delta\beta\underline{\Phi}_\sigma(\vec{r}_3)}\right)^{hom}+\left(\frac{\delta\underline{\underline{\Gamma}}(\vec{r}\,|\,\vec{r}_1)}{\delta\beta\underline{\Phi}_\sigma(\vec{r}_3)}\right)^{hom}*\underline{\underline{M}}_{ss}^{\dagger\,hom}(\vec{r}'\,|\,\vec{r}_1)+\underline{S}_\sigma^{(1)}(\vec{r}\,|\,\vec{r}'\,|\,\vec{r}_3)\right]*\Delta\beta\underline{\Phi}_\sigma(\vec{r}_3\,|\,\vec{r})\\ +O((\Delta\beta\Phi)^2)=0\,,\end{aligned}\tag{4.1}$$

where

$$\Delta\beta\underline{\Phi}_\sigma(\vec{r}_3\,|\,\vec{r})\equiv\beta[\underline{\Phi}_\sigma(\vec{r}_3)-\underline{\Phi}_\sigma(\vec{r})]_{ss}\,,\tag{4.2}$$

$$\underline{\underline{S}}^{(0)}(\vec{r}\,|\,\vec{r}')\equiv\underline{\underline{\gamma}}^{hom}(\vec{r}\,|\,\vec{r}')\,,\tag{4.3}$$

$$\underline{\underline{S}}_\sigma^{(1)}(\vec{r}\,|\,\vec{r}'\,|\,\vec{r}_3)\equiv\left(\frac{\delta\underline{\underline{\gamma}}(\vec{r}\,|\,\vec{r}')}{\delta\beta\underline{\Phi}_\sigma(\vec{r}_3)}\right)^{hom}+\left(\frac{\delta\underline{\underline{M}}_{ss}(\vec{r}\,|\,\vec{r}_1)}{\delta\beta\underline{\Phi}_\sigma(\vec{r}_3)}\right)^{hom}*\underline{\underline{\Gamma}}^{hom}(\vec{r}_1\,|\,\vec{r}')+\underline{\underline{\Gamma}}^{hom}(\vec{r}\,|\,\vec{r}_1)*\left(\frac{\delta\underline{\underline{M}}^{\dagger}(\vec{r}'\,|\,\vec{r}_1)}{\delta\beta\underline{\Phi}_\sigma(\vec{r}_3)}\right)^{hom}\,,\tag{4.4}$$

and where the repeated Greek subscript (σ) is summed. In Eqs. (4.1)–(4.4) the superscript hom implies that the superscripted quantity is evaluated in the homogeneous reference state as was described above. This means that the hom quantities have an implicit $\vec{r}$ dependence through the $\beta\underline{\Phi}_{ss}(\vec{r})$ [or equivalently through $\underline{a}_{ss}(\vec{r})$]. We stress the fact that this dependence is *local in space*. Since the hom state is translationally invariant, the various hom quantities appearing in Eq. (4.1) depend explicitly on position only through differences in the various positions.

The coefficient of each power of $\Delta\beta\Phi$ in Eq. (4.1) is set to zero separately. The nth equation which thereby results in an inhomogeneous equation for the nth functional derivative of $\underline{\underline{\Gamma}}$ (evaluated in hom). Moreover, the inhomogeneous parts of the equations are expressible solely in terms of the solutions of the lower-order equations or of other known quantities.

Consider the zeroth-order equation, written symbolically as

$$\underline{\underline{M}}*\underline{\underline{\Gamma}}+\underline{\underline{\Gamma}}*\underline{\underline{M}}^{\dagger}+\underline{\underline{S}}^{(0)}=0\,.\tag{4.5}$$

(We do not write out the hom and "ss" symbols for the remainder of this work.) From Eqs. (4.3) and (3.9) it follows that $\underline{\underline{S}}^{(0)}$ is simply the same correlation matrix on the random-force fluctuations [cf. Eq. (1.2)] as encountered in an equilibrium system. Thus, reversing the usual argument implies that $\underline{\underline{\Gamma}}^{hom}(\vec{r}-\vec{r}')$ is nothing but the matrix of static correlations found in an equilibrium system (possibly in uniform motion). The properties of the equilibrium system are linked to those of the NESS in the hom sense. For long-wavelength phenomena, these static quantities are readily expressible in terms of thermodynamic quantities (e.g., compressibility heat capactities, etc.).

The equation for the first functional derivative of $\underline{\underline{\Gamma}}$ has the formal solution:

$$\frac{\delta\underline{\underline{\Gamma}}}{\delta\beta\underline{\Phi}_\sigma}=\int_0^\infty d\tau\,\exp(\underline{\underline{M}}\tau)*\underline{\underline{S}}_\sigma^{(1)}*\exp(\underline{\underline{M}}^{\dagger}\tau)\,.\tag{4.6}$$

In order to proceed, the various functional derivatives contained in $\underline{\underline{S}}_\sigma^{(1)}$ [cf. Eq. (4.4)] are needed. It follows from Eq. (3.9) that

$$\left(\frac{\delta\underline{\underline{\gamma}}(\vec{r}\,|\,\vec{r}')}{\delta\beta\underline{\Phi}_\sigma(\vec{r}_3)}\right)^{hom}=\int_{-\infty}^{\infty}ds\,\langle\dot{\underline{A}}_D(\vec{r},s)\dot{\underline{A}}_D^{\dagger}(\vec{r}')\hat{A}_\sigma(\vec{r}_3)\rangle^{hom}\,,\tag{4.7}$$

and from Eq. (3.2),

$$\begin{aligned}\left(\frac{\delta\underline{\underline{M}}_{ss}(\vec{r}\,|\,\vec{r}')}{\delta\beta\underline{\Phi}_\sigma(\vec{r}_3)}\right)^{hom}=\Big(&\langle\dot{\underline{A}}_D(\vec{r})\hat{\underline{A}}(\vec{r}_1)\hat{A}_\sigma(\vec{r}_3)\rangle*\langle\hat{\underline{A}}(\vec{r}_1)\hat{\underline{A}}(\vec{r}')\rangle^{-1}\\ &-\int_0^\infty d\tau[\langle\dot{\underline{A}}_D(\vec{r},\tau)\dot{\underline{A}}_D(\vec{r}_1)\hat{A}_\sigma(\vec{r}_3)\rangle+\langle\dot{\underline{A}}_D(\vec{r},\tau)\hat{\underline{A}}(\vec{r}_1)\dot{A}_{\sigma,D}(\vec{r}_3)\rangle\\ &-\langle\dot{\underline{A}}_D(\vec{r},\tau)\dot{\underline{A}}_D(\vec{r}_2)\rangle*\langle\hat{\underline{A}}(\vec{r}_2)\hat{\underline{A}}(\vec{r}_4)\rangle^{-1}*\langle\hat{\underline{A}}(\vec{r}_4)\hat{\underline{A}}(\vec{r}_1)\hat{A}_\sigma(\vec{r}_3)\rangle]*\langle\hat{\underline{A}}(\vec{r}_1)\hat{\underline{A}}(\vec{r}')\rangle^{-1}\Big)^{hom}\,.\end{aligned}\tag{4.8}$$

In obtaining Eqs. (4.7) and (4.8), we used the definition of the local-equilibrium average [cf. Eq. (1.10)] and the orthogonality relation, Eq. (3.3). Furthermore, only terms to second terms to second order in $\underline{\dot{A}}$ have been retained.

Noting that the hom averages are stationary allows Eq. (4.7) to be rewritten as

$$\left(\frac{\delta\underline{\underline{\gamma}}(\vec{r}|\vec{r}')}{\delta\beta\Phi_\sigma(\vec{r}_3)}\right)^{\text{hom}} = \int_0^\infty ds\left(\langle[\underline{\dot{A}}_D(\vec{r},s)\underline{\dot{A}}_D(\vec{r}')+\underline{\dot{A}}_D(\vec{r})\underline{\dot{A}}_D(\vec{r}',s)]\hat{\underline{A}}_\sigma(\vec{r}_3)\rangle+\int_0^s d\tau\langle\underline{\dot{A}}_D(\vec{r})\underline{\dot{A}}_D(\vec{r}',s)\underline{\dot{A}}_\sigma(\vec{r}_3,\tau)\rangle\right)^{\text{hom}}. \tag{4.9}$$

The most important change in the static correlation functions in NESS was the appearance of terms proportional to k^{-2} [cf. Eq. (6.6) of Ref. 10(d) or Eq. (1.14) here] when the Fourier transform indicated by Eq. (1.8) was performed. As is easily shown, the last term on the right-hand side of Eq. (4.9) yields a term at most of order unity when used in Eq. (4.6) to compute the correction to $\underline{\underline{\Gamma}}^{\text{hom}}$. In order to see how this comes about, recall that the variables $\underline{A}(\vec{r},t)$ are densities of conserved quantities. Using Eq. (1.12a) and the fact that the integrand of Eq. (4.6) decays on the k^{-2} time scale verifies our claim. We thus neglect this term.

Next, using Eq. (3.2) (in the hom sense), we rewrite Eq. (4.8) as

$$\left(\frac{\delta\underline{\underline{M}}_{ss}(\vec{r}|\vec{r}')}{\delta\beta\Phi_\sigma(\vec{r}_3)}\right)^{\text{hom}} = \Big(\langle\dot{\underline{A}}(\vec{r})\hat{\underline{A}}(\vec{r}_1)\hat{A}_\sigma(\vec{r}_3)\rangle * \langle\hat{\underline{A}}(\vec{r}_1)\hat{\underline{A}}(\vec{r}')\rangle^{-1} - \underline{\underline{M}}^{\text{hom}}(\vec{r}-\vec{r}_2) * \langle\hat{\underline{A}}(\vec{r}_2)\hat{\underline{A}}(\vec{r}_1)\hat{A}_\sigma(\vec{r}_3)\rangle * \langle\hat{\underline{A}}(\vec{r}_1)\hat{\underline{A}}(\vec{r}')\rangle^{-1}$$
$$-\int_0^\infty d\tau\langle\underline{\dot{A}}_D(\vec{r},\tau)[\underline{\dot{A}}_D(\vec{r}_1)\hat{A}_\sigma(\vec{r}_3)+\hat{\underline{A}}(\vec{r}_1)\dot{A}_{\sigma,D}(\vec{r}_3)]\rangle * \langle\hat{\underline{A}}(\vec{r}_1)\hat{\underline{A}}(\vec{r}')\rangle^{-1}\Big)^{\text{hom}}. \tag{4.10}$$

Repeating the argument used in neglecting the last term on the right-hand side of Eq. (4.9) shows that the last term on the right-hand side of Eq. (4.10) (i.e., the term containing $\dot{A}_{\sigma,D}$) is at most $O(k^{-1})$ and may thus be neglected.

Combining Eqs. (4.10), (4.9), and (4.4) shows that

$$\underline{\underline{S}}_\sigma^{(1)}(\vec{r}|\vec{r}'|\vec{r}_3) = \langle\dot{\underline{A}}(\vec{r})\hat{\underline{A}}(\vec{r}')\hat{A}_\sigma(\vec{r}_3)\rangle^{\text{hom}} + \langle\hat{\underline{A}}(\vec{r})\dot{\underline{A}}(\vec{r}')\hat{A}_\sigma(\vec{r}_3)\rangle^{\text{hom}}$$
$$-\underline{\underline{M}}^{\text{hom}}(\vec{r}-\vec{r}_1) * \langle\hat{\underline{A}}(\vec{r}_1)\hat{\underline{A}}(\vec{r}')\hat{A}_\sigma(\vec{r}_3)\rangle^{\text{hom}} - \langle\hat{\underline{A}}(\vec{r})\hat{\underline{A}}(\vec{r}_1)\hat{A}_\sigma(\vec{r}_3)\rangle^{\text{hom}} * \underline{\underline{M}}^{\dagger\,\text{hom}}(\vec{r}'-\vec{r}_1). \tag{4.11}$$

Only terms which possibly give a k^{-2} correction to $\underline{\underline{\Gamma}}^{\text{hom}}$ have been kept in Eq. (4.11). In addition, we have used the fact that

$$\underline{\underline{\Gamma}}^{\text{hom}}(\vec{r}-\vec{r}_1) * \langle\hat{\underline{A}}(\vec{r}_1)\hat{\underline{A}}(\vec{r}')\rangle^{-1\,\text{hom}} = \underline{\underline{1}}\delta(\vec{r}-\vec{r}') \tag{4.12}$$

in obtaining Eq. (4.11). Finally using the dot-switching property of equilibrium averages (i.e., stationarity) and inserting Eq. (4.11) into (4.6), we find

$$\left(\frac{\delta\underline{\underline{\Gamma}}(\vec{r}|\vec{r}')}{\delta\beta\Phi_\sigma(\vec{r}_3)}\right)^{\text{hom}} = \langle\hat{\underline{A}}(\vec{r})\hat{\underline{A}}(\vec{r}')\hat{A}_\sigma(\vec{r}_3)\rangle^{\text{hom}}$$
$$-\int_0^\infty d\tau\,\exp[\underline{\underline{M}}^{\text{hom}}(\vec{r}-\vec{r}_1)\tau] * \langle\hat{\underline{A}}(\vec{r}_1)\hat{\underline{A}}(\vec{r}_2)\dot{A}_\sigma(\vec{r}_3)\rangle^{\text{hom}} * \exp[\underline{\underline{M}}^{\dagger\,\text{hom}}(\vec{r}'-\vec{r}_2)\tau]. \tag{4.13}$$

Since this result is valid only to $O(k^{-2})$, the first term on the right-hand side is negligible. Reconstructing the functional Taylor expansion of $\underline{\underline{\Gamma}}$ thus gives

$$\underline{\underline{\Gamma}}(\vec{r}|\vec{r}') - \underline{\underline{\Gamma}}^{\text{hom}}(\vec{r}-\vec{r}')$$
$$= -\int_0^\infty d\tau\,\exp[\underline{\underline{M}}^{\text{hom}}(\vec{r}-\vec{r}_1)\tau] * \langle\hat{\underline{A}}(\vec{r}_1)\hat{\underline{A}}^\dagger(\vec{r}_2)\dot{A}_\sigma(\vec{r}_3)\rangle^{\text{hom}} * \Delta\beta\Phi_\sigma(\vec{r}_3|\vec{r}) * \exp[\underline{\underline{M}}^{\dagger\,\text{hom}}(\vec{r}'-\vec{r}_2)\tau]. \tag{4.14}$$

This expression is equivalent to what our microscopic theory gave, but with one slight difference: the variable $\dot{A}_\sigma$ which appears on the right-hand side is not $\dot{A}_{\sigma,D}$. However, noting that in a stationary state,

$$\langle\underline{\vec{J}}\hat{\underline{A}}\rangle * \vec{\nabla}\beta\underline{\Phi}_{ss} = O(\vec{\nabla}^2\beta\underline{\Phi}_{ss}) \tag{4.15}$$

allows us to replace $\dot{A}_\sigma$ by $\dot{A}_{D,\sigma}$ to order $\vec{\nabla}\beta\underline{\Phi}_{ss}$. Lastly, writing

$$\Delta\beta\Phi_\sigma(\vec{r}_3|\vec{r}) \approx (\vec{r}_3-\vec{r})\cdot\vec{\nabla}\beta\Phi_\sigma(\vec{r}), \tag{4.16}$$

using Eqs. (1.12), and integrating by parts in Eq. (4.14) yields

$$\underline{\underline{\Gamma}}(\vec{r}|\vec{r}') - \underline{\underline{\Gamma}}^{\text{hom}}(\vec{r}-\vec{r}') = -\int_0^\infty e^{\underline{\underline{M}}^{\text{hom}}\tau} * \langle\hat{\underline{A}}\hat{\underline{A}}\vec{\underline{I}}_{\sigma,\tau}\rangle^{\text{hom}}\cdot\vec{\nabla}\beta\Phi_\sigma(r) * e^{\underline{\underline{M}}^{\dagger\,\text{hom}}\tau}\,d\tau, \tag{4.17}$$

which, when Fourier transformed as indicated in Eq. (1.8), results in Eq. (1.9). Thus, to first order in the gradients of $\beta\underline{\Phi}_{ss}$ and to $O(k^{-2})$, a fluctuating hydrodynamics description is completely equivalent to the statistical-mechanics treatment of NESS fluctuations. We refer the reader to Ref. 10 for specific examples.

We should stress that the separate Einstein relation [cf. Eq. (2.6)] assumed in Sec. II and the correlations it implied occurred naturally in the microscopic treatment [cf. Eqs. (4.9)–(4.11)]. This was no accident, since Eq. (2.6) merely restates the Green-Kubo forms for the transport coefficients. As was shown in Ref. 11(b), the Green-Kubo form is valid (in the hom sense) to first order in $\vec{\nabla}\beta\underline{\Phi}$ for the flux appearing in the hydrodynamic equations (i.e., to Navier-Stokes order).

V. DISCUSSION AND CONCLUSIONS

In this work we have shown how to extend fluctuating hydrodynamic equations to nonequilibrium steady states. Three modifications were made. In the first place macroscopic equations of motion linearized around NESS and not equilibrium were used, which introduced explicitly position-dependent parameters into the equations. Second, we allowed the noise-correlation properties to vary in space through their dependence on the NESS temperature, chemical potential, and velocity. Finally, we required that the usual connection between the Onsager coefficients and the noise correlations (i.e., Green-Kubo relations) still hold in NESS. These three ingredients were sufficient to obtain the results of our earlier work.[10]

Moreover, we have shown via a microscopic approach that these three assumptions are correct to leading order in $\vec{\nabla}\beta\underline{\Phi}$. It is interesting to note that to first order in $\vec{\nabla}\beta\underline{\Phi}$, our result would be obtained if we assumed that *both* the Onsager coefficients and the noise correlations were equal to their equilibrium values, since the effects arising from their position dependence cancel each other. In addition, if we had assumed a spatial dependence, inconsistent with Eq. (2.6), then the cancellation would *not* occur, and terms involving derivatives of transport coefficients would appear in the static averages and ultimately in the light-scattering spectrum.

Recall that the main result of the microscopic calculations is that certain static correlations in NESS are long range and, as we have shown, this leads to measurable changes in the light-scattering spectrum. The phenomenological discussion of Sec. II shows that the origin of this long-range correlation is twofold, arising from changing both the noise correlation properties, as well as the form of the hydrodynamic equations. In equilibrium, the effects of the noise are "balanced" with the form of the hydrodynamic equations such that the static correlation functions are short range. If this "balance" is disturbed (e.g., in NESS), it is not surprising that a static correlation on the hydrodynamic length scale (i.e., long range) can arise.

Perhaps phenomenological theory is most clearly summarized in Eq. (2.12). It has the form of the generalized Einstein relation, except that on the right-hand side of Eq. (2.12) the random-force correlation matrix is replaced by $\delta(\vec{r}-\vec{r}')\times\langle\hat{\underline{A}}_T\hat{\underline{A}}_T\underline{\vec{I}}_T\rangle^{\text{hom}}(\vec{r})\cdot\vec{\nabla}\beta\underline{\Phi}(\vec{r})$. This same quantity appears in the reversible nonlinear parts of the hydrodynamic equations, when they are linearized about NESS, and causes mode coupling. It is this mode coupling which plays the role of the "noise" which "excites" the new parts of the static averages, and it is completely reversible in origin. Of course, one need not make use of Eq. (2.12) and could, in any specific application, start directly from the generalized Einstein relation. This might be more convenient if a higher-order theory is desired.

We have seen that the modifications to the phenomenological, fluctuating hydrodynamic equations are plausible and can be justified by microscopic considerations. There are some limitations to the present theory. These are the following.

(i) The theory is valid only to first order in $\vec{\nabla}\beta\underline{\Phi}$. If the gradients in the NESS are too large, then we cannot work to first order in $\dot{\underline{A}}$, and the functional expansion used in Sec. IV probably diverges. Moreover, the assumed form of the phenomenological equations [cf. Eq. (2.1)] is valid only to this order. In particular, Eq. (2.11) would need modification in a higher-order theory. Analogous limitations are inherent in a first-order Chapman-Enskog solution to the Boltzmann equation.[7] Note, however, that higher-order corrections can be found using the results of Ref. 11(b).

(ii) The theory is valid only for small $\vec{k}$ if the hydrodynamic equations are to be used. If $\vec{k}$ is not small generalized hydrodynamics must be used. Also, some of the terms which we have neglected may become important.

(iii) $\vec{k}$ cannot be too small. The reason for this is twofold. Boundary effects are neglected and a first-order solution of the fluctuating hydrodynamic equations is used. Specifically, $\vec{k}$ must be much larger than the inverse macroscopic length scale.

ACKNOWLEDGMENTS

One of us (D.R.) would like to thank the Miller Institute of the University of California at Berkeley for their support and the Chemistry Department for their hospitality. I. P. acknowledges the support of the Israel Commission for basic research. He also wishes to thank the Aspen Center for Physics, where a part of this paper was written, for its hospitality. One of us (J. M.) acknowledges the support of the National Science Foundation. We also thank Irwin Oppenheim and Moshe Gitterman for useful discussions.

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