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Statistical mechanics of stationary states. IV. Far-from-equilibrium stationary states and the regression of fluctuations

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The authors present a microscopic theory for the averages of any dynamical variable and in particular of fluctuations in nonequilibrium stationary states that are arbitrarily far from equilibrium, as long as the macroscopic gradients are sufficiently small. It is argued that the dynamics of the fluctuations are governed by the linearized macroscopic equations of motion (analogous to Onsager's hypotheses for equilibrium fluctuations). The fluctuation-dissipation theorem is examined and it is found that it does not hold in its equilibrium form. The authors find that the dissipation does not contain an important part of the information about the fluctuation, and attempt an interpretation of this fact.

I. INTRODUCTION

In the previous papers in this series[1-4] (hereafter Refs. 1, 2, and 3 are referred to as I, II, and III) we have developed a statistical-mechanical theory of nonequilibrium stationary states (NESS). We developed the formalism using response theory, which implies that the results obtained are valid only for NESS that are fairly close to equilibrium (up to second order in the parameters that characterize the deviation from equilibrium). In this paper we generalize our results for NESS that are arbitrarily far from equilibrium, provided the nonuniformities occur over macroscopic distances.

Since the response technique is a perturbation expansion around the *equilibrium* state, a derivation of a far-from-equilibrium result would require working to infinite order. In order to circumvent this difficulty, we employ here projection-operator techniques.[5,6] After establishing the generalized results, we examine the average time evolution of fluctuations in the NESS and the existence of a fluctuation-dissipation theorem (of the first kind). Our main results are that the Onsager regression hypotheses holds, whereas the fluctuation-dissipation theorem must be changed considerably.

The main result of I was an expression for the nonequilibrium average of an arbitrary dynamical variable, say $B(X(t),\vec{r})$, where X is the phase point

$$X \equiv \{\vec{r}^N, \vec{p}^N\}. \tag{1.1}$$

This average is denoted by $\langle B(\vec{r},t)\rangle_{\rm NE}$, and a shorthand notation is used omitting the phase-point dependence. The result was

$$\langle B(\vec{r},t)\rangle_{\rm NE} = \langle B(\vec{r})\rangle_L(t) - \int_0^\infty d\tau \langle \hat{B}(\vec{r})\underline{I}(\vec{r}_1,-\tau)\rangle_L(t) * \vec{\nabla}\beta\underline{\Phi}(\vec{r}_1,t). \tag{1.2}$$

In this expression

$$\hat{B}(\vec{r},t) \equiv B(\vec{r},t) - \langle B(\vec{r})\rangle, \tag{1.3}$$

angular brackets stand for an equilibrium grand canonical average, and the notation $\langle\,\rangle_L(t)$ implies an average over the local equilibrium distribution function

$$f_L(X,t) = f_{\rm GC}(X)\exp[\beta\underline{\Phi}(\vec{r}_1,t)*\underline{A}(\vec{r}_1)] \times\left(\sum_N \int dX f_{\rm GC}(X) \times\exp[\beta\underline{\Phi}(\vec{r}_1,t)*\underline{A}(\vec{r}_1)]\right)^{-1}. \tag{1.4}$$

Here $f_{\rm GC}(X)$ is the grand canonical distribution function. The set of conjugate variables $\beta\underline{\Phi}(\vec{r},t)$ are chosen such that

$$\langle \underline{A}(\vec{r},t)\rangle_{\rm NE} = \langle \underline{A}(\vec{r})\rangle_L(t). \tag{1.5}$$

The theory dictates that the set $\underline{A}$ must contain all the pertinent slowly varying (on the average) variables in the system. In Eqs. (1.2) and (1.4) the symbol * denotes an integration over all space of the dummy position variable that is repeated on the two sides of *, as well as a summation over the set of variables. Finally, the quantities $\underline{I}$ in Eq. (1.2) are the dissipative parts of the microscopic fluxes and will be redefined below. The parameters $\beta\underline{\Phi}(\vec{r},t)$ were shown to measure the deviation from equilibrium. Result (1.2) was verified only to second order in $\beta\underline{\Phi}$ and to first order in $\vec{\nabla}\beta\underline{\Phi}$.

Using the appropriate stationary values of $\beta\underline{\Phi}(\vec{r})$ in Eq. (1.2) yields the NESS average of B. The first objective of this paper is to generalize Eq. (1.2) to all orders in $\beta\underline{\Phi}$. This is done in Sec. II.

In III we calculated the detailed space-time dependence of correlation functions for NESS that are close to equilibrium. *A posteriori* we have found that the results obtained could be recaptured if one assumes Onsager's regression hypothesis for NESS time correlation functions. This hypothesis implies that if

$$\delta\dot{\underline{a}}(\vec{r},t)=\underline{\underline{M}}_{ss}(\vec{r}\,|\,\vec{r}_1)*\delta\underline{a}(\vec{r}_1,t), \tag{1.6}$$

where $\delta\underline{a}$ is the deviation of the macroscopic variable from the NESS and $\underline{\underline{M}}_{ss}$ governs the linearized relaxation, then

$$\langle\dot{\underline{A}}(\vec{r},t)\underline{A}(\vec{r}_2)\rangle_{\text{NESS}} =\underline{\underline{M}}_{ss}(\vec{r}\,|\,\vec{r}_1)*\langle\underline{A}(\vec{r}_1,t)\underline{A}(\vec{r}_2)\rangle_{\text{NESS}}\ . \tag{1.7}$$

Here we justify this relation for NESS that have small spatial gradients but otherwise are arbitrarily removed from equilibrium. The importance of this result will be stressed later, but it is sufficient to say now that with this result the calculation of NESS time correlation functions reduces to the calculation of NESS static correlation functions, if $\underline{\underline{M}}_{ss}$ is known. The justification of (1.7) is presented in Sec. III.

Section IV is devoted to the examination of the fluctuation-dissipation theorem of the first kind. This theorem relates the dissipation due to an external force applied to a stationary system and the spontaneous fluctuations in the system.[7] We show that in this context there is an interesting difference between equilibrium states and NESS. Section V offers conclusions and a discussion.

In all that follows, we assert that we have a classical system whose evolution is governed by the Liouville equation. The Liouvillian is denoted by iL and the microscopic dynamical variables evolve according to

$$\dot{\underline{A}}(\vec{r},t)=iL\underline{A}(\vec{r},t). \tag{1.8}$$

The main ideas developed in this paper can be followed without reference to I, II, and III. In some cases, however, we do rely on previous results to omit technical manipulations. In such cases the reader is referred to an explicit section in one of the previous papers.

II. AVERAGES OF DYNAMICAL VARIABLES IN FAR-FROM-EQUILIBRIUM NESS

As was mentioned in the Introduction, response theory is limited to a predetermined order in the deviation from equilibrium. In order to derive results that are valid to any order in displacement from equilibrium, we resort here to projection-operator techniques and generate the generalization of Eq. (1.2).

A. Formal solution for distribution function

Consider the set of dynamical variables

$$\underline{C}(X(t),\vec{r})=\begin{Bmatrix}1\\ \underline{\tilde{A}}^t(X(t),\vec{r})\end{Bmatrix}, \tag{2.1}$$

where as usual the set $\underline{A}$ contains *all* the slow variables in the system and the symbol $\underline{\tilde{A}}^t$ is defined by

$$\underline{\tilde{A}}^t(X(t),\vec{r})=\underline{A}(X(t),\vec{r})-\langle\underline{A}(\vec{r})\rangle_L(t). \tag{2.2}$$

The set $\underline{C}$ spans the space of the slow variables, including the constants. Following the methods derived in Ref. 8, we consider now the projection operator $P(t)$ defined by

$$P(t)B(\vec{r},t) =\langle B(\vec{r},t)\underline{C}(\vec{r}_1)\rangle_L(t)*\langle\underline{C}(\vec{r}_1)\underline{C}(\vec{r}_2)\rangle_L^{-1}(t)*\underline{C}(\vec{r}_2), \tag{2.3}$$

where $B(\vec{r})$ is an arbitrary dynamical variable (when t is not specified we mean that $t=0$). Here the inverse is defined by

$$\langle\underline{C}(\vec{r}_1)\underline{C}(\vec{r}_2)\rangle_L^{-1}(t)*\langle\underline{C}(\vec{r}_2)\underline{C}(\vec{r}_3)\rangle_L(t) \equiv\underline{\underline{1}}\delta(\vec{r}_1-\vec{r}_3). \tag{2.4}$$

A second projector $Q(t)$ is defined as $1-P(t)$.

Denoting the operator $\sum_N\int dX$ by Tr, we consider the operation

$$\text{Tr}[D(\vec{r}\,')P(t)B(\vec{r},t)]=\langle B(\vec{r},t)\underline{C}(\vec{r}_1)\rangle_L(t) *\langle\underline{C}(\vec{r}_1)\underline{C}(\vec{r}_2)\rangle_L^{-1}(t) *\text{Tr}\underline{C}(\vec{r}_2)D(\vec{r}\,'), \tag{2.5}$$

where D is again an arbitrary dynamical variable. The right-hand side (RHS) of Eq. (2.5) can also be expressed as $\mathrm{Tr}([P^+(t)D(\vec{r}\,')]B(\vec{r},t))$, which is in fact a definition of the projector $P^+(t)$,

$$P^+(t)D(\vec{r}\,')=f_L(X,t)\underline{C}(\vec{r}_1)*\langle\underline{C}(\vec{r}_1)\underline{C}(\vec{r}_2)\rangle_L^{-1}(t)*\mathrm{Tr}\underline{C}(\vec{r}_2)D(\vec{r}\,'). \tag{2.6}$$

It is easy to verify that $P^+(t)$ is indeed a projection operator. Let $Q^+(t)$ be the projector $1-P^+(t)$.

These two newly defined operators are now used to derive an expression for $f(X,t)$ in terms of $f_L(X,t)$. Identically,

$$f(X,t)=[Q^+(t)+P^+(t)]f(X,t). \tag{2.7}$$

By definition [cf. Eq. (2.6)],

$$P^+(t)f(X,t)=f_L(X,t)\underline{C}(\vec{r}_1)*\langle\underline{C}(\vec{r}_1)\underline{C}(\vec{r}_2)\rangle_L^{-1}(t)*\langle\underline{C}(\vec{r}_2)\rangle_{\mathrm{NE}}(t), \tag{2.8}$$

where

$$\langle\underline{C}(\vec{r}_2)\rangle_{\mathrm{NE}}(t)\equiv\mathrm{Tr}(f(X,t)\underline{C}(X,\vec{r}_2)).$$

For the special set $\underline{C}$, however,

$$\langle\underline{C}(t)\rangle_{\mathrm{NE}}=\langle\underline{C}\rangle_L(t)=0, \tag{2.9}$$

unless $C=1$ [cf. Eqs. (2.2) and (1.5)]. Thus

$$P^+(t)f(X,t)=f_L(X,t). \tag{2.10}$$

The other term in Eq. (2.7) is denoted by

$$\chi(X,t)\equiv Q^+(t)f(X,t). \tag{2.11}$$

The next stage is to derive equations of motion for $f_L(X,t)$ and $\chi(X,t)$. To do this, consider the quantity

$$P^+(t)\frac{\partial f(X,t)}{\partial t}=f_L(X,t)\underline{C}(\vec{r})*\langle\underline{C}(\vec{r})\underline{C}(\vec{r}_1)\rangle_L^{-1}(t)*\mathrm{Tr}\underline{C}(\vec{r}_1)\dot{f}(X,t). \tag{2.12}$$

Using Eqs. (1.5) and (1.4), we find

$$\mathrm{Tr}\underline{C}(\vec{r}_1)\dot{f}(X,t)=\mathrm{Tr}\underline{C}(\vec{r}_1)\dot{f}_L(X,t)=\langle\underline{C}(\vec{r}_1)\underline{\dot{A}}^t(\vec{r}_2)\rangle_L(t)*\beta\underline{\dot{\Phi}}(\vec{r}_2,t). \tag{2.13}$$

Substituting this back into Eq. (2.12) and using Eq. (2.4), we find

$$P^+(t)\frac{\partial f(X,t)}{\partial t}=f_L(X,t)\underline{\dot{A}}^t(\vec{r})*\beta\underline{\dot{\Phi}}(\vec{r},t)=\frac{\partial}{\partial t}f_L(X,t). \tag{2.14}$$

On the other hand,

$$Q^+(t)\frac{\partial f(X,t)}{\partial t}=[1-P^+(t)]\frac{\partial f(X,t)}{\partial t}=\frac{\partial\chi(X,t)}{\partial t}, \tag{2.15}$$

which is an equation of motion for $\chi(X,t)$. Noting that the left-hand side (LHS) of Eq. (2.15) is $-Q^+iL[f_L(X,t)+\chi(X,t)]$, we see that Eq. (2.15) may be solved formally for $\chi(X,t)$:

$$\chi(X,t)=T_+\exp\left(-\int_0^t Q^+(\tau)iL\,d\tau\right)\chi(X,0)-\int_0^t ds\,T_+\exp\left(-\int_s^t Q^+(\tau)iL\,d\tau\right)\times Q^+(s)iLf_L(X,s), \tag{2.16}$$

where T_+ is the time-ordering operator. As $f(X,t)=f_L(X,t)+\chi(X,t)$, we have now an exact solution of $f(X,t)$ in terms of $f_L(X,t)$ and the initial value $\chi(X,0)$. This solution contains the full N-particle dynamics and is by itself quite useless. It may be used, however, to find the nonequilibrium average of any dynamic variable $B(\vec{r},t)$:

$$\langle B(\vec{r},t)\rangle_{\mathrm{NE}}=\langle B(\vec{r})\rangle_L(t)+\mathrm{Tr}\left[B(\vec{r})T_+\exp\left(-\int_0^t Q^+(\tau)iL\,d\tau\right)\chi(X,0)\right]-\mathrm{Tr}\left[B(\vec{r})\int_0^t ds\,T_+\exp\left(-\int_0^t Q^+(\tau)iL\,d\tau\right)Q^+(s)iLf_L(X,s)\right]. \tag{2.17}$$

To make this result useful, it is necessary to introduce approximations. To this aim it is convenient to reintroduce the projectors $P(t)$ and $Q(t)$ instead of the adjoints $P^+(t)$ and $Q^+(t)$. Noting that

$$iLf_L(X,\tau)=f_L(X,\tau)\underline{\dot{A}}(\vec{r}\,')*\beta\underline{\dot{\Phi}}(\vec{r}\,',\tau),$$

we readily see that Eq. (2.17) can be rewritten as

$$\langle B(\vec{r},t)\rangle_{\mathrm{NE}}=\langle B(\vec{r})\rangle_L(t)+\left\langle Q(0)T_-\exp\left(\int_0^t iLQ(\tau)d\tau\right)B(\vec{r})\right\rangle_{\mathrm{NE}}(t=0)-\int_0^t ds\left\langle\left[Q(s)T_-\exp\left(\int_s^t iLQ(\tau)d\tau\right)B(\vec{r})\right]\underline{\dot{A}}(\vec{r}_1)\right\rangle_L(s)*\beta\underline{\dot{\Phi}}(\vec{r}_1,s), \tag{2.18}$$

where the second term on the RHS is averaged with the true distribution at time zero, and is an "initial value" term. It is rigorously zero if $\chi(X, t=0)=0$. Since

$$Q(0)T_- \exp\left(\int_0^t iLQ(\tau)\,d\tau\right) B(\vec{r})$$

is orthogonal to the slow variables $\underline{C}$, and especially to unity in the sense that

$$\left\langle Q(0)T_- \exp\left(\int_0^t iLQ(\tau)\,d\tau\right) B(\vec{r})\right\rangle_L (t=0)=0\,, \tag{2.19}$$

we assume that its average varies quickly in time compared with the evolution of the slow set and should decay to zero on molecular time scales. Thus for the calculation of $\langle B(\vec{r},t)\rangle_{NE}$ for times that are longer than microscopic decay times, we drop this term.

The most important approximation, however, concerns the third term on the RHS of Eq. (2.18). This approximation makes explicit use of the smallness of $\dot{A}$. The term appearing in the exponent, $iLQ(\tau)$, is of course

$$iLQ(\tau)=iL-iLP(\tau)=iL+O(\underline{\dot{A}})\,. \tag{2.20}$$

Thus, to first order in the smallness of $\underline{\dot{A}}$, the third term is

$$\begin{aligned}
-\int_0^t ds\langle[Q(s)e^{iL(t-s)}B(\vec{r})]\underline{\dot{A}}(\vec{r}_1)\rangle_L(s) * \beta\underline{\Phi}(\vec{r}_1,s)\\
=-\int_0^t ds\langle[Q(s)B(\vec{r},t-s)]\underline{\dot{A}}(\vec{r}_1)\rangle_L(s) * \beta\underline{\Phi}(\vec{r}_1,s)\\
=-\int_0^t d\tau\langle[Q(t-\tau)B(\vec{r},\tau)]\underline{\dot{A}}(\vec{r}_1)\rangle_L(t-\tau)\\
*\beta\underline{\Phi}(\vec{r}_1,t-\tau)\,,
\end{aligned} \tag{2.21}$$

where $\tau=t-s$.

Since the quantity $Q(t-\tau)B(\vec{r},\tau)$ is orthogonal to the slow variables in the sense that

$$\langle[Q(t-\tau)B(\vec{r},\tau)]\underline{C}(\vec{r}_1)\rangle_L(t-\tau)=0\,,$$

we assume that the correlation function in Eq. (2.21) decays to zero for $\tau>\tau_m$. It is now consistent with the approximations previously made to rewrite Eq. (2.21) as

$$-\int_0^\infty d\tau\langle[Q(t)B(\vec{r},\tau)]\underline{\dot{A}}(\vec{r}_1)\rangle_L(t) * \beta\underline{\Phi}(\vec{r}_1,t)\,.$$

Thus our final expression for an average of an arbitrary dynamical variable in any system that has a set of variables that spans the slow evolution is

$$\langle B(\vec{r},t)\rangle_{NE}=\langle B(\vec{r})\rangle_L(t) - \int_0^\infty d\tau\langle B(\vec{r},\tau)\underline{\dot{A}}_D(\vec{r}_1)\rangle_L(t) * \beta\underline{\Phi}(\vec{r}_1,t)\,, \tag{2.22}$$

where $\underline{\dot{A}}_D(\vec{r})=Q(t)\underline{\dot{A}}(\vec{r})$.

This result is valid to all orders in $\beta\underline{\Phi}$ and to first order in the smallness parameter characterizing the average time variation of $\underline{A}(\vec{r},t)$.

B. Specialization to simple fluids

It has been argued in this series and elsewhere[9] that in simple fluids the densities of the conserved variables span the slow set $\underline{A}(\vec{r},t)$. A discussion of this choice can be found in I–III or Ref. 9. In other words, the energy, number, and momentum densities are taken, along with the constants (spanned by unity), to comprise the set $\underline{C}$. With this choice we can develop Eq. (2.22) a step further. Since the equation

$$\underline{\dot{A}}(\vec{r},t)=-\vec{\nabla}\cdot\underline{J}(\vec{r},t)$$

is fulfilled for conserved variables, where the $\underline{J}(\vec{r},t)$ are the microscopic fluxes, we can write Eq. (2.22) as

$$\langle B(\vec{r},t)\rangle_{NE}=\langle B(\vec{r})\rangle_L(t) - \int_0^\infty d\tau\langle B(\vec{r},\tau)\underline{J}_D(\vec{r}_1)\rangle_L(t) * \vec{\nabla}\beta\underline{\Phi}(\vec{r}_1,t)\,, \tag{2.23}$$

where we have used Green's theorem in the last term.

This is the generalization of the expression found in I [cf. Eq. (3.7)] to all orders in $\beta\underline{\Phi}$. The main point of difference is that the dissipative fluxes here are different from the ones defined in I. There we had

$$\begin{aligned}
\underline{I}(\vec{r},t)\equiv\underline{J}_D(\vec{r},t)\\
=\underline{J}(\vec{r},t)-\langle\underline{J}(\vec{r},t)\underline{\hat{A}}(\vec{r}_1)\rangle\\
*\langle\underline{\hat{A}}(\vec{r}_1)\underline{\hat{A}}(\vec{r}_2)\rangle^{-1} * \underline{\hat{A}}(\vec{r}_2)\,,
\end{aligned} \tag{2.24}$$

whereas here

$$\begin{aligned}
\underline{J}_D(\vec{r},t)\equiv\underline{J}(\vec{r},t)-\langle\underline{J}(\vec{r},t)\underline{C}(\vec{r}_1)\rangle_L(t)\\
*\langle\underline{C}(\vec{r}_1)\underline{C}(\vec{r}_2)\rangle_L^{-1}(t) * \underline{C}(\vec{r}_2)\,.
\end{aligned} \tag{2.25}$$

Of course

$$\langle\underline{J}_D(\vec{r},t)\underline{C}(\vec{r}_1)\rangle_L(t)=0\,. \tag{2.26}$$

C. Hydrodynamic stationary states

The manner in which a NESS arises in the context of a Liouvillian theory was discussed extensively in I and will not be repeated here. All the arguments given there pertain to the present stage of development.

The specialization of Eq. (2.23) to NESS is easily obtained by changing the $\underline{\Phi}(r,t)$ which appear explicitly and implicitly in the local averages to $\underline{\Phi}(r)$, the stationary values of these quantities. The final form for the NESS average is thus

$$\langle B(\vec{r})\rangle_{\text{NESS}} = \langle B(\vec{r})\rangle_{L,\text{ss}} - \int_0^\infty d\tau \langle B(\vec{r},\tau)\underline{J}_D(\vec{r}_1)\rangle_{L,\text{ss}} * \vec{\nabla}\beta\underline{\Phi}_{\text{ss}}(\vec{r}_1). \tag{2.27}$$

III. REGRESSION OF FLUCTUATIONS TO NESS

We now show that the regression idea as embodied in Eqs. (1.6) and (1.7) is valid. As a first step we have to derive Eq. (1.6) and find the matrix $\underline{\underline{M}}_{\text{ss}}(r|r_1)$.

A. Macroscopic equations of motion, linearized around NESS

The general equation (2.22) can be used in particular for the dynamical variables $\dot{\underline{A}}(\vec{r},t)$.

Defining

$$\underline{a}(\vec{r},t) \equiv \langle \underline{A}(\vec{r},t)\rangle_{\text{NE}} - \langle \underline{A}(\vec{r})\rangle_{\text{NESS}}, \tag{3.1}$$

we find the macroscopic equation of motion

$$\dot{\underline{a}}(\vec{r},t) = \langle \dot{\underline{A}}(\vec{r})\rangle_L(t) - \int_0^\infty d\tau \langle \dot{\underline{A}}_D(\vec{r},\tau)\dot{\underline{A}}_D(\vec{r}_1)\rangle_L(t) * \beta\underline{\Phi}(\vec{r}_1,t). \tag{3.2}$$

These equations are highly nonlinear in principle. In the vicinity of a NESS, however, we may linearize them around the stationary state. We do so by expanding in $\delta\beta\underline{\Phi}(\vec{r},t)$,

$$\delta\beta\underline{\Phi}(\vec{r},t) \equiv \beta\underline{\Phi}(\vec{r},t) - \beta\underline{\Phi}_{\text{ss}}(\vec{r}), \tag{3.3}$$

keeping linear terms only. We are guided by the fact that for any variable $B(\vec{r},t)$ that is not explicity dependent on $\beta\Phi$, the functional derivative

$$\left.\frac{\delta\langle B(\vec{r})\rangle_L(t)}{\delta\beta\underline{\Phi}(\vec{r}_1,t)}\right|_{\text{ss}} = \langle B(\vec{r},t)\tilde{\underline{A}}(\vec{r}_1)\rangle_{L,\text{ss}},$$

where

$$\tilde{\underline{A}}(\vec{r},t) = \underline{A}(\vec{r},t) - \langle \underline{A}(\vec{r})\rangle_{L,\text{ss}},$$

and, if t is not specified, $t=0$. Remembering also that $\dot{\underline{a}}_{\text{ss}}=0$, we find

$$\delta\dot{\underline{a}}(\vec{r},t) = \left(\langle \dot{\underline{A}}(\vec{r})\tilde{\underline{A}}(\vec{r}_1)\rangle_{L,\text{ss}} - \int_0^\infty d\tau[\langle \dot{\underline{A}}_D(\vec{r},\tau)\dot{\underline{A}}_D(\vec{r}_1)\rangle_{L,\text{ss}} + \langle \dot{\underline{A}}_D(\vec{r},\tau)\tilde{\underline{A}}(\vec{r}_1)\dot{\underline{A}}_D(\vec{r}_2)\rangle_{L,\text{ss}} * \beta\underline{\Phi}_{\text{ss}}(\vec{r}_2)]\right) * \delta\beta\underline{\Phi}(\vec{r}_1,t). \tag{3.4}$$

In arriving at Eq. (3.4) we have used the fact that

$$\frac{\delta\langle \dot{\underline{A}}_D(\vec{r},\tau)\dot{\underline{A}}_D(\vec{r}_1)\rangle_L(t)}{\delta\beta\underline{\Phi}(\vec{r}_2,t)} = \left\langle \frac{\delta\dot{\underline{A}}_D(\vec{r},\tau)}{\delta\beta\underline{\Phi}(\vec{r}_2,t)}\dot{\underline{A}}_D(\vec{r}_1)\right\rangle_{L,\text{ss}} + \left\langle \dot{\underline{A}}_D(\vec{r},\tau)\frac{\delta\dot{\underline{A}}_D(\vec{r}_1)}{\delta\beta\underline{\Phi}(\vec{r}_2,t)}\right\rangle_{L,\text{ss}} + \langle \dot{\underline{A}}_D(\vec{r},\tau)\tilde{\underline{A}}(\vec{r}_2)\dot{\underline{A}}_D(\vec{r}_1)\rangle_{L,\text{ss}}. \tag{3.5}$$

The second term on the RHS of Eq. (3.5) is zero by construction. The first term is of third order in $\dot{\underline{A}}$ and therefore may be neglected.

Noting that [cf. Eq. (1.5)]

$$\delta\underline{a}(\vec{r},t) = \langle \tilde{\underline{A}}(\vec{r})\tilde{\underline{A}}(\vec{r}_1)\rangle_{L,\text{ss}} * \delta\beta\underline{\Phi}(\vec{r}_1,t), \tag{3.6}$$

we can rewrite Eq. (3.4) in the form

$$\delta\dot{\underline{a}}(\vec{r},t) = \underline{\underline{M}}_{\text{ss}}(\vec{r}|\vec{r}\,') * \delta\underline{a}(\vec{r}\,',t), \tag{3.7}$$

which are the equations of motion linearized around NESS, with

$$\underline{\underline{M}}_{\text{ss}}(\vec{r}|\vec{r}\,') = \left(\langle \dot{\underline{A}}(\vec{r})\tilde{\underline{A}}(\vec{r}_1)\rangle_{L,\text{ss}} - \int_0^\infty d\tau[\langle \dot{\underline{A}}_D(\vec{r},\tau)\dot{\underline{A}}_D(\vec{r}_1)\rangle_{L,\text{ss}} + \langle \dot{\underline{A}}_D(\vec{r},\tau)\tilde{\underline{A}}(\vec{r}_1)\dot{\underline{A}}_D(\vec{r}_2)\rangle_{L,\text{ss}} * \beta\underline{\Phi}_{\text{ss}}(\vec{r}_2)]\right) * \langle \tilde{\underline{A}}(\vec{r}_1)\tilde{\underline{A}}(\vec{r}\,')\rangle^{-1}_{L,\text{ss}}. \tag{3.8}$$

Note that these equations are nonlocal in space. It has been shown previously how to transform these equations to the local partial differential equations of hydrodynamics by using the condition of small macroscopic gradients. In this work we use the nonlocal form to prove the regression theorem.

We note the interesting fact that Eq. (3.8) can be rewritten in a more compact form which is a natural generalization of the equilibrium result. In Appendix A we show that, to the appropriate order in $\dot{\underline{A}}$,

$$\underline{\underline{M}}_{ss}(\vec{r}|\vec{r}\,') = \langle \dot{\underline{A}}(\vec{r},t)\tilde{\underline{A}}(\vec{r}_1)\rangle_{L,ss} * \langle \tilde{\underline{A}}(\vec{r}_1,t)\tilde{\underline{A}}(\vec{r}\,')\rangle_{L,ss}^{-1}. \quad (3.9)$$

B. Correlation functions in NESS

The general expression (2.27) can be used to express time correlation functions in the NESS; for any pair of dynamical variables $B(X,\vec{r})$ and $D(X,\vec{r}_1)$,

$$\langle B(\vec{r},\sigma)D(\vec{r}_1)\rangle_{NESS} = \langle B(\vec{r},\sigma)D(\vec{r}_1)\rangle_{L,ss} - \int_0^\infty d\tau \langle B(\vec{r},\sigma)D(\vec{r}_1)\underline{J}_D(\vec{r}_2,-\tau)\rangle_{L,ss} * \vec{\nabla}\beta\underline{\Phi}_{ss}(\vec{r}_2). \quad (3.10)$$

The objective of this section is to prove that

$$\langle \mathring{B}(\vec{r},\sigma)D(\vec{r}_1)\rangle_{NESS} = [\underline{\underline{M}}_{ss}(\vec{r}|\vec{r}_2)]_{BA} * \langle \underline{A}(\vec{r}_2,\sigma)D(\vec{r}_1)\rangle_{NESS} \quad (3.11)$$

for any $B,D \in \underline{A}$, where $\underline{\underline{M}}_{ss}$ is the linearized matrix found in Eq. (3.8). Equation (3.11) is the generalization of Onsager's regression idea to the NESS.

There is one difference between NESS correlation functions and equilibrium ones that enters into the proof below. This is in the property of "dot switching." In equilibrium,

$$\langle \dot{B}(\vec{r},\sigma)\underline{C}(\vec{r}_1)\rangle = -\langle B(\vec{r},\sigma)\dot{\underline{C}}(\vec{r}_1)\rangle. \quad (3.12)$$

This property is modified here. Consider the local equilibrium correlation function

$$\langle \dot{B}(\vec{r},\sigma)\underline{C}(\vec{r}_1)\rangle_L(t) = \langle iLB(\vec{r},\sigma)\underline{C}(\vec{r}_1)\rangle_L(t). \quad (3.13)$$

Using the hermiticity of the Liouvillian, we obtain

$$\begin{aligned}\langle [iLB(\vec{r},\sigma)]\underline{C}(\vec{r}_1)\rangle_L(t) &= -\langle B(\vec{r},\sigma)iL\underline{C}(\vec{r}_1)\rangle_L(t) - \mathrm{Tr}[B(\vec{r},\sigma)\underline{C}(\vec{r}_1)iLf_L(X,t)] \\ &= -\langle B(\vec{r},\sigma)\dot{\underline{C}}(\vec{r}_1)\rangle_L(t) - \langle B(\vec{r},\sigma)\underline{C}(\vec{r}_1)\dot{\underline{A}}(\vec{r}_2)\rangle_L(t) * \beta\underline{\Phi}(\vec{r}_2,t);\end{aligned} \quad (3.14)$$

when $\underline{\Phi}(\vec{r})=0$, we recapture Eq. (3.12).

The proof of Eq. (3.11) is divided into two parts. First we analyze the regression of the local equilibrium correlation function and then turn to the full correlation function.

C. Decay of local equilibrium correlation function

We derive now the microscopic equations of motion for $\dot{\underline{A}}$. Using the projectors P_{ss} and Q_{ss}, where

$$P_{ss}B(\vec{r},t) = \langle B(\vec{r},t)\underline{C}(\vec{r}_1)\rangle_{L,ss} * \langle \underline{C}(\vec{r}_1)\underline{C}(\vec{r}_2)\rangle_{L,ss}^{-1} * \underline{C}(\vec{r}_2),$$

$$Q_{ss} = 1 - P_{ss},$$

we write

$$\dot{\underline{A}}(\vec{r},t) = e^{iLt}iL\underline{A}(\vec{r}) = e^{iLt}(P_{ss}+Q_{ss})iL\underline{A}(\vec{r}). \quad (3.15)$$

Again we separate the RHS into two parts,

$$\dot{\underline{A}}(\vec{r},t) = \langle \dot{\underline{A}}(\vec{r})\rangle_{L,ss} + \langle \dot{\underline{A}}(\vec{r})\tilde{\underline{A}}(\vec{r}_1)\rangle_{L,ss} * \langle \tilde{\underline{A}}(\vec{r}_1)\tilde{\underline{A}}(\vec{r}_2)\rangle_{L,ss}^{-1} * \tilde{\underline{A}}(\vec{r}_2,t) + e^{iLt}Q_{ss}iL\underline{A}(\vec{r}). \quad (3.16)$$

The last term on the RHS is rewritten by invoking the well-known operator identity

$$e^{Q_{ss}iLt} = e^{iLt} - \int_0^t e^{iL(t-\tau)}P_{ss}iLe^{Q_{ss}iL\tau}d\tau. \quad (3.17)$$

Thus

$$e^{iLt}Q_{ss}iL\underline{A} = \underline{F}^{\neq}(\vec{r},t) + \int_0^t d\tau \langle iL\underline{F}^{\neq}(\vec{r},t)\underline{C}(\vec{r}_1)\rangle_{L,ss} * \langle \underline{C}(\vec{r}_1)\underline{C}(\vec{r}_2)\rangle_{L,ss}^{-1} * \underline{C}(\vec{r}_2,t-\tau), \quad (3.18)$$

where

$$\underline{F}^{\neq}(\vec{r},t) \equiv e^{Q_{ss}iLt}Q_{ss}iL\underline{A}(\vec{r}). \quad (3.19)$$

Using Eq. (3.14) for the dot switching property and Eqs. (2.1) and (3.18) in Eq. (3.16), we get the final result for the microscopic equation of motion:

$$\underline{\dot{A}}(\vec{r},t) = \underline{F}^{\ddagger}(\vec{r},t)+\langle\underline{\dot{A}}(\vec{r})\rangle_{L,ss}+\langle\underline{\dot{A}}(\vec{r})\underline{\tilde{A}}(\vec{r}_1)\rangle_{L,ss}*\langle\underline{\tilde{A}}(\vec{r}_1)\underline{\tilde{A}}(\vec{r}_2)\rangle_{L,ss}^{-1}*\underline{\tilde{A}}(\vec{r}_2,t)-\int_0^t d\tau\langle\underline{F}^{\ddagger}(\vec{r},\tau)\underline{\dot{A}}(\vec{r}_1)\rangle_{L,ss}*\beta\underline{\Phi}_{ss}(\vec{r}_1)$$
$$-\int_0^t d\tau\{[\langle\underline{F}^{\ddagger}(\vec{r},\tau)\underline{\dot{A}}(\vec{r}_2)\rangle_{L,ss}+\langle\underline{F}^{\ddagger}(\vec{r},\tau)\underline{\tilde{A}}(\vec{r}_1)\underline{\dot{A}}(\vec{r}_2)\rangle_{L,ss}*\beta\underline{\Phi}_{ss}(\vec{r}_2)]*\langle\underline{\tilde{A}}(\vec{r}_2)\underline{\tilde{A}}(\vec{r}_3)\rangle_{L,ss}^{-1}*\underline{\tilde{A}}(\vec{r}_3,t-\tau)\}\,. \tag{3.20}$$

Note that owing to the fact that our correlation functions are computed in the local equilibrium ensemble, this equation is more lengthy than the analogous one which is usually obtained for equilibrium ensembles.[6] With the help of Eq. (3.2) and the fact that $\dot{a}$ is zero in the NESS, we see that the second and fourth terms on the RHS of Eq. (3.20) disappear. Furthermore, in the stationary state, $\underline{\dot{A}}(r)*\beta\underline{\Phi}_{ss}(r)=\underline{\dot{A}}_D(r)*\beta\underline{\Phi}_{ss}(r)+O(\dot{A}^2)$. This fact allows us to replace $\dot{A}$ by $\dot{A}_D$ in all integrals that are $O(\overline{A}^2)$ already.

We can now find the rate of change of the local equilibrium correlation function simply by multiplying Eq. (3.20) by $\underline{\tilde{A}}(\vec{r})$ and averaging over $f_{L,ss}(X)$. We find

$$\langle\underline{\dot{A}}(\vec{r},t)\underline{\tilde{A}}(\vec{r}')\rangle_{L,ss}=\langle\underline{\dot{A}}(\vec{r})\underline{\tilde{A}}(\vec{r}_1)\rangle_{L,ss}*\langle\underline{\tilde{A}}(\vec{r}_1)\underline{\tilde{A}}(\vec{r}_2)\rangle_{L,ss}^{-1}*\langle\underline{\tilde{A}}(\vec{r}_2,t)\underline{\tilde{A}}(\vec{r}')\rangle_{L,ss}$$
$$-\int_0^t\{[\langle\underline{F}^{\ddagger}(\vec{r},\tau)\underline{\dot{A}}_D(\vec{r}_1)\rangle_{L,ss}+\langle\underline{F}^{\ddagger}(\vec{r},\tau)\underline{\tilde{A}}(\vec{r}_1)\underline{\dot{A}}_D(\vec{r}_3)\rangle_{L,ss}*\beta\underline{\Phi}_{ss}(\vec{r}_3)]$$
$$*\langle\underline{\tilde{A}}(\vec{r}_1)\underline{\tilde{A}}(\vec{r}_2)\rangle_{L,ss}^{-1}*\langle\underline{\tilde{A}}(\vec{r}_2,t-\tau)\underline{\tilde{A}}(\vec{r}')\rangle_{L,ss}\}\,d\tau\,. \tag{3.21}$$

In obtaining this equation we have made use of the fact that

$$\langle\underline{F}^{\ddagger}(\vec{r},t)\underline{\tilde{A}}(\vec{r}_1)\rangle_{L,ss}=0\,, \tag{3.22}$$

which stems from the fact that $\underline{F}^{\ddagger}(\vec{r},t)=Q_{ss}\underline{F}^{\ddagger}(\vec{r},t)$. This last equality is obtained by noting that $e^{Q_{ss}iLt}Q_{ss}=Q_{ss}e^{Q_{ss}iLt}Q_{ss}$, which follows from the series expansion of the exponentials and from the fact that $Q_{ss}^2=Q_{ss}$.

In fact, Eq. (3.22) can be used now to simplify Eq. (3.21) further. The meaning of Eq. (3.22) is that $\underline{F}^{\ddagger}$ has no component on the slow set, and thus the correlation function $\langle\underline{F}^{\ddagger}(\vec{r},t)\underline{\dot{A}}(\vec{r}_1)\rangle_{L,ss}$ is short lived. As is usually done, we extend the limit of integration in Eq. (3.21) to infinity and drop the τ dependence in the last correlation function. We can then rewrite Eq. (3.21) in the form

$$\langle\underline{\dot{A}}(\vec{r},t)\underline{\tilde{A}}(\vec{r}')\rangle_{L,ss}=\underline{\underline{M}}_{ss}(\vec{r}|\vec{r}_2)*\langle\underline{\tilde{A}}(\vec{r}_2,t)\underline{\tilde{A}}(\vec{r}')\rangle_{L,ss}\,, \tag{3.23}$$

where

$$\underline{\underline{M}}_{ss}(\vec{r}|\vec{r}_2)=\langle\underline{\dot{A}}(\vec{r})\underline{\tilde{A}}(\vec{r}_1)\rangle_{L,ss}*\langle\underline{\tilde{A}}(\vec{r}_1)\underline{\tilde{A}}(\vec{r}_2)\rangle_{L,ss}^{-1}$$
$$-\int_0^\infty\{[\langle\underline{F}^{\ddagger}(\vec{r},t)\underline{\dot{A}}_D(\vec{r}_1)\rangle_{L,ss}+\langle\underline{F}^{\ddagger}(\vec{r},\tau)\underline{\tilde{A}}(\vec{r}_1)\underline{\dot{A}}_D(\vec{r}_3)\rangle_{L,ss}*\beta\underline{\Phi}_{ss}(\vec{r}_3)]*\langle\underline{\tilde{A}}(\vec{r}_1)\underline{\tilde{A}}(\vec{r}_2)\rangle_{L,ss}^{-1}\}\,d\tau\,. \tag{3.24}$$

Equation (3.24) can be brought to a form that makes the identity to the previously found [Eq. (3.9)] $\underline{\underline{M}}_{ss}$ obvious. As was mentioned, Eq. (3.9) is valid to second order in $\dot{A}$. To that order, one can replace $\underline{F}^{\ddagger}$ by $e^{iLt}\overline{Q}_{ss}\,iL\underline{A}$ for the reason described by Eq. (2.16). Thus, whenever $\underline{F}^{\ddagger}$ appears in a product with $\dot{A}$, we may replace it by $\underline{\dot{A}}_D(\vec{r},t)=\underline{\dot{A}}(\vec{r},t)-P_{ss}\underline{\dot{A}}(\vec{r},t)$. With this change it is obvious that Eq. (3.24) is identical to Eq. (3.8).

D. Regression of fluctuations in NESS

Here we prove Eq. (3.11). According to the general formalism [cf. Eq. (2.22)],

$$\langle\underline{\dot{A}}(\vec{r},t)\underline{\tilde{A}}(\vec{r}_1)\rangle_{NE}=\langle\underline{\dot{A}}(\vec{r},t)\underline{\tilde{A}}(\vec{r}_1)\rangle_{L,ss}$$
$$-\int_0^\infty d\tau\langle\underline{\dot{A}}(\vec{r},t)\underline{\tilde{A}}(\vec{r}_1)\underline{J}_D(\vec{r}_2,-\tau)\rangle_{L,ss}*\vec{\nabla}\beta\underline{\Phi}_{ss}(\vec{r}_2)\,. \tag{3.25}$$

The first term on the RHS was examined in Sec. III C. We treat the second by expressing $\dot{A}(\vec{r},t)$ as

$$\underline{\dot{A}}(\vec{r},t)\equiv Q_{ss}^t\,\underline{A}(\vec{r},t)+\langle\underline{\dot{A}}(\vec{r},t)\rangle_{L,ss}+\langle\underline{\dot{A}}(\vec{r},t)\underline{\tilde{A}}(\vec{r}_1)\rangle_{L,ss}*\langle\underline{\tilde{A}}(\vec{r}_1,t)\underline{\tilde{A}}(\vec{r}_2)\rangle_{L,ss}^{-1}*\underline{\tilde{A}}(\vec{r}_2,t)\,, \tag{3.26}$$

where Q^t_{ss} is defined by Eq. (3.26).

Substituting this expression in the second term on the RHS of (3.25), we find two terms [cf. Eq. (2.26)]:

$$-\int_0^\infty d\tau\langle[Q^t_{ss}\,\underline{A}(\vec{r},t)]\tilde{A}(\vec{r}_1)J_D(\vec{r}_2,-\tau)\rangle_{L,ss}*\vec{\nabla}\beta\underline{\Phi}_{ss}(\vec{r}_2)$$
$$-\langle\dot{\underline{A}}(\vec{r},t)\underline{\tilde{A}}(\vec{r}')\rangle_{L,ss}*\langle\underline{\tilde{A}}(\vec{r}',t)\underline{\tilde{A}}(\vec{r}'')\rangle^{-1}_{L,ss}*\int_0^\infty d\tau\langle\underline{\tilde{A}}(\vec{r}'',t)\underline{\tilde{A}}(\vec{r}_1)\underline{J_D}(\vec{r}_2,-\tau)\rangle_{L,ss}*\vec{\nabla}\beta\underline{\Phi}(\vec{r}_2). \tag{3.27}$$

First we dispose of the first term. It contains two dissipative variables that decay very quickly The correlation function peaks when all the time arguments are the same. For any t that is nonzero its largest value is obtained when $\tau=0$. If t is larger than some τ_D, it has never any significant magnitude and thus can be dropped.

Using Eq. (3.9), the second term becomes

$$-\underline{\underline{M}}_{ss}(\vec{r}|\vec{r}'')*\int_0^\infty d\tau\langle\underline{\tilde{A}}(\vec{r}'',t)\underline{\tilde{A}}(\vec{r}_1)\underline{J_D}(\vec{r}_2,-\tau)\rangle_{L,ss}*\nabla\beta\underline{\Phi}(\vec{r}_2). \tag{3.28}$$

Collecting Eqs. (3.23), (3.25), and (3.28), we find our final result:

$$\langle\dot{\underline{A}}(\vec{r},t)\underline{\tilde{A}}(\vec{r}_1)\rangle_{\rm NESS}=\underline{\underline{M}}_{ss}(\vec{r}|\vec{r}_2)*\langle\underline{\tilde{A}}(\vec{r}_2,t)\underline{\tilde{A}}(\vec{r}_1)\rangle_{\rm NESS}\,. \tag{3.29}$$

We remind the reader that $\langle\ \rangle_{\rm NESS}$ is the average in the true nonequilibrium ensemble. We have thus proved the regression hypotheses for NESS. The only property of the set $\underline{A}$ that we have used is that it spans the slow variables. Thus the proof holds for any NESS—hydrodynamic or not.

IV. FLUCTUATION-DISSIPATION THEOREM

The usual fluctuation-dissipation theorem of the first kind states that there is a simple relation between the spontaneous fluctuations which occur in an equilibrium system and the dissipation which occurs when that system is linearly displaced from equilibrium.[7]

Using linear-response theory, the response of a system initially at equilibrium with respect to H_0 at time $t=-\infty$, subject to the Hamiltonian

$$H_T(t)=H_0-\underline{A}(\vec{r})*\underline{F}(\vec{r},t), \tag{4.1}$$

is[7]

$$\langle\underline{\tilde{A}}(\vec{r},t)\rangle_{\rm NE}=-\int_{-\infty}^t dt'\,\phi_{\underline{AA}}(\vec{r}|\vec{r}';t-t')*\underline{F}(\vec{r}',t'). \tag{4.2}$$

Here H_0 is the Hamiltonian of the unperturbed system; $\underline{F}(\vec{r},t)$ are weak external forces which couple to the set of variables $\underline{A}$, $\underline{F}(\vec{r},-\infty)=0$, and ϕ is the after-effect function. The quantity $\langle\underline{\tilde{A}}\rangle_{\rm NE}$ is the difference between the average values of $\underline{A}$ in a system subject to the Hamiltonian H_T and in a system subject to the Hamiltonian H_0.

It is advantageous to define the Fourier transform of ϕ in space and time,

$$\chi_{\underline{AA}}(\vec{k},\omega)\equiv\lim_{\epsilon\to 0}\int_0^\infty d\Delta t\int d\Delta\vec{r}\,\phi_{\underline{AA}}(\Delta\vec{r},\Delta t)\times e^{i\vec{k}\cdot\Delta\vec{r}-i\omega\Delta t}e^{-\epsilon\Delta t}, \tag{4.3}$$

where $\Delta r=\vec{r}-\vec{r}'$ and $\Delta t=t-t'$. If the system is not translationally invariant, this function will have a parametric dependence on $\vec{r}$.

It has been shown that, if the dissipation is defined as the work averaged over one cycle of a monochromatic force, then this dissipation $Q(\vec{k},\omega)$ is proportional to the imaginary part of $\chi(\vec{k},\omega)$:

$$Q(\vec{k},\omega)=-V\tfrac{1}{2}\omega\chi''_{\underline{AA}}(\vec{k},\omega)\,|F_{\vec{k}\omega}|^2. \tag{4.4}$$

The proof of the fluctuation-dissipation theorem in equilibrium reduces then to showing that $\chi_{\underline{AA}}$ is the Fourier transform of the autocorrelation function $\langle\underline{A}_{\vec{k}}(t)\underline{A}_{-\vec{k}}\rangle$.[7]

We examine now the analog of this theorem in the NESS and we shall clarify the differences that occur between a NESS and equilibrium states.

A. After-effect function

Consider again the Hamiltonian in Eq. (4.1). The corresponding Liouvillian is

$$L_T=L_0+L_1(t),$$
$$L_0=i[H_0,\,];\;L_1(t)=-i[\underline{A}*\underline{F},\,], \tag{4.5}$$

where the symbols [,] denote Poisson brackets. The Liouville equation is

$$\frac{\partial f(X,t)}{\partial t}=-iL_T(t)f(X,t). \tag{4.6}$$

The formal solution of this equation is

$$f(X,t)=T_+\exp\left(-i\int_{-\infty}^{t}dt_1\,L_T(t_1)\right)f^0(X,-\infty), \tag{4.7}$$

where $-\infty$ is the time when the forces have been switched on. We take here $f^0(X,-\infty)$ to be a stationary-state distribution function.

Equation (4.7) can be written

$$f(X,t)=\left[\exp\left(-\int_{\infty}^{t}L_0\,d\tau\right)-\int_{-\infty}^{t}d\tau\exp[-iL_0(t-\tau)]\,iL_1(\tau)T_+\exp\left(-i\int_{-\infty}^{\tau}d\sigma\,L_T(\sigma)\right)\right]f^0(X,-\infty)$$
$$=f^0(X,t)-\int_{-\infty}^{t}d\tau\exp[-iL(t-\tau)]\,iL_1(\tau)f(X,\tau). \tag{4.8}$$

Here $f^0(X,t)$ is the true distribution at time t, if the forces were never switched on. One must remember here that our stationary state is not an equilibrium state, and

$$e^{-iL_0t}f^0(X,-\infty)\neq f^0(X,-\infty). \tag{4.9}$$

Equation (4.8) can be iterated, and, if we retain terms up to linear order in the forces, we find

$$f(X,t)=f^0(X,t)-\int_{-\infty}^{t}e^{-iL_0(t-\tau)}iL_1(\tau)f^0(X,\tau)d\tau$$
$$=f^0(X,t)+\int_{-\infty}^{t}d\tau\,e^{-iL_0(t-\tau)}$$
$$\times[f^0(\tau),\underline{A}(\vec{r}_1)]*\underline{F}(\vec{r}_1,\tau), \tag{4.10}$$

where we have used Eq. (4.5).

It is possible now to find the after-effect function. The response of $\underline{A}$ due to the forces is

$$\mathrm{Tr}\{\underline{A}(r)[f(X,t)-f^0(X,t)]\}$$
$$=\int_{-\infty}^{t}d\tau\,\mathrm{Tr}(\underline{A}(r)\{e^{-iL_0(t-\tau)}[f^0(X,\tau),\underline{A}(\vec{r}_1)]\})$$
$$*\underline{F}(\vec{r}_1,\tau). \tag{4.11}$$

Thus the NESS after-effect function is

$$\phi_{\underline{AA}}=-\mathrm{Tr}(\underline{A}(\vec{r})\{e^{-iL_0(t-\tau)}[f^0(X,\tau),\underline{A}(\vec{r}_1)]\}). \tag{4.12}$$

We can clarify now the differences between the situations in equilibrium and here. In the equilibrium case (take a canonical ensemble, for example) $f^0(\tau)$ is a function of the Hamiltonian H_0 only. Poisson brackets appearing there can be transformed as follows[10]:

$$[f_{\mathrm{eq}}(H_0),\underline{A}(\vec{r}_1)]=\frac{\partial f_{\mathrm{eq}}(H_0)}{\partial H_0}[H_0,\underline{A}(\vec{r}_1)]$$
$$=\frac{-1}{k_BT}f_{\mathrm{eq}}\mathring{\underline{A}}(\vec{r}_1). \tag{4.13}$$

The after-effect function becomes then

$$\phi^{\mathrm{eq}}_{\underline{AA}}=\frac{-1}{k_BT}\langle\dot{\underline{A}}(\vec{r},t)\underline{A}(\vec{r}_1)\rangle, \tag{4.14}$$

which, upon Fourier transforming and taking the imaginary part, yields

$$\chi''_{\underline{AA}}\propto\omega\langle\underline{A}_{\vec{k}\omega}\underline{A}_{-\vec{k}}\rangle,$$

which, in conjunction with Eq. (4.4), yields the fluctuation-dissipation theorem. The situation here is different. The quantity $f^0(X,t)$ is *not* a function of H_0 only, and therefore transformation (4.13) is not possible. We may transform $\phi_{AA'}$, however, to a more familiar form by integrating by parts the phase-space integral, finding

$$\phi_{\underline{AA}}=\mathrm{Tr}\{f^0(X,\tau)[\underline{A}(\vec{r},t-\tau),\underline{A}(\vec{r}_1)]\}. \tag{4.15}$$

B. Dissipation

We consider here the extra dissipation (beyond the spontaneous, constant dissipation that exists in the NESS) that is due to the forces. From Eq. (4.11) and (4.15) we see that the response in any variable A_α, $\delta a_\alpha(\vec{r},t)$, can be written as

$$\delta a_\alpha(r,t)=\int_{-\infty}^{t}d\tau\,\phi_{\alpha\gamma}(\vec{r}|\vec{r}_1;t-\tau)*F_\gamma(\vec{r}_1,\tau), \tag{4.16}$$

where repeated Greek indices are to be summed upon and stationarity has been used. The dissipation rate is $Q(t)=dH_T/dt=-\underline{A}*\partial\underline{F}(\vec{r},t)/\partial t$. The macroscopic dissipation is

$$\overline{Q}(t)=-[\langle\underline{A}(\vec{r})\rangle_{\mathrm{NESS}}+\delta\underline{a}(\vec{r},t)]*\frac{\partial\underline{F}(\vec{r},t)}{\partial t}. \tag{4.17}$$

Choosing a monochromatic force, i.e.,

$$\underline{F}(\vec{r},t)=\mathrm{Re}[\underline{F}(\vec{r})e^{-i\omega t}], \tag{4.18}$$

we find the average of the dissipation over one cycle:

$$\bar{\bar{Q}}_\omega = \frac{\omega}{2\pi}\int_0^{2\pi/\omega} \bar{Q}(t)dt$$
$$= \frac{\omega}{2\pi}\int_0^{2\pi/\omega} dt \int_{-\infty}^{t} d\tau\, \phi_{\alpha\gamma}(\vec{r}|\vec{r}_1; t-\tau)$$
$$\ddagger \mathrm{Re}[F_\gamma(\vec{r}_1)e^{-i\omega\tau}]\mathrm{Re}[i\omega F_\alpha(\vec{r})e^{-i\omega t}]. \quad (4.19)$$

Equation (4.19) is simplified by writing $\mathrm{Re}[F_\gamma(\vec{r}_1)e^{-i\omega\tau}]$ as $\frac{1}{2}[F_\gamma(\vec{r}_1)e^{-i\omega\tau}+\text{c.c.}]$, $\mathrm{Re}[iF_\alpha(\vec{r})e^{-i\omega\tau}]$ as $(-1/2i)[F_\alpha(\vec{r}_1)e^{-i\omega\tau}-\text{c.c.}]$. Multiplying these two factors together, changing the variable of integration to $s=t-\tau$, and then performing the integration over t, we obtain the result

$$\bar{\bar{Q}}_\omega = -\tfrac{1}{2}\omega\, \mathrm{Im}\int_0^\infty ds\, e^{-i\omega s}\phi_{\alpha\gamma}(\vec{r}|\vec{r}_1; s)^*_* F^*_\gamma(\vec{r}_1)F_\alpha(\vec{r}), \quad (4.20)$$

where the superscript * denotes complex conjugation. Defining in the usual fashion

$$\chi_{\alpha\gamma}(\vec{r}|\vec{r}_1;\omega) = \int_0^\infty ds\, e^{-i\omega s}\phi_{\alpha\gamma}(\vec{r}|\vec{r}_1; s), \quad (4.21)$$

we find

$$\bar{\bar{Q}}_\omega = \tfrac{1}{2}\omega\, \mathrm{Im}[F_\alpha(\vec{r})*\chi_{\alpha\gamma}(\vec{r}|\vec{r}_1;\omega)*F^*_\gamma(\vec{r}_1)]. \quad (4.22)$$

As is commonly done in equilibrium theories,[7] the space dependence of the force is chosen to be a plane wave:

$$F_\alpha(\vec{r}) = e^{i\vec{k}\cdot\vec{r}}F_\alpha. \quad (4.23)$$

Thus

$$\bar{\bar{Q}}_{\vec{k}\omega} = -\tfrac{1}{2}\omega\, \mathrm{Im}\int d\vec{r}\, d\Delta\vec{r}\, e^{i\vec{k}\cdot\Delta\vec{r}}\chi_{\alpha\gamma}(\vec{r}|\vec{r}-\Delta\vec{r};\omega)F_\alpha F^*_\gamma$$
$$= -\tfrac{1}{2}\omega\, \mathrm{Im}\int d\vec{r}\, \chi_{\alpha\gamma}(\vec{k},\omega|\vec{r})F_\alpha F^*_\gamma. \quad (4.24)$$

It is natural to define the local dissipation as

$$\bar{\bar{Q}}_{\vec{k}\omega}(\vec{r}) = -\tfrac{1}{2}\omega\, \mathrm{Im}\chi_{\alpha\gamma}(\vec{k},\omega|\vec{r})F_\alpha F^*_\gamma. \quad (4.25)$$

Clearly, if the system is translationally invariant, Eq. (4.25) reduces to the usual equilibrium result, and $\bar{\bar{Q}}_{\vec{k}\omega}(\vec{r}) = (1/V)\bar{\bar{Q}}_{\vec{k}\omega}$.

C. Relation between dissipation and fluctuations

The after-effect function for systems in NESS was found in Sec. IV A to be $\langle[\underline{A}(\vec{r},s),\underline{A}(\vec{r}_1)]\rangle_{\text{NESS}}$. The theory of Sec. II (and papers I–III) can be used to evaluate this quantity for NESS whose spatial gradients are not too large. Using the methods of Sec. IV A 3 of I, we can modify Eq. (2.23) to read in this case

$$\phi_{\alpha\alpha}(\vec{r}|\vec{r}_1;s) = \langle[A_\alpha(\vec{r},s),A_\alpha(\vec{r}_1)]\rangle_L^{\text{hom}}(\vec{r})$$
$$-\int_0^\infty d\tau\langle[A_\alpha(\vec{r},s),A_\alpha(\vec{r}_1)]$$
$$\times \underline{I}_T(-\tau)\rangle_L^{\text{hom}}(\vec{r})\cdot\vec{\nabla}\beta\underline{\Phi}(\vec{r}), \quad (4.26)$$

where $\underline{I}_T \equiv \int \underline{J}_D(\vec{r})d\vec{r}$.

The superscript "hom" is a prescription for evaluating the correlation function in a uniform system whose $\beta\underline{\Phi} = \beta\underline{\Phi}(\vec{r})$. In other words, it is computed with the distribution function

$$f_L^{\text{hom}}(X) = f_{\text{GC}}\exp[\beta\underline{\Phi}(\vec{r})\cdot\underline{A}_T]\Big/\left(\sum_N \int dX f_{\text{GC}}\exp[\beta\underline{\Phi}(\vec{r})\cdot\underline{A}_T]\right)^{-1}, \quad (4.27)$$

where the subscript T stands for the variable "integrated over all space" (see I for more details).

Denoting the first term on the RHS of Eq. (4.26) by $\phi^L_{\alpha\alpha}$, one may readily verify that

$$\phi^L_{\alpha\alpha}(\vec{r},\vec{r}_1;s)$$
$$= -\beta(\vec{r})\langle A_\alpha(\vec{r},s)[A_\alpha(\vec{r}_1),H_0-\vec{P}_T\cdot\vec{v}(\vec{r})]\rangle_L^{\text{hom}}(\vec{r}). \quad (4.28)$$

Fourier transforming in space and time according to

$$A_{\alpha,\vec{k},\omega} = \int dr\, e^{i\vec{k}\cdot\vec{r}}\int_0^\infty dt\, e^{-i\omega t}A_\alpha(\vec{r},t), \quad (4.29)$$

we find

$$\chi^L_{\alpha\alpha}(\vec{k},\omega|\vec{r})$$
$$= -(\beta(\vec{r})/V)\langle A_{\alpha,\vec{k},\omega}[A_{\alpha,-\vec{k}},H_0-\vec{P}_T\circ\vec{v}(\vec{r})]\rangle_L^{\text{hom}}(\vec{r}). \quad (4.30)$$

In Appendix B we prove that the imaginary part of $\chi^L_{\alpha\alpha}$ is

$$[\chi^L_{\alpha\alpha}(\vec{k},\omega|\vec{r})]''$$
$$= (1/V)[\omega-\vec{k}\cdot\vec{v}(\vec{r})]\beta(\vec{r})\mathrm{Re}\langle A_{\alpha,\vec{k},\omega}A_{\alpha,-\vec{k}}\rangle_L^{\text{hom}}(\vec{r}). \quad (4.31)$$

The term proportional to $\vec{k}\cdot\vec{v}(\vec{r})$ is of course the Doppler shift seen in light scattering from convecting systems. In order to make our point more simply, we consider now a NESS without convection, that is, driven by a single force (F_α).

Further, we decompose $\bar{\bar{Q}}(\vec{r})$ itself into two contributions, $\bar{\bar{Q}}^L(\vec{r})$ and $\bar{\bar{Q}}^D(\vec{r})$, where $\bar{\bar{Q}}^L(\vec{r})$ corresponds to the contribution of $\chi^L(\vec{k},\omega|\vec{r})$ and $\bar{\bar{Q}}^D(\vec{r})$ stands for the rest. Comparing Eq. (4.31) with Eq. (4.25), we may conclude that the space-dependent local equilibrium part of the dissipation measures the local equilibrium part of the time correlation function, in much the same way as in equilibrium:

$$\overline{\overline{Q}}^{L}_{\vec{k}\omega}(\vec{r}) = -\tfrac{1}{2}\omega^2 \,\mathrm{Re}[\langle A_{\alpha,\vec{k},\omega} A_{\alpha,-\vec{k}}\rangle_L^{\mathrm{hom}}(\vec{r})/V] \times\beta(\vec{r})|F_\alpha(\vec{k},\omega)|^2 . \qquad (4.32)$$

From the discussion given in Sec. III we see that for small k and ω the LHS of Eq. (4.32) may be computed by using the regression hypothesis. This approach is standard[3,7,9] and gives

$$\overline{\overline{Q}}^{L}_{\vec{k}\omega}(\vec{r}) = (-\omega^2/2V)\beta(\vec{r})\,|F_\alpha(\vec{k},\omega)|^2 \times \mathrm{Re}\{[1/(i\omega\underline{\underline{1}} - \underline{\underline{M}}_{\vec{k}})]\cdot\langle \underline{A}_{\vec{k}}\underline{A}_{-\vec{k}}\rangle\}^{\mathrm{hom}}_{\alpha\alpha}(\vec{r}) . \qquad (4.33)$$

The remaining term in the expression for the dissipation is related by Eq. (4.25) to the second term on the RHS of Eq. (4.26). We define

$$\begin{aligned}\chi^{D}_{\alpha\gamma}(\vec{k},\omega|\vec{r}) &\equiv -\frac{1}{V}\int_0^\infty ds\, e^{-i\omega s} \times \int_0^\infty d\tau \langle [A_{\alpha,\vec{k}}(s), A_{\gamma,-\vec{k}}]\underline{I}_T(-\tau)\rangle_L^{\mathrm{hom}}(\vec{r})\circ\vec{\nabla}\beta\underline{\Phi}(\vec{r})\\ &= -\frac{1}{V}\int_0^\infty ds\, e^{-i\omega s} \times \int_0^\infty d\tau\langle [A_{\alpha,\vec{k}}(s+\tau), A_{\gamma,-\vec{k}}(\tau)]\underline{I}_T\rangle_L^{\mathrm{hom}}(\vec{r}) \circ\vec{\nabla}\beta\underline{\Phi}(\vec{r}) ,\end{aligned} \qquad (4.34)$$

where the second equality follows from the invariance of the Poisson bracket to canonical transformations.[11] Hence for the case at hand,

$$\overline{\overline{Q}}^{D}_{\vec{k}\omega}(\vec{r}) = -\tfrac{1}{2}\omega|F_\alpha(\vec{k},\omega)|^2[\chi^{D}_{\alpha\alpha}(\vec{k},\omega|\vec{r})]'' . \qquad (4.35)$$

In Appendix C we show that for the hydrodynamic variables

$$\chi^{D}_{\alpha\gamma}(\vec{k},\omega|\vec{r}) = -\left[\left(\frac{1}{i\omega\underline{\underline{1}} - \underline{\underline{M}}_{\vec{k}}}\right)_{\alpha E}\frac{i}{V}\vec{k}\cdot\int_0^\infty dt_1\langle I_{\gamma,T}(t_1)I_{\gamma,T}\rangle \cdot\vec{\nabla}\beta\underline{\Phi}_\gamma(\vec{r})\right]^{\mathrm{hom}}(\vec{r}),$$

$$\gamma = E, P ,\quad \chi^{D}_{\alpha\gamma}(\vec{k},\omega|\vec{r}) = 0 ,\quad \gamma = N , \qquad (4.36)$$

to leading order in k. Recognizing the time integral appearing in the expression above as the Green-Kubo form for the Onsager coefficients,[7,9] it is a simple matter to show that for systems under thermal constraint, for $\vec{k}$ in the light scattering regime, and for small macroscopic gradients,

$$\overline{\overline{Q}}^{D}_{\vec{k}\omega}(\vec{r})/\overline{\overline{Q}}^{L}_{\vec{k}\omega}(\vec{r}) \sim [\vec{k}\circ\vec{\nabla}\ln\beta(\vec{r})]/k^2 \lesssim 10^{-4} , \qquad (4.37)$$

which follows by using Eqs. (4.36), (4.35), and (4.33). A similar estimate holds for the case in which convection is present and hence the contribution of the dissipative part of the aftereffect function to the average dissipation is negligible, and so

$$\overline{\overline{Q}}_{\vec{k}\omega}(\vec{r}) \approx \overline{\overline{Q}}^{L}_{\vec{k}\omega}(\vec{r}) . \qquad (4.38)$$

In other words, a *measurement of the dissipation resulting from perturbing a NESS yields only the local "hom" part of a time correlation function.* Hence none of the new effects predicted in II and III would be seen in this fashion, since the full NESS time correlation function is not involved in the dissipation. In this sense the fluctuation-dissipation theorem is less useful in NESS than in equilibrium states, since it gives only partial information about the fluctuations.

In fact, from Appendix B [cf. Eq. (B5)] we see that the information contained in the NESS dissipation is rather simply connected to equilibrium quantities which are themselves directly measurable in an equilibrium-dissipation measurement.

A couple of remarks concerning Eq. (4.38) are in order. In the first place, the light scattering predictions made in Refs. 1–4 are consistent with the fluctuation-dissipation theorem as given above. This is true since the dissipative contribution to the NESS time correlation function gives zero when integrated over frequencies and angles. Thus it does not contribute to the total scattering intensity (i.e., all angles and frequencies). This means that the total scattering intensity (which is simply related to the dissipation) involves only the local part of the NESS time correlation function as shown above. It will be seen in our future publications that the same remark pertains to light scattering from NESS with a velocity field. Although the spectrum is changed considerably compared with local equilibrium, the integrated effect (over all k and ω) contains only the local equilibrium contributions.

The second remark is that Eq. (4.38) is not really a surprising result. We have all along been restricted to wave vectors k larger than the macroscopic gradients. In this regime the terms in the hydrodynamic equations proportional to the $\vec{\nabla}\beta\underline{\Phi}_{ss}$ are completely negligible. Hence the dissipation could be computed by solving the hydrodynamic equations in a system subjected to some monochromatic disturbance, all the while neglecting the explicit $\vec{\nabla}\beta\underline{\Phi}_{ss}$ terms. For small perturbations the dissipation computed in this fashion will equal $\bar{\bar{Q}}^{L}_{\vec{k}\omega}$ as the hydrodynamics is the same as in equilibrium.

V. DISCUSSION

We discuss first the significance of the generalization to all orders in $\beta\underline{\Phi}$ with the restriction that the final result, Eq. (2.22), is valid to first order in $\vec{\nabla}\beta\underline{\Phi}$. This generalization is important, since macroscopic systems tend to be fairly large (think about the ocean, for example, . . .) and even with very small gradients one can achieve large deviations from equilibrium if the system is large enough. The level at which the theory pertains now is similar to the level of the first-order "ϵ expansion" in the Chapman-Enskog method. Namely, the reference state is a local equilibrium state and not an equilibrium one, and the smallness parameter is the spatial variation of the local properties and not the deviation from equilibrium.

We have proven the regression hypotheses for the NESS correlation functions. This result is fairly important, since Eq. (3.29) can be cast into the form

$$\langle \underline{\tilde{A}}(\vec{r},t)\underline{\tilde{A}}(\vec{r}_1)\rangle_{\text{NESS}} = e^{\underline{\underline{M}}_{ss}(\vec{r}|\vec{r}_2)t} * \langle \underline{\tilde{A}}(\vec{r}_2)\underline{\tilde{A}}(\vec{r}_1)\rangle_{\text{NESS}} \;, \tag{5.1}$$

which means that once the linearized (macroscopic) equations of motion are known, the computation of NESS time correlation functions reduces to the calculation of the static-correlation functions. As was shown in III, the static correlations themselves can be evaluated once $\underline{\underline{M}}_{ss}$ is known. We stress here that one must be extremely careful not to employ the local equilibrium value of the static correlation functions, since this leads to important omissions. A complete calculation of the type suggested in III (or II) is needed. Another word of caution: Eq. (5.1) is valid only for positive time, and time-reversal symmetry is broken in the NESS. The method used to extend Eq. (5.1) to negative time should be based on the stationarity property of the correlation functions (cf. III).

The examination of the fluctuation-dissipation theorem gave the following results: The dissipation is related to the local equilibrium part of the fluctuation and not to the true fluctuation. Thus, when the difference between the local part and the complete form of the fluctuation is large, (as it is for small k's), the fluctuation-dissipation theorem does not hold. In other words, the dissipation does not contain all the information about the fluctuations. Our explicit calculations of NESS correlation functions agree with these results. In the two cases of a NESS with a temperature gradient[3] and with a velocity field[12] we found that the spectra of light scattered from a NESS differed considerably from equilibrium spectra. However the total scattered light obtained by integrating the differential intensity over all angles and frequencies was completely determined by the local equilibrium part of the fluctuations alone.

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APPENDIX A: PROOF OF EQ. (3.8)

Consider the quantity

$$\langle \underline{\dot{A}}(\vec{r},t)\underline{\tilde{A}}(\vec{r}_1)\rangle_{L,ss} * \langle \underline{\tilde{A}}(\vec{r}_1,t)\underline{\tilde{A}}(\vec{r}\,')\rangle^{-1}_{L,ss} \,. \tag{A1}$$

Any function $f(t)$ can be written as $f(0)+\int_0^t f(\tau)d\tau$. The above quantity is written as

$$\langle \underline{\dot{A}}(\vec{r})\underline{\tilde{A}}(\vec{r}_1)\rangle_{L,ss} * \langle \underline{\tilde{A}}(\vec{r}_1)\tilde{A}(\vec{r}\,')\rangle^{-1}_{L} + \int_0^t \Big[\langle \underline{\ddot{A}}(\vec{r},\tau)\underline{\tilde{A}}(\vec{r}_1)\rangle_{L,ss} * \langle \underline{\tilde{A}}(\vec{r}_1,\tau)\underline{\tilde{A}}(\vec{r}\,')\rangle^{-1}_{L,ss} + \langle \underline{\dot{A}}(\vec{r},\tau)\underline{\tilde{A}}(\vec{r}_1)\rangle_{L,ss} * \frac{\partial}{\partial\tau}\langle \underline{\tilde{A}}(\vec{r}_1,\tau)\tilde{A}(\vec{r}\,')\rangle^{-1}_{L,ss}\Big]d\tau \,. \tag{A2}$$

Since

$$\langle \underline{\tilde{A}}(\vec{r},t)\underline{\tilde{A}}(\vec{r}_1)\rangle^{-1}_{L,ss} * \langle \underline{\tilde{A}}(\vec{r}_1,t)\underline{\tilde{A}}(\vec{r}\,')\rangle_{L,ss} = \underline{\underline{1}}\delta(\vec{r}-\vec{r}\,'),$$

$$\langle \underline{\mathring{A}}(\vec{r}_1,\tau)\underline{\tilde{A}}(\vec{r}\,')\rangle^{-1}_{L,ss} = -\langle \underline{\tilde{A}}(\vec{r}_1,\tau)\underline{\tilde{A}}(\vec{r}_2)\rangle^{-1}_{L,ss} * \langle \underline{\dot{A}}(\vec{r}_2,\tau)\underline{\tilde{A}}(\vec{r}_3)\rangle_{L,ss} * \langle \underline{\tilde{A}}(\vec{r}_3,\tau)\underline{\tilde{A}}(\vec{r}\,')\rangle^{-1}_{L,ss} \,. \tag{A3}$$

Substituting Eq. (A3) in Eq. (A2), we find, after some trivial algebra,

$$\langle \underline{\dot{A}}(\vec{r},t)\underline{\tilde{A}}(\vec{r}_1)\rangle_{L,ss} * \langle \underline{\tilde{A}}(\vec{r}_1,t)\underline{\tilde{A}}(\vec{r}\,')\rangle^{-1}_{L,ss} = \langle \underline{\dot{A}}(\vec{r})\underline{\tilde{A}}(\vec{r}_1)\rangle_{L,ss} * \langle \underline{\tilde{A}}(\vec{r}_1)\underline{\tilde{A}}(\vec{r}\,')\rangle^{-1}_{L,ss} + \int_0^t \left\langle \frac{\partial}{\partial\tau}\underline{\mathring{A}}_D(\vec{r},\tau)\underline{\tilde{A}}(\vec{r}_1)\right\rangle_L * \langle \underline{\tilde{A}}(\vec{r}_1,\tau)\underline{\tilde{A}}(\vec{r}\,')\rangle^{-1}_{L,ss}\, d\tau \,. \tag{A4}$$

Using the rule for dot switching [Eq. (3.14)] and noticing that $\langle \underline{A}(\vec{r},t)\underline{\tilde{A}}(\vec{r}_1)\rangle^{-1}_{L,ss}$ is slowly varying in time, we conclude that the last expression is nothing but $\underline{\underline{M}}_{ss}(\vec{r}|\vec{r}\,')$.

APPENDIX B: PROOF OF EQ. (4.31)

The form of $\chi^L_{\alpha\alpha}(\vec{k},\omega|\vec{r})$ is

$$\chi^L_{\alpha\alpha}(\vec{k},\omega|\vec{r}) = -\frac{\beta(\vec{r})}{V}\langle A_{\alpha,\vec{k},\omega}[A_{\alpha,-\vec{k}},H_0-\vec{P}_T\cdot\vec{v}(\vec{r})]\rangle_L^{\text{hom}}(\vec{r}). \tag{B1}$$

It can be rewritten as

$$\chi^L_{\alpha\alpha}(\vec{k},\omega|\vec{r}) = -\beta(\vec{r})\frac{1}{V}\left(\langle A_{\alpha,\vec{k},\omega}\dot{A}_{\alpha,-\vec{k}}\rangle_L^{\text{hom}}(\vec{r}) - \left\langle A_{\alpha,\vec{k},\omega}\sum_j\frac{\partial}{\partial\vec{r}_j}A_{\alpha,-\vec{k}}\right\rangle_L^{\text{hom}}(\vec{r})\cdot\vec{v}(\vec{r})\right), \tag{B2}$$

where the sum extends over all the particles in the system and $\vec{r}_j$ is the position of the jth particle.

In the "hom" average we can dot switch (cf. Sec. IIIB) and rewrite Eq. (B2) as

$$\chi^L_{\alpha\alpha}(\vec{k},\omega|\vec{r}) = \frac{\beta(\vec{r})}{V}\left(i\omega\langle A_{\alpha,\vec{k},\omega}A_{\alpha,-\vec{k}}\rangle_L^{\text{hom}}(\vec{r}) - \langle A_{\alpha,\vec{k}}A_{\alpha,-\vec{k}}\rangle_L^{\text{hom}}(\vec{r}) + \vec{v}(\vec{r})\cdot\left\langle A_{\alpha,\vec{k},\omega}\sum_j\frac{\partial}{\partial\vec{r}_j}A_{\alpha,-\vec{k}}\right\rangle_L^{\text{hom}}(\vec{r})\right). \tag{B3}$$

It is easy to verify that

$$\sum\frac{\partial}{\partial\vec{r}_j}A_{\alpha,-\vec{k}} = -i\vec{k}A_{\alpha,-\vec{k}},$$

and thus

$$[\chi^L_{\alpha\alpha}(\vec{k},\omega|\vec{r})]'' = [\omega-\vec{k}\cdot\vec{v}(\vec{r})]V^{-1}\beta(\vec{r}) \times\text{Re}[\langle A_{\alpha,\vec{k},\omega}A_{\alpha,-\vec{k}}\rangle_L^{\text{hom}}(\vec{r})]. \tag{B4}$$

Further, for the important case in which $\alpha=N$, introducing the Gallilean transformation $\vec{p}_j\to\vec{p}_j+m(\vec{v})_{\text{NE}}$ allows Eq. (B4) to be rewritten as

$$[\chi^L_{NN}(\vec{k},\omega|\vec{r})]'' = \{[\omega-\vec{k}\cdot\vec{v}(\vec{r})]/V\} \times\beta(\vec{r})\langle N_{\vec{k},\omega-\vec{k}\cdot\vec{v}(\vec{r})}N_{-\vec{k}}\rangle|_{T=T(\vec{r}),\ \mu=\mu(\vec{r})+1/2mv2(\vec{r})}, \tag{B5}$$

where we have used the fact that $\langle N_{\vec{k},\omega}N_{-\vec{k}}\rangle$ is real. In the same manner it can be shown that a relationship like that given in Eq. (B5) holds whenever $A_{\alpha,\vec{k}}$ does not explicitly depend on the particle's momentum. Should this not be the case, $\langle A_{\alpha,\vec{k},\omega}A_{\alpha,-\vec{k}}\rangle^{\text{hom}}$ will contain terms explicitly proportional to $\vec{v}(\vec{r})$.

APPENDIX C: PROOF OF EQ. (4.36)

For equilibrium correlations it is useful to write

$$\underline{A}_{\vec{k}}(t) = e^{\underline{\underline{M}}_{\vec{k}}t}\cdot\underline{A}_{\vec{k}}(0) + \int_0^t dt_1\, e^{\underline{\underline{M}}_{\vec{k}}(t-t_1)}\underline{I}_{\vec{k}}(t_1)\cdot i\vec{k}, \tag{C1}$$

which is valid for $t>\tau_D$ and where $\underline{\underline{M}}_{\vec{k}}$ is given by Eq. (3.24) evaluated at equilibrium. The set $A_{\vec{k}}$ is taken to be the set of hydrodynamic variables.

Let

$$V\phi^D_{\alpha\gamma}(\vec{k},s|\vec{r}) \equiv -\int_0^\infty d\tau\langle[A_{\alpha,\vec{k}}(s+\tau),A_{\gamma,-\vec{k}}(\tau)]\underline{I}_T\rangle_L^{\text{hom}}(\vec{r})\cdot\vec{\nabla}\beta\Phi(\vec{r}); \tag{C2}$$

then using (C1) we have

$$\begin{aligned} V\phi^D_{\alpha\gamma}(\vec{k},s|\vec{r}) = -\int_0^\infty d\tau\Big(& (e^{\underline{\underline{M}}_{\vec{k}}(s+\tau)})_{\alpha\alpha'}(e^{\underline{\underline{M}}_{\vec{k}}\tau})^*_{\gamma\gamma'}\langle[A_{\alpha',\vec{k}},A_{\gamma',-\vec{k}}]\underline{I}_T\rangle\cdot\vec{\nabla}\beta\underline{\Phi}(\vec{r}) \\ & + \int_0^{\tau+s} dt_1(e^{\underline{\underline{M}}_{\vec{k}}(s+\tau-t_1)})_{\alpha\alpha'}(e^{\underline{\underline{M}}_{\vec{k}}\tau})^*_{\gamma\gamma'}\langle[i\vec{k}\cdot I_{\alpha',\vec{k}}(t_1),A_{\gamma',-\vec{k}}]\underline{I}_T\rangle\cdot\vec{\nabla}\beta\underline{\Phi}(\vec{r}) \\ & - \int_0^{\tau} dt_1(e^{\underline{\underline{M}}_{\vec{k}}(s+\tau)})_{\alpha\alpha'}(e^{\underline{\underline{M}}_{\vec{k}}(\tau-t_1)})^*_{\gamma\gamma'}\langle[A_{\alpha',\vec{k}},i\vec{k}\cdot I_{\gamma',-\vec{k}}(t_1)]\underline{I}_T\rangle\cdot\vec{\nabla}\beta\underline{\Phi}(\vec{r})\Big), \end{aligned} \tag{C3}$$

where the term containing two explicit factors of ik has been neglected and where the superscript "hom" has been dropped. To leading order in k, the k dependence of the variables appearing in the last two terms on the RHS of Eq. (C3) can be neglected. Further noting that

$$[I_{\alpha',T}(t_1),A_{\gamma',T}] = \dot{I}_{\alpha',T}(t_1)\delta_{\gamma',E} \tag{C4}$$

allows Eq. (C3) to be rewritten as

$$V\phi^D_{\alpha\gamma}(\vec{k},s|\vec{r}) = -\int_0^\infty d\tau\Big((e^{\underline{M}_{\vec{k}}(s+\tau)})_{\alpha\alpha'}(e^{\underline{M}_{\vec{k}}\tau})^*_{\gamma\gamma'}\langle[A_{\alpha',\vec{k}},A^*_{\gamma',\vec{k}}]\underline{I}_T\rangle + \int_0^{\tau+s} dt_1 (e^{\underline{M}_{\vec{k}}(s+\tau-t_1)})_{\alpha\alpha'}(e^{\underline{M}_{\vec{k}}\tau})^*_{\gamma E}\langle i\vec{k}\cdot\dot{I}_{\alpha',T}(t_1)\underline{I}_T\rangle + \int_0^{\tau} dt_1 (e^{\underline{M}_{\vec{k}}(s+\tau)})_{\alpha E}(e^{\underline{M}_{\vec{k}}(\tau-t_1)})^*_{\gamma\gamma'}\langle i\vec{k}\cdot\dot{I}_{\gamma',T}(t_1)\underline{I}_T\rangle\Big)\cdot\vec{\nabla}\beta\underline{\Phi}(\vec{r}). \quad (C5)$$

Performing the above integrations over t_1 by parts and neglecting terms containing more than one explicit factor of k (remember $M_{\vec{k}}\sim\vec{k}$), we find that

$$V\phi^D_{\alpha\gamma}(\vec{k},s|\vec{r}) = -\int_0^\infty d\tau\Big((e^{\underline{M}_{\vec{k}}(s+\tau)})_{\alpha\alpha'}(e^{\underline{M}_{\vec{k}}\tau})^*_{\gamma\gamma'}\{\langle[A_{\alpha',\vec{k}},A_{\gamma',-\vec{k}}]\underline{I}_T\rangle - \delta_{\gamma',E}\langle i\vec{k}\cdot I_{\alpha',T}\underline{I}_T\rangle - \delta_{\alpha,E}\langle i\vec{k}\cdot I_{\gamma',T}\underline{I}_T\rangle\} + (e^{M_{\vec{k}}\tau})^*_{\gamma,E}\langle i\vec{k}\cdot I_{\alpha',T}(\tau+s)\underline{I}_T\rangle + (e^{M_{\vec{k}}(s+\tau)})_{\alpha E}\langle i\vec{k}\cdot I_{\gamma,T}(\tau)\underline{I}_T\rangle\Big)\cdot\vec{\nabla}\beta\underline{\Phi}(\vec{r}). \quad (C6)$$

Using the explicit forms of the hydrodynamic variables,[9] it is straightforward to show that the terms in braces in Eq. (C6) cancel to $O(k^2)$. Since the time correlation function $\langle I(t)I\rangle$ is assumed to decay to zero when $t>\tau_D$, this implies that the next-to-last term on the RHS of (C6) may be dropped for $s>\tau_D$. Finally, putting τ to zero in $e^{\underline{M}_{\vec{k}}(s+\tau)}$, which appears in the last term, results in

$$V\phi^D_{\alpha\gamma}(\vec{k},s|\vec{r}) = -(e^{\underline{M}_{\vec{k}}s})_{\alpha E}\int_0^\infty d\tau\, i\vec{k}\cdot\langle I_{\gamma,T}(\tau)I_T\rangle\cdot\vec{\nabla}\beta\underline{\Phi}(\vec{r}), \quad (C7)$$

which, on taking the half-sided Fourier transform and using the forms of the $I_{\gamma,T}$, gives Eq. (4.36).

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