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Asymptotic Time Behavior of Correlation Functions. III. Local Equilibrium and Mode-Coupling Theory

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The decay to equilibrium is discussed from a general point of view based on the assumed rapid approach to local equilibrium for well-chosen initial states. The assumption is applied to the problem of time correlation functions and it is shown that the mode-coupling formula describes the asymptotics of the so-called projected wavenumber-dependent correlation functions. The local equilibrium assumption thus provides a general basis for the $t^{-3/2}$ behavior of correlation functions derived in previous papers in this series, as well as for the infinite series of correction terms $t^{-(2-Pn)}$ $(n \geqslant 2)$, with $Pn = 2^{-n}$, and for the corresponding series of corrections of order k^{3-Pn} $(n \geqslant 1)$ to Navier–Stokes hydrodynamics.

KEY WORDS: Nonequilibrium statistical mechanics; local equilibrium assumptions; generalized hydrodynamics; Green–Kubo formulas; mode-coupling formulas; long-time tails.

1. INTRODUCTION

In 1970 Alder and Wainwright$^{(1)}$ published strong computer evidence for the existence of a tail in the velocity correlation function, decaying like an inverse power of time. This remarkable discovery triggered a multitude of theoretical studies[4] on the asymptotic behavior of the general class of time correlation functions appearing in the Green–Kubo formulas (see, e.g., Ref. 3) for the transport coefficients.

Several different lines of argument have been pursued. The most fundamental one, that of generalized kinetic theory, was initiated by Dorfman and Cohen.$^{(4)}$ They derived a low-density expression for the tails in Green–Kubo

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[4] See Ref. 2 for a recent review.

integrands, and extending the results to higher densities in the spirit of the Enskog theory, they were able to reproduce the computer data, amplitudes included. Generalizations of these results in several directions using kinetic theory have since appeared.[5]

A formal kinetic theory for arbitrary densities has recently been presented by Résibois *et al.*(9) Exploiting certain plausible assumptions, which are checked for self-consistency, they use diagrammatic techniques in a derivation of the asymptotic validity of the so-called mode-coupling formula for the correlation functions. Their argument is sufficiently general to provide a basis not only for the leading asymptotic tails, but also for an infinite series of closely spaced corrections, discussed by Pomeau(10) and by Ernst and Dorfman.(11)

Other approaches,(2) which we will not consider here, are based on fluctuating hydrodynamics, on hydrodynamics generalized to include product modes, or on the nonlinear Boltzmann equation.

In the present series of papers[6] we discuss the asymptotics of the time correlation functions on the basis of one crucial *local equilibrium assumption*: that of a (relatively) fast approach of a carefully constructed initial nonequilibrium ensemble describing a local fluctuation to a state close to local equilibrium. By a "local fluctuation" in this context we mean a fluctuation taking place in a region which is large compared to a characteristic microscopic size, taken to be the equilibrium correlation length ξ, but small compared to the macroscopic size. The concept "rapidly" is not quantitatively defined. It essentially means faster than the slow processes which we keep. One may well ask why such a phenomenological point of view can be of interest at the present time, considering the recent results obtained by the more fundamental approach of generalized kinetic theory. The reason is the following: The derivations from first principles suffer from a high degree of complexity. They involve resummations over infinite subclasses of diagrams, complicated arguments on the magnitude of the remainder, etc. Furthermore, the arguments have not yet attained the impeccable status of mathematical rigor. It seems therefore meaningful to parallel the diagrammatic approach by a phenomenological one, which, although still complex, has the virtue of relative simplicity in addition to focusing directly on the physical mechanism involved.

In paper I(12) we considered the kinetic terms of the time correlation functions. For those terms the local equilibrium assumption was only needed

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[5] See, e.g., Refs. 5–8. A more complete set of references is given in Ref. 2.

[6] See Ref. 12 (referred to hereafter as I; see also Ref. 13) and Ref. 14 (referred to as II; see also Ref. 15).

on the level of the one-particle distribution function. When potential terms are included, as in paper II,(14) an assumption on distributions in Γ-space is required. A discussion of local equilibrium ideas on the Γ-space level was not given in II, however. Instead, the calculations were, without detailed motivation, based directly on the mode-coupling formula.(16)

The function of the present paper is to derive the mode-coupling formula for the asymptotic time dependence of correlation functions on the basis of the local equilibrium assumption. We shall also try to put the local equilibrium ideas into a more general perspective. To that end we recall, in Section 3 (after having defined notation in Section 2), the derivation of generalized hydrodynamics from the local equilibrium assumption. The arguments, which are in the spirit of the Chapman–Enskog development of the Boltzmann equation, provide a necessary background for the subsequent discussion of the correlation functions, especially in Section 4.

In addition to the correlation functions which enter in the Green–Kubo formulas, we consider a more general class of so-called projected time correlation functions $U_{\mathbf{k}}(t)$, where the currents are wavelength dependent, and where the time dependence is not described by the full Liouville propagator $\exp(-t\mathscr{L})$ but by a projected operator $\exp(-tP_{\perp}\mathscr{L})$. We shall argue, in Section 4, that a consistent use of the assumption on the approach to local equilibrium, necessary for the derivation of hydrodynamics in Section 3, unambiguously shows that the mode-coupling formula describes the asymptotic time behavior of the *projected* wavelength-dependent time correlation $U_{\mathbf{k}}(t)$ and not the asymptotics of the *true* wavelength-dependent time correlation $C_{\mathbf{k}}(t)$ obtained from $U_{\mathbf{k}}(t)$ by replacing the projected Liouville propagator by the true one. This point has barely been noticed in the literature.

Sections 5 and 6 mainly focus on the delicate problem of how to decompose the initial equilibrium ensemble into components on which a local equilibrium assumption can consistently be made. In order that the subtlety of the problem can be appreciated, we have chosen a stepwise approach. In Section 5 we define a decomposition which is close in spirit to one previously proposed by Pomeau.(17) On the basis of the corresponding local equilibrium assumption, the mode-coupling formula is derived. Nevertheless, we show by a counter example that this method leads to inconsistencies, even on the two-mode level approximation.

Consequently, a refined version of the decomposition and the associated local equilibrium assumption is called for. We present it in Section 6. The mode-coupling formula is rederived, and the counterexample against the method of Section 5 is shown not to apply to the refined version.

We close, in Section 7, with an extended discussion.

2. NOTATION

The objects on which our discussion is based are the hydrodynamic variables of a simple fluid, i.e., the low Fourier components of number, momentum, and energy density. Their microscopic definitions have been given in I and II and various useful linear combinations were discussed in II. In the present paper only very general properties of the hydrodynamic variables will be used. We consequently refrain from repeating their detailed definitions here.

We shall be content with stating that it is convenient from the start to work with those linear combinations of the densities of conserved quantities that are eigenfunctions of linearized hydrodynamics. They are the hydrodynamic modes $a_{\mathbf{k}}^{i}(\Gamma)$, where the superscript labels two shear modes, two sound modes, and a heat mode. The subscript $\mathbf{k}$ is, as usual, the Fourier variable, and Γ indicates the dependence on the phase space variable (for details, see II). We shall restrict ourselves to modes with $\mathbf{k}$ values corresponding to wavenumbers below a (small) cutoff k_c, the number M of allowed k values being equal to $Vk_c^3\pi^{-3}$ (V is the volume of the system). The normalization is chosen as in II and reads, to zeroth order in k,

$$\langle a_{\mathbf{k}}^{i} a_{\mathbf{l}}^{j*}\rangle = V\,\delta_{ij}\,\delta_{\mathbf{kl}} \tag{1}$$

where the bracket denotes an average over an equilibrium grand canonical ensemble. With periodic boundary conditions in V, the average product of modes with $\mathbf{k} \neq \mathbf{l}$ automatically vanishes due to translational invariance.

In order to simplify the notation, we collect these $5M$ modes in a column vector $a(\Gamma)$. The corresponding row vector is denoted by $\tilde{a}(\Gamma)$ and its complex conjugate by $\tilde{a}^*(\Gamma) = a^\dagger(\Gamma)$. As a short-hand notation the components of $a(\Gamma)$ will, when needed, simply be written as $a_{\mathbf{k}}(\Gamma)$. The orthonormality relations (1) can now be summarized as

$$\langle aa^\dagger\rangle = V\mathbf{1} \tag{2}$$

where $\mathbf{1}$ is the $5M \times 5M$ unit matrix.

The modes were defined as Fourier components of deviations of densities from their equilibrium average. Consequently

$$\langle a\rangle = 0 \tag{3}$$

The (classical) mechanical equation of motion for $a(\Gamma)$ reads

$$da(\Gamma)/dt = \mathscr{L}a(\Gamma) \tag{4}$$

where $\mathscr{L}$ is the Liouville operator. Since $a(\Gamma)$ is the density of a conserved quantity, $-\mathscr{L}a(\Gamma)$ is the divergence of the current associated with $a(\Gamma)$. It follows that $\mathscr{L}a(\Gamma)$ vanishes as $|\mathbf{k}|$ when $\mathbf{k} \to 0$.

The local equilibrium distribution $\rho_l(\Gamma|B(t))$ in Γ space plays a crucial role in the subsequent arguments. It has the form (see, e.g., Ref. 18)

$$\rho_l(\Gamma|B(t)) = \rho_{\text{eq}}(\Gamma)\exp[a^\dagger(\Gamma)\cdot B(t)]/\langle\exp[a^\dagger\cdot B(t)]\rangle \tag{5}$$

where $\rho_{\text{eq}}(\Gamma)$ is the *total* equilibrium distribution, both ρ_{eq} and ρ_l being normalized to unity. The inner product should be interpreted as

$$a^\dagger\cdot B = \sum_{\mathbf{k},i}{}' a_{\mathbf{k}}^{i*}B_{\mathbf{k}}^{i} = B^\dagger\cdot a \tag{6}$$

with the summation over every Cartesian component of $\mathbf{k}$ restricted by $|k_{x,y,z}| < k_c$. The second equality in (6) follows from the (cubical) symmetry[7] of the region in $\mathbf{k}$ space over which we sum. The absence of a factor V^{-1} in (6) (which will prove a formal convenience later) is due to the convention chosen for Fourier transforms of *fields* (or rather, of the deviations from their equilibrium values)

$$B_{\mathbf{k}}^{i} = (1/V)\int d\mathbf{r}[\exp(-i\mathbf{k}\cdot\mathbf{r})]B^i(\mathbf{r}) \tag{7}$$

The fields $B(t)$ in (5) (with components $B_{\mathbf{k}}$ and $|k_{x,y,z}| < k_c$) are parameters with (otherwise) arbitrary values and specify the local equilibrium state completely. By varying the $B(t)$ one can give the local equilibrium average of the hydrodynamic modes $\langle a\rangle_l$ any desired value. When the low Fourier components of the averages of the hydrodynamic modes in some nonequilibrium ensemble are given, the *associated* local equilibrium state is constructed such that

$$\langle a\rangle_{\text{noneq}} = \langle a\rangle_l = (\partial/\partial B^\dagger)\ln\langle\exp(a^\dagger\cdot B)\rangle \tag{8}$$

This equation defines the associated hydrodynamic fields B.

The Fourier language developed above is the convenient one in the context of Sections 3 and 4. The problems taken up in Sections 5 and 6, however, are more easily elucidated in terms of a cell picture in real space. Since the Fourier description has been restricted to the $M = k_c{}^3\pi^{-3}V$ lowest components, a real space description in terms of a cubic lattice with $M = V/v_c$ cells of volume v_c is completely equivalent.

The hydrodynamic fields in cell α with center at r_α are then, by (7),

$$B^i(\alpha) = \sum_{\mathbf{k}}{}' [\exp(i\mathbf{k}\cdot\mathbf{r}_\alpha)]B_{\mathbf{k}}^{i} \tag{9}$$

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[7] It would be somewhat more natural to restrict the summation over $\mathbf{k}$ to a spherical region. The cubical one is more convenient for our purpose, however, and the difference is of no physical consequence.

In cell language the *integrated* densities over a cell α are the natural objects to work with, so we define[8]

$$a^i(\alpha) = (v_c/V) \sum_{\mathbf{k}}{}' [\exp(i\mathbf{k}\cdot\mathbf{r}_\alpha)] a_{\mathbf{k}}^i \tag{10}$$

With these definitions the dot product (6) can be read in two equivalent ways:

$$a^\dagger \cdot B = \sum_{\mathbf{k},i}{}' a_{\mathbf{k}}^{i*} B_{\mathbf{k}}^i = \sum_{\alpha,i} a^i(\alpha) B^i(\alpha) \tag{11}$$

with two corresponding interpretations of the local equilibrium state (5).

3. LOCAL EQUILIBRIUM AND HYDRODYNAMICS

3.1. Initial Condition

The importance of local equilibrium states is that they are, in a coarse-grained sense, asymptotically approximate solutions of the Liouville equation for a certain class of initial states. In this section we shall consider the hydrodynamic equations for such initial states.

The fundamental assumption underlying this paper is that special[9] (to be specified later) initial states will approach a local equilibrium state rapidly. On the basis of this assumption we focus, in the spirit of the Chapman–Enskog development of the Boltzmann equation (see, e.g., Ref. 19) on the small *deviation* from the *local* equilibrium state, and derive hydrodynamic equations with transport kernels slightly generalized beyond the Navier–Stokes order. Such a procedure is, of course, not new, and our version is particularly close to the one proposed by Piccirelli.(20) It will be restated here in a condensed form as a necessary background for the arguments in subsequent sections.

Our special initial distribution $\rho_s(\Gamma; 0)$ can be written in the form [defining $\phi(\Gamma)$]

$$\rho_s(\Gamma; 0) = \rho_l(\Gamma | B)[1 + \phi(\Gamma)] \tag{12}$$

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[8] In Γ-space variables, $a^i(\alpha)$ is to be interpreted as

$$a^i(\Gamma; \alpha) = \sum_j a_j^i(\Gamma)$$

where the sum goes over all *particles j* with centers $\mathbf{r}_j$ within cell α. When a^i is the energy density, $a_j^i(\Gamma)$ will depend on the position of particles within the interaction range of particle j; i.e., $a(\Gamma; \alpha)$ may depend on the positions of particles outside (but close to) cell α.

[9] Such refinements of the local equilibrium assumption by restricting the initial states is only relevant if one is interested in nonlinear hydrodynamics (e.g., Section 4). In the case of linear hydrodynamics one may allow an essentially arbitrary initial state.

where B, in accordance with (8), is defined by

$$\int d\Gamma\, \rho_s(\Gamma; 0)a(\Gamma) = \int d\Gamma\, \rho_l(\Gamma|B)a(\Gamma) \tag{13}$$

The local equilibrium assumption can now be formulated as follows: The distribution $\rho_s(\Gamma; t)$ resulting from our special initial states will rapidly evolve (in a coarse-grained sense) into a distribution of the *normal* form

$$\rho_n(\Gamma; t|B) = \rho_l(\Gamma|B(t))[1 + \psi_B(\Gamma; t)] \tag{14}$$

where the long-lived deviation ψ_B from strict local equilibrium is generated by the gradients in the hydrodynamic fields $B(t)$ themselves. [The contribution to $\rho_s(\Gamma; t)$ resulting from the initial $\phi(\Gamma)$ decays after a short initial period has elapsed.] Again in accordance with (8), these fields are, for *all* times, defined by

$$\int d\Gamma\, \rho_s(\Gamma; t)a(\Gamma) = \int d\Gamma\, \rho_l(\Gamma|B(t))a(\Gamma) \tag{15}$$

and, in particular, their initial values are given by (13). The normalization of ρ_n in (14) together with (15) immediately yields the local equilibrium averages

$$\langle\psi_B(t)\rangle_l = 0; \qquad \langle\psi_B(t)a\rangle_l = 0 \tag{16}$$

Below we shall derive (hydrodynamic) equations for $B(t)$ assuming the Γ-space distribution to be of the normal form (14). As a consequence, the equations will only be valid *after* the initial state ρ_s has decayed to ρ_n. The precise duration of the initial transient will depend on the detailed properties of ρ_s but is, by assumption, *short*. The question is then whether $B(t)$ by backward hydrodynamic extrapolation to $t = 0$, i.e., through the initial transient, coincides with the true value of $B(0)$ as given by (13). In general, the answer is *no* and the nontrivial problem of constructing corrected initial conditions for hydrodynamics, using the information contained in $\phi(\Gamma)$ of (12), is known as the "connection problem across the initial boundary layer."(21),[10]

However, the difference between the true $B(0)$ and the appropriate initial data is only significant in the context of hydrodynamic equations of $O(k^3)$ or higher. In the following we shall only generalize Navier–Stokes [i.e., $O(k^2)$] hydrodynamics by less than one order in the wavenumber. *Within* this approximation it is, indeed, consistent to neglect the information contained in $\phi(\Gamma)$ and use B from (13) as initial data.

Effectively, then, the local equilibrium assumption enables us, within the approximation needed, to base the subsequent discussion on (14) for *all* $t \geqslant 0$, supplemented with the initial condition $\psi_B(\Gamma; 0) = 0$.

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[10] For a simple example see Ref. 22.

3.2. The Gradient Term

The first step in the derivation of hydrodynamics from (14)–(15) consists in expressing $\psi_B(\Gamma; t)$ as a functional of local equilibrium states, i.e., as a functional of $B(t)$. To this end, rewrite the Liouville equation for ρ_n as follows:

$$[(\partial/\partial t) + \mathscr{L}]\rho_l(t)\psi_B(t) = -[(\partial/\partial t) + \mathscr{L}]\rho_l(t) \tag{17}$$

The local equilibrium state depends on time through $B(t)$ only, and using (5) and (8), we have

$$\begin{aligned} \frac{\partial \rho_l(t)}{\partial t} &= \rho_l(t)\left[a^\dagger \cdot \frac{\partial B}{\partial t} - \frac{\partial}{\partial t}\ln\langle \exp(a^\dagger \cdot B)\rangle\right] \\ &= \rho_l(t)[a^\dagger(\Gamma) - \langle a^\dagger\rangle_l]\cdot\frac{\partial B}{\partial t} \end{aligned} \tag{18}$$

In general, $\partial B/\partial t$ is related to $\partial\langle a\rangle_l/\partial t$ by the matrix relation

$$\frac{\partial B}{\partial t} = \frac{\partial B}{\partial\langle a\rangle_l}\cdot\frac{\partial\langle a\rangle_l}{\partial t} \tag{19}$$

The Liouville equation gives for the latter quantity

$$\begin{aligned} \partial\langle a\rangle_l/\partial t &= -\int d\Gamma\, a(\Gamma)\mathscr{L}\rho_n(\Gamma; t\,|\,B) \\ &= -\int d\Gamma\, [a(\Gamma) - \langle a\rangle_l]\mathscr{L}\rho_n(\Gamma; t\,|\,B) \end{aligned} \tag{20}$$

The last term in (20) could be added since it vanishes due to the time invariance of the normalization of ρ_n.

Equations (18)–(20) can be written in the compact form

$$\partial\rho_l(t)/\partial t = -P'(t)\mathscr{L}\rho_n(t) \tag{21}$$

where the projector[23] $P'(t)$ is defined by

$$\begin{aligned} P'(t)\rho_l h(\Gamma) = \rho_l[a^\dagger(\Gamma) &- \langle a^\dagger\rangle_l]\cdot\partial B/\partial\langle a\rangle_l \\ &\cdot\int d\Gamma'[a(\Gamma') - \langle a\rangle_l]\rho_l(\Gamma')h(\Gamma') \end{aligned} \tag{22}$$

Due to the weight factor ρ_l the integral in (22) converges [i.e., the definition of $P'(t)$ is meaningful] for a large class of functions $h(\Gamma)$. The fact that $[P'(t)]^2 = P'(t)$ is a consequence of the local equilibrium version of (2). Differentiation of (8) gives

$$\langle[a(\Gamma) - \langle a\rangle_l][a^\dagger(\Gamma) - \langle a^\dagger\rangle_l]\rangle = \partial\langle a\rangle_l/\partial B \tag{23}$$

Since, by (7), B is of $O(V^{-1})$, the right-hand side of (23) is of $O(V)$, as it should be. Use of (21) and (14) transforms (17) into

$$[(\partial/\partial t) + P_\perp'(t)\mathscr{L}]\rho_l(t)\psi_B(t) = -P_\perp'(t)\mathscr{L}\rho_l(t) \tag{24}$$

where $P_\perp' = 1 - P'$.

Equation (24) is still an exact consequence of (17). In particular, no linearization has yet been performed. At this point, however, we shall take formal advantage of the basic assumption already introduced, and, in the spirit of the Chapman–Enskog method, linearize around states of local equilibrium. Note first that

$$\mathscr{L}\rho_l = \rho_l\mathscr{L}a^\dagger\cdot B \tag{25}$$

is of $O(k_c B)$, i.e., linear in B and k_c, since $\mathscr{L}a^\dagger$ is linear in k for small k, and the summation implied by the dot product in (25) is restricted to $k < k_c$. Similarly,

$$\partial\rho_l/\partial t = \rho_l a^\dagger\cdot\partial B/\partial t \tag{26}$$

is of $O(k_c B)$, as follows from (18)–(20). Finally, since $\psi_B(\Gamma; 0) = 0$, $\psi_B(\Gamma; t)$ will be of the order of the inhomogeneous term in (24), i.e., again of $O(k_c B)$. To linear order in the gradients, (24) consequently reduces to[11]

$$[(\partial/\partial t) + P_\perp(t)\mathscr{L}]\psi_B(t) = -P_\perp(t)\mathscr{L}\ln\rho_l(t) + O(k_c^2 B^2) \tag{27}$$

with the projector $P_\perp(t) = 1 - P(t)$ defined by

$$\rho_l(t)P(t) = P'(t)\rho_l(t) \tag{28}$$

The formal solution of (27) with initial condition $\psi_B(\Gamma; 0) = 0$ reads

$$\psi_B(\Gamma; t) = -\int_0^t dt'\, G(t, t')P_\perp(t')\mathscr{L}\ln\rho_l(t') \tag{29}$$

where the Green's function (or rather, the integral operator on Γ space) $G(t, t')$ solves the equation

$$[(\partial/\partial t) + P_\perp(t)\mathscr{L}]G(t, t') = 0 \tag{30}$$

subject to the initial condition $G(t, t) = 1$. Equation (29) accomplishes our first goal: it provides an expression for ψ_B in terms of ρ_l or, equivalently, in terms of B. Its validity is restricted by the fact that terms of $O(k_c^2 B^2)$ have been neglected. For our purposes, however, (29) will prove sufficiently accurate.

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[11] From (27) the *nonlinear* Navier–Stokes equations can be derived consistently, but not the *nonlinear* Burnett equations. The *linear* Burnett equations are still completely contained in (27). However, the initial condition, $\psi_B(\Gamma; 0)$, should be modified to this order of approximation in view of the connection problem across the initial boundary layer.

3.3. Nonlinear Hydrodynamics

As a second step one must derive hydrodynamic equations for $B(t)$ by appealing to the conservation laws together with (19) and (29). Equation (20) can be rewritten as

$$\partial\langle a\rangle_l/\partial t = -\int d\Gamma\, a(\Gamma)\mathscr{L}\rho_n = \int d\Gamma\, \rho_n\mathscr{L}a = \langle\mathscr{L}a\rangle_l + \langle\psi_B\mathscr{L}a\rangle_l \tag{31}$$

The first term [of $O(k_cB)$] has the form

$$\langle\mathscr{L}a\rangle_l = -\int d\Gamma\, a\mathscr{L}\rho_l = -\langle a\mathscr{L}a^\dagger\rangle_l\cdot B(t) \equiv -V\Omega_B(t)\cdot B(t) \tag{32}$$

The subscript stresses the dependence of Ω_B on the local equilibrium parameters B.

The second term in (31) is of $O(k_c{}^2B)$ [and use of (29) implies neglect in (31) of terms of $O(k_c{}^3B^2)$]. As a preliminary to writing it down, note that

$$\langle\psi_B(t)P(t)\mathscr{L}a\rangle_l = 0 \tag{33}$$

which follows from the definitions (28) and (22), and the relations (16). Insertion of (29) and use of (25) then yields

$$\langle\psi_B(t)\mathscr{L}a\rangle_l = -\int_0^t dt'\, \langle[P_\perp(t)\mathscr{L}a]G(t,t')P_\perp(t')\mathscr{L}a^\dagger\rangle_l\cdot B(t') \tag{34}$$

Define the correlation matrix $U_B(t, t')$ as

$$U_B(t,t') = V^{-1}\langle[P_\perp(t)\mathscr{L}a]G(t,t')P_\perp(t')\mathscr{L}a^\dagger\rangle_l \tag{35}$$

Equation (31) can then be written in the form

$$\frac{1}{V}\frac{\partial\langle a\rangle_l}{\partial t} = -\Omega_B(t)\cdot B(t) - \int_0^t dt'\, U_B(t,t')\cdot B(t') \tag{36}$$

Supplemented with (19), Eq. (36) constitutes a closed set of equations of motion for the hydrodynamic fields $B(t)$. If the second term on the right is neglected, (36) reduces to the usual nonlinear Euler equations. Dissipation is included with the second term and from its form it is clear that non-Markovian effects have not been altogether ignored. In fact, (36) mildly generalizes the *nonlinear* Navier–Stokes equations [$O(k_c{}^2)$] in that it is consistent up to, but not including, $O(k_c{}^3)$.

3.4. Linearized Hydrodynamics

Finally, we perform a *second* linearization. This time we shall linearize in small deviations (measured by B) from *total* equilibrium; i.e., we shall replace the matrices $\partial\langle a\rangle_l/\partial B$, $\Omega_B(t)$, and $U_B(t, t')$ by their total equilibrium forms. The first one becomes trivial due to (23) and (2),

$$\partial\langle a\rangle_l/\partial B \to V\mathbf{1} \tag{37}$$

In equilibrium, the Euler matrix becomes strictly diagonal in **k**. With the current $j_{\mathbf{k}}^i$ defined as

$$-ikj_{\mathbf{k}}^i = \mathscr{L}a_{\mathbf{k}}^i \tag{38}$$

we extract the factor ik and define the equilibrium 5×5 Euler matrix as

$$ik\Omega_{\mathbf{k}} = (1/V)\langle a_{\mathbf{k}}\mathscr{L}a_{\mathbf{k}}^\dagger\rangle = ik(1/V)\langle a_{\mathbf{k}}j_{\mathbf{k}}^\dagger\rangle \tag{39}$$

To find the dissipative matrix in equilibrium we first note that the Green's function (30) reduces to

$$G(t, t') = \exp[-(t - t')P_\perp\mathscr{L}] \tag{40}$$

where the equilibrium form of $P_\perp = 1 - P$ follows from (22), (28), (37), and translational invariance as

$$Pg_{\mathbf{k}} = (1/V)\sum_i a_{\mathbf{k}}^i(\Gamma)\langle a_{\mathbf{k}}^{i*}g_{\mathbf{k}}\rangle \tag{41}$$

Projected quantities will be distinguished by a caret,

$$\hat{j}_{\mathbf{k}} = P_\perp j_{\mathbf{k}} = -(ik)^{-1}P_\perp\mathscr{L}a_{\mathbf{k}}; \qquad \hat{\mathscr{L}} = P_\perp\mathscr{L} \tag{42}$$

Second, the dissipative matrix also becomes strictly diagonal in **k** in total equilibrium, and extracting the factor $|ik|^2$, we define the 5×5 matrix

$$U_{\mathbf{k}}(t) = V^{-1}\langle\hat{j}_{\mathbf{k}}[\exp(-t\hat{\mathscr{L}})]\hat{j}_{\mathbf{k}}^\dagger\rangle \tag{43}$$

The linearized version of (36) then reads

$$\partial B_{\mathbf{k}}/\partial t = -ik\Omega_{\mathbf{k}}\cdot B_{\mathbf{k}}(t) - k^2\int_0^t dt'\, U_{\mathbf{k}}(t - t')\cdot B_{\mathbf{k}}(t') \tag{44}$$

where the dot products have the same meaning as previously, except that no summation over **k** is implied.

In terms of Laplace transforms

$$B_z = \int_0^\infty dt\, e^{-zt}B(t) \tag{45}$$

(44) can be cast into its final form

$$zB_{\mathbf{k}z} + ik\Omega_{\mathbf{k}}\cdot B_{\mathbf{k}z} + k^2U_{\mathbf{k}z}\cdot B_{\mathbf{k}z} = B_{\mathbf{k}}(0) \tag{46}$$

We close this section with a summary and some comments:

(i) For certain special initial states, which are assumed to evolve rapidly into states close to local equilibrium, we have derived hydrodynamic equations by a two-step linearization. In the first place we found an expression, (29), for the small difference between the normal solution and strict local equilibrium, by linearizing in small gradients. The result, (36), was a complicated set of nonlinear, non-Markovian equations for the hydrodynamic fields. (Some remarks on the linear and nonlinear hydrodynamic equations for arbitrary initial states will be made in Section 7.) A second linearization around *total* equilibrium resulted in the simpler equation (46).

(ii) The dissipative kernels $U_{\mathbf{k}}(t)$ are projected correlation functions and reduce, in the limits $V \to \infty$, $\mathbf{k} \to 0$, and after integration over t, to the appropriate combinations of the standard Green–Kubo expressions$^{(3)}$ for the linear Navier–Stokes transport coefficients. Their $\mathbf{k}$-dependent versions, as derived here, have been shown by Pomeau$^{(10)}$ and Ernst and Dorfman$^{(11)}$ to contain terms of orders $k^{1/2}$, $k^{3/4}$, $k^{7/8}$, etc.; i.e., although the consistency of (46) breaks down at $O(k^3)$, those equations still contain an infinite series of correction terms beyond the Navier–Stokes level of $O(k^2)$.

(iii) At the beginning of this section we argued that in the context of hydrodynamic equations of lower order than $O(k^3)$ the "true" initial values $B(0)$, as determined by (13), can consistently be used; i.e., they are sufficient with regard to (46). For hydrodynamics of $O(k^3)$, however, the true initial values should be corrected by terms of $O(k)$.

4. TIME CORRELATION FUNCTIONS AND THE GRADIENT TERM

In the previous section we derived, on the basis of a local equilibrium assumption, hydrodynamic equations with dissipative kernels expressed as projected current–current time correlation functions. We now turn to the problem of how this type of assumption can be exploited in the *evaluation* of such correlation functions.

The quantity of central interest is the dissipative matrix $U_{\mathbf{k}}(t)$ of (43), which contains the projected propagator $\exp(-t\hat{\mathscr{L}})$. Since, however, local equilibrium ideas are associated with evolution according to the *full* Liouville propagator $\exp(-t\mathscr{L})$, their immediate implications relate to the *true* (5 × 5) time correlation matrix

$$C_{\mathbf{k}}(t) = V^{-1}\langle \hat{j}_{\mathbf{k}} e^{-t\mathscr{L}} \hat{j}_{\mathbf{k}}^{\dagger}\rangle = V^{-1}\langle \hat{j}_{\mathbf{k}}(t) \hat{j}_{\mathbf{k}}^{\dagger}\rangle \tag{47}$$

In this section we show that the long-time asymptotics of $U_{\mathbf{k}}(t)$ can be found by a calculation of $C_{\mathbf{k}}^{l}(t)$, the local equilibrium contribution to $C_{\mathbf{k}}(t)$.

The actual calculation of $C_{\mathbf{k}}^{l}(t)$ is taken up in subsequent sections. For our present purposes all that is needed is the method in broad outline. The first step is to split the equilibrium ensemble in (47) into special initial ensembles $\rho_s(\Gamma; 0)$ such that $C_{\mathbf{k}}(t)$ can be written as[12]

$$C_{\mathbf{k}}(t) = \int ds\, h(s)\langle \hat{j}_{\mathbf{k}}(t)\rangle_s \tag{48}$$

where $\langle\cdots\rangle_s$ is an average with $\rho_s(\Gamma; 0)$, and $h(s)$ is the product of the initial current and the appropriate weight factor. By assumption the Γ-space distribution of these special ensembles rapidly approaches the normal form ρ_n of (13) and subsequently evolves according to the hydrodynamic equations of Section 3. With the known form of ρ_n one can calculate the current at time t. Finally, the special ensembles are recombined according to Eq. (48) and $C_{\mathbf{k}}(t)$ is obtained.

Two differences with Section 3 should be noted. First, our present objective is the derivation of expressions for $C_{\mathbf{k}}(t)$, or, by virtue of (48), for the current $\langle \hat{j}_{\mathbf{k}}(t)\rangle_s$. Consequently, rather than eliminate currents in favor of hydrodynamic quantities, as in Section 3, we shall choose the opposite strategy. Second, it will prove useful to keep (formally) the rapidly decaying initial contributions to the current.

Evaluation of $C_{\mathbf{k}}(t)$ is essentially the calculation of the current in (48), which can be expressed as

$$\langle \hat{j}_{\mathbf{k}}(t)\rangle_s \equiv J_{\mathbf{k}}(t) = J_{\mathbf{k}}^{i}(t) + J_{\mathbf{k}}^{l}(t) + J_{\mathbf{k}}^{g}(t) \tag{49}$$

Here we have written $\rho_s(t) = \rho_i(t) + \rho_n(t)$, where ρ_n is the normal form (14) and $\rho_i(t)$ is a (by assumption) rapidly decaying initial term. The average of $\hat{j}_{\mathbf{k}}$ calculated with $\rho_i(\Gamma; t)$ is indicated by $J_{\mathbf{k}}^{i}(t)$. The average of $\hat{j}_{\mathbf{k}}$ with the normal form gives according to (14) a local equilibrium current, $J_{\mathbf{k}}^{l}(t) = \langle \hat{j}_{\mathbf{k}}\rangle_l$, and a contribution from the gradient term, reading

$$J_{\mathbf{k}}^{g}(t) = \langle \psi_B(t)\hat{j}_{\mathbf{k}}\rangle_l = -ik\int_0^t dt'\, U_{\mathbf{k}}(t - t')\cdot\langle a_{\mathbf{k}}(t')\rangle_s + R(t) \tag{50}$$

This expression, where only the linear term is written explicitly and the remainder is denoted by $R(t)$, can be deduced from (34) by the procedure of Section 3.4. The linearization is based on the following considerations. There are two smallness parameters in the problem: The wavenumber k and the deviations from equilibrium as measured by $\langle a_{\mathbf{k}}\rangle_s$, or equivalently by $B_{\mathbf{k}}$. The first term on the right-hand side of (49) is of $O(1)$ as far as both k and B are concerned. As will be shown in Section 5, $J_{\mathbf{k}}^{l}$ is to leading order of $O(B^2)$ (i.e., nonlinear in the hydrodynamic fields); higher order corrections such as

[12] The details of the prescription for the splitting do not affect the arguments of this section, which apply equally well whether the procedure of Section 5 or that of Section 6 is chosen.

$O(kB^2)$ or $O(B^3)$ will be neglected. From (50) it is also clear that $J_{\mathbf{k}}^{g}$ is of $O(kB)$; the correction term $R(t)$ is again of $O(kB^2)$ and consistency thus requires that it be neglected.

As mentioned already, we intend to eliminate $\langle a_{\mathbf{k}}(t)\rangle_s$ from (50) and observe that $(a_{\mathbf{k}}(t)\rangle_s$ is related to $\langle \hat{j}_{\mathbf{k}}(t)\rangle_s = J_{\mathbf{k}}(t)$ through the hydrodynamic equations, i.e.,

$$\partial\langle a_{\mathbf{k}}\rangle_s/\partial t = \int d\Gamma\, \rho_s \mathscr{L} a_{\mathbf{k}} = \int d\Gamma\, \rho_s(P\mathscr{L}a_{\mathbf{k}} + P_{\perp}\mathscr{L}a_{\mathbf{k}}) \tag{51}$$

Due to (39) and (41) the first term reads

$$\int d\Gamma\, \rho_s P\mathscr{L}a_{\mathbf{k}} = -ik\int d\Gamma\, \rho_s \Omega_{\mathbf{k}}\cdot a_{\mathbf{k}} = -ik\Omega_{\mathbf{k}}\cdot\langle a_{\mathbf{k}}(t)\rangle_s \tag{52}$$

and with (42) and (49) we have for the second term $\langle P_{\perp}\mathscr{L}a_{\mathbf{k}}\rangle_s = -ikJ_{\mathbf{k}}(t)$. Taking Laplace transforms of (51) and solving for $\langle a_{\mathbf{k}z}\rangle_s$, we obtain

$$\langle a_{\mathbf{k}z}\rangle_s = (z + ik\Omega_{\mathbf{k}})^{-1}\cdot(\langle a_{\mathbf{k}}\rangle_s - ikJ_{\mathbf{k}z}) \tag{53}$$

where $\langle a_{\mathbf{k}}\rangle_s$ is the initial value at $t = 0$. [Equation (53) is actually a nonlinear hydrodynamic equation, since $J_{\mathbf{k}}(t)$ contains $J_{\mathbf{k}}^{l}(t)$, which is quadratic in the hydrodynamic fields $B(t)$.] By inserting $\langle a_{\mathbf{k}z}\rangle_s$ in the Laplace transform of (50), multiplying it by $h(s)$, and integrating over s, we have expressed the contribution of the gradient term in $U_{\mathbf{k}z}$ and $C_{\mathbf{k}z}$ themselves

$$C_{\mathbf{k}z}^{g} = -k^2 U_{\mathbf{k}z}\cdot(z + ik\Omega_{\mathbf{k}})^{-1}\cdot C_{\mathbf{k}z} \tag{54}$$

$C_{\mathbf{k}z}^{a}$ is defined in the obvious manner

$$C_{\mathbf{k}z}^{a} = \int ds\, h(s) J_{\mathbf{k}z}^{a}, \qquad a = g, l, i \tag{55}$$

and the relation $\int ds\, h(s)\langle a_k\rangle_s = \langle \hat{j}_{\mathbf{k}}^{*} a_{\mathbf{k}}\rangle = 0$, which is a consequence of (41)–(42), was used in the derivation of (54). Since $C_{\mathbf{k}z} = C_{\mathbf{k}z}^{i} + C_{\mathbf{k}z}^{l} + C_{\mathbf{k}z}^{g}$, it follows directly from (54) that

$$C_{\mathbf{k}z} = (z + ik\Omega_{\mathbf{k}})\cdot(z + ik\Omega_{\mathbf{k}} + k^2 U_{\mathbf{k}z})^{-1}\cdot(C_{\mathbf{k}z}^{i} + C_{\mathbf{k}z}^{l}) \tag{56}$$

The remarkable fact is that the relation (56) is of precisely the same form as the identity connecting the matrix of true time correlation functions, $C_{\mathbf{k}z}$ in (47), with the dissipative matrix, $U_{\mathbf{k}z}$ in (43), reading^{(24)}

$$C_{\mathbf{k}z} = (z + ik\Omega_{\mathbf{k}})\cdot(z + ik\Omega_{\mathbf{k}} + k^2 U_{\mathbf{k}z})^{-1}\cdot U_{\mathbf{k}z} \tag{57}$$

This identity may be derived directly from the matrix relations

$$U_{\mathbf{k}z} = C_{\mathbf{k}z} + ikU_{\mathbf{k}z}\cdot N_{\mathbf{k}z}, \qquad zN_{\mathbf{k}z} = -ikC_{\mathbf{k}z} - ik\Omega_{\mathbf{k}}\cdot N_{\mathbf{k}z} \tag{58}$$

with $N_{\mathbf{k}z} = V^{-1}\langle a_{\mathbf{k}z}\hat{j}_{\mathbf{k}}^{\dagger}\rangle$, as can be obtained from (39)–(43).

By comparing (55) and (57) we deduce that

$$U_{\mathbf{k}}(t) = C_{\mathbf{k}}^{i}(t) + C_{\mathbf{k}}^{l}(t) \tag{59}$$

Since the initial contribution $C_{\mathbf{k}}^{i}(t)$ is rapidly decaying, we have shown that the long-time asymptotic behavior of the projected correlation function $U_{\mathbf{k}}(t)$ is precisely given by the local equilibrium contribution to the full correlation function, i.e.,

$$U_{\mathbf{k}}(t) \simeq C_{\mathbf{k}}^{l}(t) \tag{60}$$

We emphasize again that the main results (59) and (60) are independent of the details of the splitting in special ensembles, and apply as long as the time evolution of these special ensembles is governed by hydrodynamics.

In summary, we have proved the following statement: Suppose that the long-time behavior of $C_{\mathbf{k}}(t)$ is calculated on the basis of a local equilibrium assumption (14), i.e., $C_{\mathbf{k}}(t) \simeq C_{\mathbf{k}}^{l}(t) + C_{\mathbf{k}}^{g}(t)$, where $C_{\mathbf{k}}^{l}$ and $C_{\mathbf{k}}^{g}$ are, respectively, the contributions from the local equilibrium term and that from the gradient term ψ_B in (14). Then, (i) $C_{\mathbf{k}}^{g}(t)$ can be expressed in terms of $U_{\mathbf{k}}(t)$ and of $C_{\mathbf{k}}(t)$ itself. (ii) From the identity (57) relating $U_{\mathbf{k}}(t)$ and $C_{\mathbf{k}}(t)$ it follows that the long-time asymptotic behavior of the projected correlation matrix $U_{\mathbf{k}}(t)$ is given by the local equilibrium contribution $C_{\mathbf{k}}^{l}(t)$ to the true correlation matrix.

5. DECOMPOSITION IN BARE HYDRODYNAMIC ENSEMBLES

5.1. Motivation

We now turn to the delicate problem of how to construct a decomposition (48) of the initial equilibrium ensemble appearing in $C_{\mathbf{k}}(t)$ in such a way that each component can be consistently assumed to develop rapidly into a state close to local equilibrium. Strictly speaking, the decomposition we consider in the present section will prove a failure in this respect. We shall nevertheless discuss it in detail, for the following reasons:

(i) It has a certain intuitive appeal and is close in spirit to the procedure used in I.

(ii) It amounts to a somewhat simplified version of a derivation already given by Pomeau(17) of the mode-coupling formula, and consequently (given the results of Section 4) implies all the results on long-time tails, etc., found in Refs. 10–14.

(iii) The fact that it nevertheless, when applied to *other* correlation functions than those encountered in Sections 3 and 4, leads to serious inconsistencies, is a striking demonstration of the subtlety of the local equilibrium assumption. Thus, the successes and failures of the decomposition to be used in the present section motivate the discussion in Section 6 and lead to the refined decomposition and final form of the local equilibrium assumption found there.

For simplicity we consider a typical (diagonal) term of the matrix $C_{\mathbf{k}}(t)$ in (47). The arguments used are independent of this restriction, and the results generalize immediately to the complete matrix. The object to be considered, then, is

$$C_{\mathbf{k}}(t) = V^{-1}\langle \hat{j}_{\mathbf{k}} e^{-t\mathscr{L}} \hat{j}_{\mathbf{k}}^{*} \rangle \tag{61}$$

where $\hat{j}_{\mathbf{k}} = \sum_i \hat{j}_i \exp(-i\mathbf{k}\cdot\mathbf{r}_i)$ is *one* of the components of the column vector (42), and where $\hat{j}_i$ is the part of the total current associated with the ith particle. The notation is slightly deceptive: With potential terms included, $\hat{j}_i$ will depend not only on the velocity of particle i, but also on the relative positions of all other particles within the range of the interaction.

5.2. Cell Description of $C_{\mathbf{k}}(t)$

As pointed out in Section 2, a slowly varying function with periodic boundary conditions in V can be equivalently represented by its Fourier components up to a finite cutoff $|k_{x,y,z}| \leqslant k_c$ or by the set of values it takes on a cubic lattice with lattice constant $l = \pi k_c^{-1}$ in real space. Each lattice point can be considered the center of a cubic cell with volume $v_c = l^d$, the total number of cells being $M = V/v_c = Vk_c^3/\pi^3$. Our discussion is confined to hydrodynamic, i.e., small, wavenumbers and we shall measure l on the scale of the equilibrium correlation length ξ. The cell size will be chosen as small as possible, but sufficiently large that the open subsystem associated with each cell can be considered thermodynamic to a good approximation. This choice implies that equilibrium cell–cell correlations should, for consistency, be neglected. One can think of l as being typically of the order of 10ξ.

With a notation analogous to that of (10), we can then write

$$\hat{j}_{\mathbf{k}} = \sum_{\alpha} \hat{j}(\alpha) \exp(-i\mathbf{k}\cdot\mathbf{r}_{\alpha}) \tag{62}$$

where $\hat{j}(\alpha) = \hat{j}(\Gamma; \alpha)$ is the total current of cell α. It depends, apart from the phases of the particles inside cell α, also on the positions of particles in neighboring cells close to the (mathematical) surface of cell α. The relative importance of this surface effect, as compared to the bulk contribution to $\hat{j}(\alpha)$, decreases with increasing cell size. For $l \approx 10\xi$ it should already be negligible.

In the same spirit $C_{\mathbf{k}}(t)$ can be expressed in terms of the cell–cell correlation function $C_{\alpha\beta}(t)$ as

$$C_{\mathbf{k}}(t) = (v_c/V) \sum_{\alpha,\beta} \{\exp[i\mathbf{k}\cdot(\mathbf{r}_\alpha - \mathbf{r}_\beta)]\} C_{\alpha\beta}(t) \tag{63}$$

with

$$C_{\alpha\beta}(t) = (1/v_c)\langle \hat{j}(\beta) e^{-t\mathscr{L}} \hat{j}(\alpha)\rangle \tag{64}$$

Consistent neglect of equilibrium cell–cell correlations implies that $C_{\alpha\beta}(0) = \text{const} \times \delta_{\alpha\beta}$. We shall mostly use cell language in what follows, since it is conceptually clearer. We stress, however, that the formal manipulations are equivalent in Fourier language.

5.3. Bare Hydrodynamic Ensembles

We shall now construct a specific decomposition of the type (48) of the initial equilibrium ensemble and formulate the corresponding local equilibrium assumption. The conceptual advantage of a cell description in this context is the possibility of realizing the decomposition (48) by imposing *local* constraints.

Since we are discussing current–current correlation functions, we first have to remove the factor $\hat{j}(\Gamma; \alpha)$, because it would lead to a nonpositive probability density. (For an alternative treatment see Section 7.4.) This can be done by constraining the total current in cell α to have the value J at time $t = 0$. The corresponding decomposition amounts to slicing Γ space into shells of constant $\hat{j}(\Gamma; \alpha)$. The normalized distribution on a shell at $t = 0$ reads

$$\rho_J(\Gamma; 0) = \rho_{\text{eq}}(\Gamma)\, \delta(\hat{j}(\Gamma; \alpha) - J)/f_{\text{eq}}(J) \tag{65}$$

with the equilibrium current distribution given as

$$f_{\text{eq}}(J) = \langle \delta(j(\Gamma) - J)\rangle \tag{66}$$

Hence, the cell–cell correlation function can be written

$$C_{\alpha\beta}(t) = (1/v_c) \int dJ\, f_{\text{eq}}(J) J \langle \hat{j}(\beta; t)\rangle_J \tag{67}$$

where $\langle \hat{j}(\beta; t)\rangle_J$ is the average of the current in cell β at time t taken over the initially constrained ensemble (65).

However, the decomposition (67) is not sufficiently detailed for the application of a local equilibrium assumption. The reason is that all averages of conserved densities and thus all hydrodynamic fields vanish in the constrained ensemble (65). Therefore, a further decomposition of $\rho_J(\Gamma; 0)$ with respect to its hydrodynamic content is needed, and in addition to $\hat{j}(\alpha)$, the conserved quantities $a^r(\alpha)$ $(r = 1,\ldots, d+2)$ in cell α are given specified values (namely A^r) at $t = 0$.(25) The corresponding generalizations of (65)–(67) read

$$\rho_{JA}(\Gamma;0) = \rho_{\mathrm{eq}}(\Gamma)\,\delta(\hat{j}(\alpha) - J)\prod_r \delta(a^r(\alpha) - A^r)/f_{\mathrm{eq}}(J, A) \tag{68}$$

$$f_{\mathrm{eq}}(J, A) = \left\langle \delta(\hat{j}(\alpha) - J)\prod_r \delta(a^r(\alpha) - A^r)\right\rangle \tag{69}$$

$$C_{\alpha\beta}(t) = (1/v_c)\int\cdots\int dJ\,dA\,f_{\mathrm{eq}}(J, A)J\langle \hat{j}(\beta; t)\rangle_{JA} \tag{70}$$

We shall call the distribution $\rho_{JA}(\Gamma; 0)$ a "bare" hydrodynamic ensemble since the corresponding variance of a conserved quantity in cell α vanishes at $t = 0$. "Dressed" hydrodynamic ensembles with a nonvanishing variance in the initial distribution of $a^r(\alpha)$ will be consideted in Section 6.

With the initial decomposition defined by (68)–(70) we are ready to formulate the associated local equilibrium assumption. We *assume* that $\rho_{JA}(\Gamma; t)$ rapidly takes the normal form

$$\rho_{JA}(\Gamma; t) \simeq \rho_n(\Gamma; t\,|\,B) = \rho_l(\Gamma|B(t))[1 + \psi_B(\Gamma; t)] \tag{71}$$

Before investigating the consequences of (71), we shall summarize the reasons why the decomposition (68)–(70) and the associated local equilibrium assumption (71) appear to form a sound foundation upon which a phenomenological theory can be built. The plausibility of (71) is based on the fact that the constraints imposed on ρ_{JA} are:

(a) *Local:* They are restricted to a single cell, since with $l \sim 10\xi$, equilibrium fluctuations in different cells are, essentially, uncorrelated.

(b) *Few:* In d dimensions the number of constraints is $d + 3$.[13] Roughly speaking, the fewer the constraints, the closer will the initial distribution be to a local equilibrium form, and the shorter the time required to approach the local equilibrium state.

(c) *Mild:* The distribution $f_{\mathrm{eq}}(J, A)$ effectively limits the values of J and A to the fluctuation range. Consequently, the initial inhomogeneities will be small in magnitude.

In addition, the constraints in (68) will, in the next subsection, be shown to be sufficient in the sense that (68)–(71) allow a derivation of the mode-coupling formula. Independent evidence(4,9) exists for the correctness of the long-time, small-k asymptotics of the mode-coupling formula. Such evidence clearly strengthens the case for (68)–(71). Nevertheless, as we shall see,

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[13] This is the point where the decomposition of this section differs from that used by Pomeau.(17)

(68)–(71) also lead to serious inconsistencies. As a result, we shall be forced to reconsider the precise *nature* of the constraints in Section 6.

5.4. Mode-Coupling Formula

The immediate task, however, is to investigate the consequences of Eqs. (68)–(71) and, in particular, to derive the mode-coupling formula from those equations. According to (60) it is the contribution to $C_{\mathbf{k}}(t)$ from $\rho_l(\Gamma|B(t))$ that determines the long-time asymptotics of $U_{\mathbf{k}}(t)$. Consequently, we replace ρ_{JA} by ρ_l to find for the current $\langle \hat{j}(\beta; t)\rangle_{JA}$

$$\begin{aligned}\langle \hat{j}(\beta)\rangle_l &= \langle \hat{j}(\beta)\exp[a^\dagger\cdot B(t)]\rangle/\langle\exp[a^\dagger\cdot B(t)]\rangle\\ &= \langle \hat{j}(\beta)\exp[a(\beta)\cdot B(\beta;t)]\rangle/\langle\exp[a(\beta)\cdot B(\beta;t)]\rangle \qquad (72)\end{aligned}$$

Here we have interpreted the dot product according to (11) in cell language and consistently neglected equilibrium correlations between different cells. Since the initial values of the decaying fields $B(\beta; t)$ are kept within the fluctuation range, an expansion of (72) in powers of B makes sense. Using the relations $\langle \hat{j}(\beta)\rangle = 0$ and $\langle a(\beta)\hat{j}(\beta)\rangle = 0$, which by translational invariance are immediate consequences of (41)–(42), we calculate the leading term to be

$$\langle \hat{j}(\beta)\rangle_l = \tfrac{1}{2}\sum_{r,s}\langle \hat{j}(\beta)a^r(\beta)a^s(\beta)\rangle B^r(\beta;t)B^s(\beta;t) + \cdots \qquad (73)$$

In paper I (Appendix A) we have explicitly shown that higher terms in (73) can be neglected for large times.

By virtue of (8) and (2), with consistent neglect of cell–cell correlations, the values of the hydrodynamic fields in cell β are, to lowest order in B, given by $v_c B^r(\beta, t) = \langle a^r(\beta; t)\rangle_{JA}$, with initial values $\langle a^r(\beta; 0)\rangle_{JA} = \delta_{\alpha\beta} A^r$. We now take formal advantage of the fact that the a's are chosen so as to diagonalize the hydrodynamic matrix, i.e., in cell language

$$B^r(\beta; t) = G^r(\beta; t|\alpha)B^r(\alpha; 0) \qquad (74a)$$

Or, in Fourier language,

$$B_{\mathbf{k}}^r(t) = G_{\mathbf{k}}^r(t)B_{\mathbf{k}}^r(0) \qquad (74b)$$

The hydrodynamic propagators[14] in (74a) and (74b) are related in the standard way [cf. (10)],

$$G^r(\beta; t|\alpha) = (v_c/V){\sum_{\mathbf{k}}}'\{\exp[i\mathbf{k}(\mathbf{r}_\beta - \mathbf{r}_\alpha)]\}G_{\mathbf{k}}^r(t) \qquad (75)$$

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[14] An explicit expression for the propagators is not needed for the general arguments. However, if one assumes that a dispersion relation for the "frequency" z_k^r of the hydrodynamic mode $a_{\mathbf{k}}^r$ exists up to, but not necessarily including, $O(k^3)$, one can write $G_{\mathbf{k}}^r(t) = \exp(z_k^r t)$.

After insertion of (74) into (73), it remains to average over initial conditions, i.e., to recombine the constrained ensembles into a total equilibrium one in the manner of (70). With (69) and the initial condition $B^r(\beta; 0) = v_c^{-1}\,\delta_{\alpha\beta}\,A^r$, the initial average yields the two-mode amplitude

$$\begin{aligned}\mathscr{A}^{rs} &\equiv v_c^{-1}\int\cdots\int dJ\,dA\,f_{\text{eq}}(J, A)JA^rA^s \\ &= v_c^{-1}\langle\hat{j}(\alpha)a^r(\alpha)a^s(\alpha)\rangle = V^{-1}\langle\hat{j}_{\mathbf{0}}a_{\mathbf{0}}{}^r a_{\mathbf{0}}{}^s\rangle \qquad (76)\end{aligned}$$

The subscript **0** indicates the Fourier component with $\mathbf{k} = 0$. The last equality in (76) is a consequence of the fact that $\mathscr{A}^{rs}$ becomes independent of v_c when $l \gg \xi$, i.e., the thermodynamic limit of $\mathscr{A}^{rs}$ exists. (For explicit expressions, see paper II.)

The local equilibrium contribution $C^l_{\alpha\beta}(t)$, which, as shown in Section 4, becomes asymptotically equal to $U_{\alpha\beta}(t)$ for long times, follows from (70)–(76) as

$$C^l_{\alpha\beta}(t) \simeq U_{\alpha\beta}(t) \simeq (2v_c)^{-1}\sum_{r,s}(\mathscr{A}^{rs})^2G^r(\beta; t\,|\,\alpha)G^s(\beta; t\,|\,\alpha) \qquad (77)$$

This is the *mode-coupling formula*, written in cell language. By (63) and (75) it can immediately be rewritten in the more familiar form

$$C_{\mathbf{k}}{}^l(t) \simeq U_{\mathbf{k}}(t) \simeq (2V)^{-1}\sum_{\mathbf{q}}{}'\sum_{r,s}{}'(\mathscr{A}^{rs})^2G_{\mathbf{q}}{}^r(t)G^s_{\mathbf{k}-\mathbf{q}}(t) \qquad (78)$$

The correlation functions are, strictly speaking, defined in the limit $V \to \infty$. In (78) this limiting process is reduced to $V^{-1}\sum_{\mathbf{q}}{}' \to (2\pi)^{-d}\int d\mathbf{q}$. In addition, the amplitudes $\mathscr{A}^{rs}$ are used in their limiting form.

The result (77)–(78) applies to one typical diagonal element of the matrix C or U. However, the various matrix elements only differ as far as the amplitudes (76) are concerned, and the generalization to the whole matrix is therefore immediate.

5.5. Counterexample

Encouraging as the results of the previous subsection may be, we shall now show by a counterexample that the local equilibrium assumption in the context of bare hydrodynamic ensembles may lead to serious inconsistencies.

Consider the quantity $\Xi_0(t) \equiv v_c^{-1}\langle\hat{j}(\beta, t)\rangle$, which vanishes by definition. [The current $\hat{j}(\beta, t)$ is not the only possible choice for the present argument; see, e.g., Section 6.1.] By virtue of (63)–(64), we are free to interpret it as the Fourier transform (with $\mathbf{k} = 0$) of the correlation function

$$\Xi_{\alpha\beta}(t) = v_c^{-1}\langle\hat{j}(\beta)e^{-t\mathscr{L}}\Delta(\alpha)\rangle \qquad (79)$$

where

$$\Delta(\alpha) = \begin{cases} 1 & \text{in cell } \alpha \\ 0 & \text{otherwise} \end{cases}$$

According to the procedure of Section 5.3, we decompose the initial equilibrium ensemble

$$\Xi_{\alpha\beta}(t) = v_c^{-1} \int dA\, f_{\mathrm{eq}}(A) \langle \hat{j}(\beta; t) \rangle_A \tag{80}$$

where $\langle \cdots \rangle_A$ is an average over the bare hydrodynamic ensemble

$$\rho_A(\Gamma; 0) = \rho_{\mathrm{eq}}(\Gamma) \prod_r \delta(a^r(\alpha) - A^r)/f_{\mathrm{eq}}(A) \tag{81}$$

with

$$f_{\mathrm{eq}}(A) = \left\langle \prod_r \delta(a^r(\alpha) - A^r) \right\rangle \tag{82}$$

In order to extract the long-time asymptotics of $\Xi_{\alpha\beta}(t)$, we appeal to the local equilibrium assumption on $\rho_A(\Gamma; t)$, in precise analogy with (71), and follow the steps of Section 5.4 to obtain

$$\Xi_0(t) \simeq (2V)^{-1} \sum_{\mathbf{q}}{}' \sum_r \mathscr{A}^{rr} G_{\mathbf{q}}^r(t) G_{-\mathbf{q}}^r(t) \tag{83}$$

where we have used the relation

$$v_c^{-1} \int dA\, f_{\mathrm{eq}}(A) A^r A^s = v_c^{-1} \langle a^r(\alpha) a^s(\alpha) \rangle = \delta_{rs} \tag{84}$$

If $\hat{j}_0$ represents any current except the longitudinal momentum current (for explicit expressions, see II), the amplitude $\mathscr{A}^{rr}$ and thereby the long-lived ($\sim t^{-3/2}$) terms of $\Xi_0(t)$ vanish identically. When $\hat{j}_0$ contains the longitudinal momentum current, however, $\mathscr{A}^{rr}$ does not vanish for all r. On the basis of (83) one would then conclude that $\Xi_0(t)$ has a $t^{-3/2}$ tail.

Although blatantly false, this conclusion was based on precisely the same procedure, and, in particular, on precisely the same assumption[15] of a rapid decay toward local equilibrium of bare hydrodynamic ensembles that was successfully exploited in the derivation of the mode-coupling formula.

It is hard to avoid the conclusion that bare hydrodynamic ensembles *cannot* decay rapidly to local equilibrium after all. That such ensembles contain slowly decaying initial terms is confirmed by the identity

$$0 \equiv \Xi_0(t) \equiv \Xi_0^l(t) + \Xi_0^i(t) \tag{85}$$

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[15] The fact that the constraint on $\hat{j}(\alpha)$ has been removed in ρ_A should make it an even *better* candidate for a local equilibrium assumption according to the criteria in Section 5.3.

(compare the case for $\mathbf{k} \neq 0$ in Section 4). Clearly the "initial" term $\Xi_0^i(t)$ decays just as slowly as $\Xi_0^l(t)$ itself!

Strictly speaking, the existence of a counterexample on the level of two-mode processes against the method of bare hydrodynamic decomposition invalidates the derivation of the mode-coupling formula given above. In the next section, however, we shall construct a refined decomposition which permits local equilibrium assumptions to be made without leading to inconsistencies. The derivation of the mode-coupling formula is correspondingly modified, and it will become clear why, for current–current correlation functions, the results are unaffected by the refinements.

6. DECOMPOSITION IN DRESSED HYDRODYNAMIC ENSEMBLES

6.1. The Need for Refinements

From the discussion of the previous section one can draw the following conclusions: (i) The local equilibrium assumption in the context of bare hydrodynamic ensembles must be in error. (ii) This error may or may not manifest itself, depending on the correlation function studied, i.e., depending on which moments of the distribution function are actually calculated.

In fact, in all strictly linear theories of nonequilibrium phenomena, errors involving higher moments of conserved quantities will not have any consequences. The asymptotic behavior of the current–current correlation functions, however, is, as we have seen, determined by two-mode processes; i.e., the *variance* in the distribution of conserved quantities plays a crucial role.

As a result, the naive expectation that almost any constrained ensemble rapidly develops into a normal state is in need of revision. We shall argue below that the failure of the decomposition used in the previous section is due to a mismatch of the initial variance and that of a local equilibrium state. The argument leads to a definite proposal for a refined decomposition. As an implication, we shall state our local equilibrium assumption in its ultimate version. Finally, the computational consequences of these refinements will be considered.

The counterexample of the previous section can be sharpened so that it points directly to the error introduced by the local equilibrium assumption in the context of bare hydrodynamic ensembles. Replace the current $\hat{j}$ in (79) by the quantity

$$[aa] \equiv aa - v_c^{-1}\langle aaa\rangle \cdot a - \langle aa\rangle \tag{86}$$

where $a = a(\beta)$. Clearly, $\langle [aa]\rangle$ and, in addition, $\langle [aa]a\rangle$ vanish identically. The corresponding "correlation function" $\Xi_{\alpha\beta}(t) = v_c^{-1}\langle [aa]e^{-t\mathscr{L}}\Delta(\alpha)\rangle$

measures the (vanishing) deviation of the variance of a from its equilibrium value $\langle aa \rangle$.

However, by the arguments of the previous section, one again finds a $t^{-3/2}$ tail in the local equilibrium part of $\Xi_{\alpha\beta}(t)$, given by (83) with $\mathscr{A}^{rr} = v_c^{-1}\langle [aa] a^r a^r \rangle$. From the identity (85) one then concludes that the approach of the locally vanishing variance to its local equilibrium value, contrary to assumption, cannot be fast. Physically, the buildup of a variance in the initially sharp local distribution of conserved quantities involves *transport* of such quantities and is therefore itself a hydrodynamic process.

We are thus led to the conclusion that only constrained ensembles in which the variances (and more generally, *all* higher moments) of conserved quantities already have their local equilibrium values should be expected to approach a local equilibrium state rapidly. The problem is then how (if possible) to decompose an arbitrary nonequilibrium ensemble into ensembles with this special property.

6.2. The Distribution $f(A)$

In order to make the above considerations precise, we shall consider the distribution function of conserved quantities $f(A)$ associated with an arbitrary initial nonequilibrium ensemble $\rho(\Gamma; 0)$. By definition,

$$f(A) = \int d\Gamma\, \rho(\Gamma; 0)\, \delta(a(\Gamma) - A) \tag{87}$$

The corresponding distribution associated with a local equilibrium state is given as

$$f_l(A|B) = \langle \delta(a(\Gamma) - A) \rangle_l = \langle \delta(a - A) \exp(a^\dagger \cdot B) \rangle / \langle \exp(a^\dagger \cdot B) \rangle \tag{88}$$

Although they will not be of direct relevance to the arguments below, we make some remarks on f_l of (88).

(i) In (88), $a(\Gamma)$ can be interpreted as the column vector of M Fourier variables $\{a_{\mathbf{k}}^s(\Gamma)\}$ *or* of M cell variables $\{a^s(\Gamma; \alpha)\}$.

(ii) The cell size was chosen such that cell–cell correlations can be neglected; i.e., in cell language f_l factorizes

$$f_l(A(\alpha), A(\beta),\ldots | B(\alpha), B(\beta),\ldots) = \prod_\alpha f_l(A(\alpha) | B(\alpha)) \tag{89}$$

Given the factorization (89), f_l does not, however, factorize with respect to Fourier variables.

(iii) From the definition (88) and with (89) one readily shows that f_l for a cell has the form

$$f_l(A(\alpha) | B(\alpha; t)) = f_{\mathrm{eq}}(A(\alpha) - \langle a(\alpha) \rangle_l | b_0 + B(\alpha; t)) \tag{90}$$

i.e., f_l has the form of an equilibrium distribution $f_{\mathrm{eq}}(A|b_0) = \langle\delta(a(\alpha) - A)\rangle$, where b_0 stands for the temperature and chemical potential in equilibrium.

(iv) As can be seen from (90), both the mean *and* the variances will shift slowly (by one-, two-mode, etc., processes) toward their values in total equilibrium.

6.3. The Decomposition

We return to the problem of decomposition of an arbitrary initial ensemble $\rho(\Gamma; 0)$, and write it as a linear superposition

$$\rho(\Gamma; 0) = \int dB\, g(B)\rho_B(\Gamma; 0) \tag{91}$$

where the weight function $g(B)$, which integrates to unity, does not have to be semi-positive definite. The so-called "B components" ρ_B must fulfill the requirements

$$\begin{aligned}\int dA\, A^n f_l(A|B) &= \int d\Gamma\, a^n(\Gamma)\rho_B(\Gamma; 0)\\ &= \int dA\, A^n \int d\Gamma\, \delta(a(\Gamma) - A)\rho_B(\Gamma; 0)\end{aligned} \tag{92}$$

i.e., *all* hydrodynamic moments of ρ_B must have the same values as in the local equilibrium state defined by the fields B. This property motivates the term "dressed hydrodynamic ensemble" for the B component ρ_B, as opposed to the bare hydrodynamic ensembles[16] used in Section 5.

Equation (92) does not define ρ_B uniquely. *One* possible form of ρ_B can be constructed as follows. Write $\rho_B(\Gamma; 0) = \rho(\Gamma; 0)f_l(a(\Gamma)|B)C(a(\Gamma))$. Consistency with (92) then requires that $C(a)$ be chosen such that $C(A) \times \int d\Gamma\, \rho(\Gamma; 0)\, \delta(a(\Gamma) - A) = 1$. By (87), the resulting ρ_B has the form

$$\rho_B(\Gamma; 0) = \rho(\Gamma; 0)f_l(a(\Gamma)|B)/f(a(\Gamma)) \tag{93}$$

Operation with $\int d\Gamma\, \delta(a(\Gamma) - A)$ on both sides of (91) gives, for any ρ_B satisfying (92),

$$f(A) = \int dB\, g(B)f_l(A|B) \tag{94}$$

This integral equation defines $g(B)$. [We assume that $g(B)$ exists for the initial ensembles used in this paper.] As a simple example, the B component of a local equilibrium state with $f(A) = f_l(A|B')$ is clearly the local equilibrium

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[16] There is close, but not perfect, analogy between the terms bare and dressed used here for the distributions in a single cell, and the standard ones of micro and grand used in the context of equilibrium ensembles in the total volume.

state itself. From (94) one finds $g(B) = \delta(B - B')$ in that case. The solution of (94) is not known in general, however. Fortunately, the full solution is not needed. For the purpose of calculating the long-time asymptotics of the correlation functions, knowledge of the first few moments of $g(B)$ suffices.

6.4. The Local Equilibrium Assumption

Associated with the decomposition (91), the final form of our local equilibrium assumption reads:

Any B component, or dressed hydrodynamic ensemble, $\rho_B(\Gamma; 0)$, satisfying (92) will rapidly approach a normal solution $\rho_n(\Gamma; t \,|\, B)$ of the form (14).

By construction, the distribution of conserved quantities in ρ_B already equals that of the associated local equilibrium state at $t = 0$. If one postulates that *all* slow decay is due to hydrodynamic processes involving combinations of modes, the above assumption takes on its strongest form. The key word "rapidly" should then be read as meaning "exponentially fast."

We shall not insist on this extreme interpretation of the assumption, however. In the context of the asymptotics of the time correlation functions, one does not need moments of $f(A)$ higher than the second. An interpretation of the crucial word "rapidly," sufficiently stringent for our purposes, is thus: faster than the slowest two-mode processes. In this context, the requirements (92) on all moments of ρ_B beyond the second-order ones should be viewed as a formal convenience rather than a real necessity.

It is instructive to review the intuitive criteria listed in Section 5 for the soundness of a local equilibrium assumption on the background of the refinements of the present section. The criteria are clearly not sufficient and, in particular, the second one must be revised. The states on which the present local equilibrium assumption is formulated are restricted by many *more* constraints than those of Section 5. The additional constraints are, however, of a special nature: they force the initial distribution $\rho(\Gamma; 0)$ to be *closer* to the local equilibrium form.

6.5. Application to $C_{\mathbf{k}}(t)$

The final task is the application of the decomposition (91), and the correspondingly refined version of the local equilibrium assumption, to the calculation of $C_{\alpha\beta}(t)$ in the form (67)[17]

$$C_{\alpha\beta}(t) = v_c^{-1}\langle \hat{j}(\beta) e^{-t\mathscr{L}} \hat{j}(\alpha)\rangle = v_c^{-1} \int dJ\, f_{\text{eq}}(J) J \langle \hat{j}(\beta; t)\rangle_J \tag{95}$$

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[17] Again we use cell language for simplicity. The argument can, of course, be given an equivalent formulation in Fourier language.

The average $\langle \hat{j} \rangle_J$ involves the constrained ensemble $\rho_J(\Gamma; 0)$ of (65). The problem is then to decompose $\rho_J(\Gamma; 0)$ into B-components $\rho_{JB}(\Gamma; 0)$, or, specifically, to calculate the necessary moments of $g_J(B)$, which solves the equation

$$f_J(A) = \int dB\, g_J(B) f_l(A|B) \tag{96}$$

This equation is a special case of (94), with $f_J(A) = \langle \delta(a - A) \rangle_J$. Assuming that $g_J(B)$ has been determined in sufficient detail, we appeal to the local equilibrium assumption of Section 6.4 and, with (91), write

$$\rho_J(\Gamma; t) = \int dB\, g_J(B) \rho_{JB}(\Gamma; t) \simeq \int dB\, g_J(B) \rho_n(\Gamma; t|B) \tag{97}$$

The decay of the B-components toward the normal solution is assumed to be sufficiently uniform in B that the integration in (97) causes no difficulties.

The contribution to $C_{\alpha\beta}$ from the local equilibrium part of ρ_n is easily calculated, by the same steps as in Section 5.4, to have the asymptotic form

$$\begin{aligned} C_{\alpha\beta}(t) \simeq v_c^{-1} \int dJ\, f_{\text{eq}}(J) J \int dB\, g_J(B) \tfrac{1}{2} \sum_{r,s} \langle \hat{j}(\beta) a^r(\beta) a^s(\beta) \rangle \\ \times G^r(\beta; t|\alpha) G^s(\beta; t|\alpha) B^r B^s \end{aligned} \tag{98}$$

This formula shows that only the second moments of $g_J(B)$ (actually only weighted averages of these moments) are needed in the calculation of the long-time behavior of $C_{\alpha\beta}(t)$. In the appendix it is shown that the moments are given uniquely by

$$\begin{aligned} v_c \int dJ\, f_{\text{eq}} J \int dB\, g_J(B) BB &= v_c^{-1} \langle \hat{j}[aa] \rangle \\ &\equiv v_c^{-1} \langle \hat{j} \{ aa - v_c^{-1} \langle aaa \rangle \cdot a - \langle aa \rangle \} \rangle \end{aligned} \tag{99}$$

where all the quantities in the average refer to cell α. By virtue of the orthogonality relations $\langle \hat{j} \rangle = \langle \hat{j} a \rangle = 0$, it follows directly that

$$v_c \int dJ\, f_{\text{eq}}(J) J \int dB\, g_J(B) B^r B^s = v_c^{-1} \langle \hat{j} a^r a^s \rangle = \mathscr{A}^{rs} \tag{100}$$

With $\mathscr{A}$ defined as in (76). Insertion of (100) into (98) yields the mode-coupling formula in the form (77), or equivalently (78).

The crucial difference in the calculations of Sections 5 and 6 leading to the mode-coupling formula lies in the recombination of the initially decomposed ensembles as expressed by Eqs. (76) and (99), respectively. Due to the orthogonality of $\hat{j}$ to 1 and to a, the result is the same in both cases.

However, the counterexample of Sections 5.5 and 6.1 against the decomposition in bare hydrodynamic ensembles is harmless in the context of dressed ensembles. The difference from the current–current correlation functions amounts to the replacement in (76) and (99) of $\hat{j}$ by unity. In (76), the result is still nonzero in certain cases, resulting in serious inconsistencies. The corresponding expression obtained from (99), however, vanishes identically.

In conclusion, with the decomposition in dressed hydrodynamic ensembles, the mode-coupling formula has been given a phenomenological derivation by a method free from internal inconsistencies.

7. DISCUSSION

In this paper we have studied the long-time and small-wavenumber behavior of true correlation functions C and projected correlation functions U on the basis of essentially *one* assumption, that of a rapid decay to a "normal" state close to local equilibrium of a carefully constructed class of initial states. The central result is the asymptotic validity of the self-consistent mode-coupling formula (78) for the dissipative matrix U. The asymptotic behavior of C follows through the identity (57) connecting U and C.

To bring out the subtlety of the local equilibrium assumption, we have chosen a presentation of stepwise refinement. The failure of the seemingly sensible approach of Section 5 is analyzed and provides motivation for our final version of the crucial assumption, as presented in Section 6.

The remainder of this paper is organized as follows. The relations between various local equilibrium derivations of the mode-coupling formula are reviewed in Sections 7.1–7.3. A more general class of correlation functions is touched upon in Section 7.4. Sections 7.5 and 7.6 concern the form of the hydrodynamic equations beyond the linear and the Navier–Stokes regimes, respectively. Finally, the status of the mode-coupling formula in two dimensions and in the critical region is considered under points in Sections 7.7 and 7.8.

7.1. Earlier Derivations

We first summarize our views, based on the results of this paper, on the various types of local equilibrium derivations of the mode-coupling formula. A common feature of all such derivations is a decomposition of the form (48)

$$C(t) = \int ds\, h(s) \langle \hat{j}(t) \rangle_s \tag{101}$$

The asymptotically relevant parts of the projected current in (101) can be written as two-mode terms

$$\hat{j} \sim \sum_{i,j} \langle \hat{j} a^i a^j \rangle a^i a^j \tag{102}$$

and the constrained average of the current can then be expressed as a sum of terms of the type

$$\langle \hat{j}(t) \rangle_s \sim \sum_{i,j} \langle \hat{j} a^i a^j \rangle \langle a^i(t) a^j(t) \rangle_s \tag{103}$$

If the constrained averages with nonvanishing coefficients can be decoupled into a product of averages $\langle a^i(t) \rangle_s \langle a^j(t) \rangle_s$, then, since time evolution according to linear hydrodynamics does not affect this factorization, one has

$$\langle \hat{j}(t) \rangle_s \sim G^i(t) G^j(t) \langle a^i \rangle_s \langle a^j \rangle_s = G^i(t) G^j(t) \langle a^i a^j \rangle_s \tag{104}$$

Insertion of this expression into (101) leads to the standard mode-coupling result for the asymptotic behavior. Clearly, such a factorization holds only provided that

$$\langle a^i a^j \rangle_s = \langle a^i \rangle_s \langle a^j \rangle_s \tag{105}$$

This requirement is fulfilled in the following cases. (i) The calculation of the kinetic terms of the correlation functions as performed in I. In that case the two-mode terms always involve such combinations of vector and scalar fields that factorization is an immediate consequence of symmetry. (ii) The bare hydrodynamic specification of Section 5 in this paper. There factorization holds by construction. (iii) The same can be said for the initial decomposition used by Pomeau,(17) namely complete specification in the Γ space associated with one cell. (iv) The extreme example along this line is complete initial specification in the Γ space of the *whole* system, i.e., $\rho_s(\Gamma; 0) = \delta(\Gamma - \Gamma_s)$. Again, (105) holds by construction. In all these cases, a local equilibrium assumption, right or wrong, leads to the mode-coupling results.

On the other hand, the requirement (105) is *not* fulfilled if one generalizes the method of I to include potential terms by specifying, in addition to the position and velocity of particle 1, also the relative position $\mathbf{r}_{12}$ of particle 2. With this specification, the factorization holds in every case except for the bulk viscosity, where two scalar fields are involved (for details, see II). On the basis of the (too) simple picture underlying the above arguments, one would attribute the failure of this decomposition to an insufficient resolution of the initial ensemble with respect to its hydrodynamic content; i.e., different values of, say, the initial number densities would not be properly separated, but would in some inconsistent sense be treated on the average.

7.2. The Exclusive Status of *B* Components

The line of reasoning sketched above is, however, not sufficiently discriminating. The factorization property (101) cannot serve as a criterion of validity for local equilibrium derivations of the mode-coupling formula. This is already obvious from the extreme example mentioned. An initial ensemble $\delta(\Gamma - \Gamma_s)$ will always retain its form and will never develop into a state of local equilibrium. (It follows that, in general, approach to local equilibrium cannot be pointwise, but can only hold in a coarse-grained sense.)

More illuminating than the failure of this extreme example is the counterexample against the decomposition in bare hydrodynamic ensembles discussed in Sections 5 and 6. The relevance of that counterexample to the derivation of the mode-coupling formula is stressed by the fact that the failure of the corresponding local equilibrium assumption manifests itself already on the crucial level of two-mode processes. As a result, one is forced to revise drastically the naive expectation that almost any constrained initial ensemble rapidly decays to local equilibrium. In fact, we argue in Section 6 that rapid decay to local equilibrium is an *exclusive* property of a special class of initial states.

This exclusiveness can be given further support by the following argument. Any constrained initial state can be written as a sum of two states

$$\rho_s(\Gamma; 0) = c_1\rho_2(\Gamma; 0) + c_2\rho_2(\Gamma; 0) \tag{106}$$

with $c_1 + c_2 = 1$ and $c_1, c_2 > 0$. By the linearity of the Liouville equation, the evolutions of the two states are mutually independent. With the state $\rho_s(t)$ are associated the fields $B_s(t)$ according to (15). Correspondingly, $B_1(t)$ and $B_2(t)$ are associated with $\rho_1(t)$ and $\rho_2(t)$. A local equilibrium assumption on ρ_s would yield

$$\rho_s(\Gamma; 0) \to \rho_n(\Gamma; t \,|\, B_s) \tag{107}$$

On the other hand, the same assumption on ρ_1 and ρ_2 would yield

$$\rho_s(\Gamma; 0) \to c_1\rho_n(\Gamma; t \,|\, B_1) + c_2\rho_n(\Gamma; t \,|\, B_2) \tag{108}$$

Equations (107) and (108) are only compatible[18] in the degenerate case when $B_s = B_1 = B_2$. In conclusion, the local equilibrium assumption can only be made on a *special class* of initial states.

The *form* of this special class is postulated on the basis of the experience gained by the study of the counterexample against the decomposition used in Section 5. The final formulation of our local equilibrium assumption, as given in Section 6, applies to *dressed* hydrodynamic initial states, for which

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[18] Note again that the problem of incompatibility between (107) and (108) does not show up if one sticks to *linear* order in *B*.

all moments of conserved quantities already have their local equilibrium values.

We have, of course, no *proof* of the unique status of the B components, or dressed hydrodynamic ensembles, as being the only states with a truly rapid decay toward local equilibrium. Such a proof would have to come from a more fundamental theory. We would like to point out, however, that in its weakest form (which is all that is needed for the derivation of the mode-coupling theory) our postulate is sufficiently precise to be tested by computer experiments. The issue is then to verify that dressed hydrodynamic ensembles decay *faster* than the slowest two-mode processes, whereas bare ensembles *do* have components as slow as those processes.

7.3. Refined Version of Factorization

With the refined local equilibrium assumption associated with B components, insistence on the simple factorization property (105) must be relaxed. From the results of the appendix, however, one has, in the case of decomposition in dressed hydrodynamic ensembles, for all times ($t = 0$ included),

$$\begin{aligned}\langle aa\rangle_{JB} = \langle aa\rangle + \langle aaa\rangle\cdot B + v_c{}^2 BB + \tfrac{1}{2}\langle aaaa\rangle_c : BB \\ + \text{three-mode terms} + \cdots \qquad (109)\end{aligned}$$

The last term is $O(v_c^{-1})$ with respect to $v_c{}^2BB$ and can be neglected. When a properly weighted average over J and B has been performed, the first term on the right of (109) vanishes since $\langle \hat{J}\rangle = 0$. The second term similarly vanishes since $\langle \hat{J}a\rangle = 0$. *Effectively*, then, one has

$$\langle aa\rangle_{JB} \sim v_c{}^2 BB \qquad (110)$$

This more refined factorization property replaces (105) in the case of dressed ensembles. The essential fact is that the deviations, in (109), from true factorization are either higher order (in v_c^{-1} or B), or they are constants or one-mode terms.

The reason for the failure of the straightforward generalization of paper I to include potential terms, sketched in Section 7.1, must be revised in the light of (109)–(110). The point is not that the initial ensemble in that case is insufficiently resolved with respect to its hydrodynamic content, but that it is *incorrectly* resolved. It is true that the initial specification of $\mathbf{v}_1$, $\mathbf{r}_1$, and $\mathbf{r}_{12}$ does not uniquely fix, say, the total number of particles. Consequently, it amounts to treating initial states with different numbers of particles together. But the same can be said about the initial decomposition into B components. Only in the latter case, however, does the initial distribution of conserved quantities coincide with the local equilibrium distribution. Thus, by our fundamental assumption of Section 6, only in the latter case is the approach to the associated normal state rapid.

In short, specification of $\mathbf{v}_1$, $\mathbf{r}_1$, and $\mathbf{r}_{12}$ does *not* imply relations of the type (109)–(110) in general. For reasons of symmetry, however, this defect only manifests itself when one considers the correlation function pertaining to the bulk viscosity. A similar error is made with *all* the decompositions mentioned in Section 7.1. But since the factorization (105) holds, the error goes unnoticed as far as derivations of mode-coupling results are concerned.

On this background one should not regard the method of paper I as a general one, by which, after straightforward extensions, the asymptotics of any desired correlation function can be calculated. Rather, that method should be viewed as a simple procedure, valid in the specific cases where it is actually used, but with no claim on generality. The *general* framework for the application of local equilibrium assumptions is provided by the present paper.

7.4. Dressed Current Distribution

In evaluating the current correlation function in Section 6, we first decomposed the equilibrium ensemble into initial ensembles $\rho_J(\Gamma; 0)$ with a sharp value of the current (with, in other words, a bare current distribution). A decomposition of $\rho_J(\Gamma; 0)$ into B components, as carried out in Section 6.3, is conceptually only possible if the current $\hat{j}(\Gamma)$ cannot be expressed completely in terms of powers of $a(\Gamma)$'s. This issue is avoided if one uses an initial ensemble with a dressed current distribution, which has basically an equilibrium variance in the currents, namely

$$\rho_X(\Gamma; 0) = \rho_{\mathrm{eq}}(\Gamma) \exp[\hat{j}^\dagger(\Gamma) \cdot X] / \langle \exp[\hat{j}^\dagger \cdot X] \rangle \tag{111}$$

and calculates the correlation function from $C_X(t) = V^{-1} \langle \hat{j}(t) \rangle_X$. Here $\hat{j}(\Gamma)$ and X stand either for the cell variables $\hat{j}(\Gamma; \alpha)$ and $X(\alpha)$ or for the Fourier variables $\hat{j}_{\mathbf{k}}(\Gamma)$ and $X_{\mathbf{k}}$. The correlation function of interest $C(t)$ is then obtained as the coefficient of the first power of X in the X expansion of $C_X(t)$. The calculation in the appendix requires minor adaptations for that case.

7.5. General Nonequilibrium Ensemble

The postulate that local equilibrium assumptions can only be consistently made on B components also has consequences for the derivation of *nonlinear* hydrodynamics. By this postulate, the validity of the derivation in Section 3 of hydrodynamic equations is restricted to initial B components $\rho_B(\Gamma; 0)$. An arbitrary initial ensemble $\rho(\Gamma; 0)$ would thus have to be decomposed as in Section 6.2,

$$\rho(\Gamma; 0) = \int dB\, g(B) \rho_B(\Gamma; 0) \tag{112}$$

with $g(B)$ determined from (94). The results of Section 3, valid for each $\rho_B(\Gamma; 0)$, would finally have to be recombined according to (112). What the precise form of the corresponding nonlinear hydrodynamic equations would be, and to what extent they would depend on the initial ensemble, still has to be investigated. The above procedure has no consequences at the level of *linear* hydrodynamic equations, where one can still make the local equilibrium assumption for an essentially arbitrary initial state.

7.6. Corrections to Navier–Stokes and $C(t)$

On the basis of the local equilibrium assumption, we have in Section 4 unambiguously associated the self-consistent mode-coupling formula with the projected correlation functions U rather than with the true correlation functions C. As was noted already in the introduction, the difference between the two disappears in the limit $\mathbf{k} \to 0$. The calculations in I and II apply in precisely that limit. However, for small but finite $\mathbf{k}$, the mode-coupling formulas can be viewed as a set of coupled integral equations, which can be solved iteratively in successive approximations. In three dimensions they yield an infinite series of corrections to Navier–Stokes hydrodynamics [of $O(k^{3-2^{-n}})$, with $n \geqslant 1$, i.e., all of them below the Burnett order, $O(k^3)$], and to the $t^{-3/2}$ tails in the Green–Kubo integrands [of $O(t^{-(2-2^{-n})})$ with $n \geqslant 2$]. Calculations along these lines have been carried out by Pomeau⁽¹⁰⁾ and Ernst and Dorfman.⁽¹¹⁾

Basic to such calculations is a definite choice (and the two possibilities are mutually exclusive): *either* the mode-coupling formula is assumed to be valid for $C_{\mathbf{k}z}$ *or* it is valid for $U_{\mathbf{k}z}$. The calculations mentioned are based on the second choice, which we have now shown to be the correct one. In Refs. 10 and 11 the actually contributing integrals are all such that $z \gtrsim k$, and thus the distinction between U and C, although conceptually basic to the method used, is not important in that range of the variables z and k [see (57)]. The situation is different for self-diffusion, however. The corresponding mode-coupling calculations in three dimensions have been carried out by de Schepper and Ernst,⁽²⁶⁾ and in that case, integrals with $z = O(k^2)$ do contribute. For such values of z there is not only a conceptual, but also a numerical difference between U and C.

Finally, we note that aside from the phenomenological derivation given in this paper, independent evidence exists for the validity of the mode-coupling formula. We refer, in particular, to arguments based on kinetic theory. For low densities, the $k^{5/2}$ correction to hydrodynamics, the $t^{-7/4}$ correction to the tail in the velocity autocorrelation function, and the $t^{1/4}$ correction to the time-dependent super-Burnett coefficient have been calculated explicitly,(6,27) and they are all in perfect agreement with the low-density limit of the mode-coupling results. For higher densities the arguments of Résibois *et al.*(9) can serve as an alternative basis for mode-coupling calculations.

7.7. Two Dimensions

It is hard to see why the derivation of the mode-coupling formula (78), without explicit expressions of the Green's functions, should only be sound for dimensionality $d \geqslant 3$. If, on the other hand, the Green's functions are assumed to be of the Navier–Stokes form (to lowest order), (78) leads to nonintegrable Green–Kubo integrands, i.e., to serious inconsistencies in two dimensions. It is tempting to blame this failure, not on the mode-coupling formula itself, but on the explicit representation used for the Green's functions.

However, the self-consistent asymptotic behavior(2) that can be derived from (78) is only of very limited interest when one wants to compare with existing computer experiments for $d = 2$. The time needed to approach *that* asymptotic behavior is presumably very long indeed. It is much more realistic for times up to 80–100 mean free times to base the calculations on a set of "bare" transport coefficients (usually chosen to have their Enskog values) as a first approximation, and to proceed from there by iteration. In fact, the lowest order result shows remarkable agreement with experiments(28) in this time regime, both for the velocity autocorrelation function(4) and for the time-dependent super-Burnett coefficient!(27)

This astonishing agreement can be "understood" on the basis of the plausible postulate that higher iterates have not yet, on the relevant time scale, obtained their asymptotic form (which would have amounted to important corrections!). Plausible as this postulate may be, it has not yet been backed up by convincing calculations, and consequently, the status of the mode-coupling formula in two dimensions remains somewhat obscure.

7.8. Critical Phenomena

The mode-coupling formula with k-dependent amplitudes has been widely used as a basis for calculations of the dynamical properties of fluids close to critical points. Our present derivation of the formula is restricted by the assumption that $k \ll \xi^{-1}$. Thus it is valid in the so-called hydrodynamical region, even close to the critical point. It remains to be seen, however, whether the critical region, where $k \gg \xi^{-1}$, can be understood on the basis of a suitably modified version of the fundamental assumption in this paper.

APPENDIX

For the evaluation of the moments in (98), i.e.,

$$\overline{B \cdots B} \equiv \int dB\, g_J(B) B \cdots B \tag{A.1}$$

one does not need to solve Eq. (96). It is sufficient to expand $f_l(A|B)$ in powers of B (measuring the small deviations from total equilibrium), and thus determine the moments (A.1). For the sake of definiteness we use cell language with the current in cell α specified. Since the distribution function for the different cells factorizes at $t = 0$, we only need to consider cell α. The expansion of $f_l(A(\alpha)|B(\alpha))$ in (88)–(89) in powers of $B(\alpha)$ takes the form

$$f_l(A|B) = f_{\mathrm{eq}}(A)[1 + A \cdot B + \{\tfrac{1}{2}AA - \langle aa\rangle\} : BB + \cdots] \tag{A.2}$$

Inserting (A.2) into (96), multiplying both sides of the equation by, respectively, A, AA,..., and integrating over A, we find an infinite set of coupled linear equations relating the moments $\langle a\rangle_J$, $\langle aa\rangle_J$,... with $\bar{B}$, $\overline{BB}$,...:

$$\begin{aligned} \langle a\rangle_J &= \langle aa\rangle \cdot \bar{B} + \tfrac{1}{2}\langle aaa\rangle : \overline{BB} + \cdots \\ \langle aa\rangle_J &= \langle aa\rangle + \langle aaa\rangle \cdot \bar{B} + \tfrac{1}{2}\{\langle aaaa\rangle - \langle aa\rangle\langle aa\rangle\} : \overline{BB} + \cdots \end{aligned} \tag{A.3}$$

where we used $\int dA\, f_J(A) A \cdots A = \langle a \cdots a\rangle_J$ and a similar expression for $\langle a \cdots a\rangle$.

In order to solve Eqs. (A.3) we have to determine the volume dependence of the fluctuation formulas

$$\langle aa \cdots a\rangle = \int \cdots \int_{v_c} d\mathbf{r}_1\, d\mathbf{r}_2 \cdots d\mathbf{r}_n\, F(12 \cdots n) \tag{A.4}$$

where

$$F(12 \cdots n) = \langle \delta a(\mathbf{r}_1)\, \delta a(\mathbf{r}_2) \cdots \delta a(\mathbf{r}_n)\rangle \tag{A.5}$$

with $F(1) = \langle \delta a(\mathbf{r}_1)\rangle = 0$. This will be done by introducing cluster functions. The functions $F(12 \cdots n)$ have the product property for separated configurations, i.e., if the smallest distance between two groups of particles $(12 \cdots l)$ and $(l + 1,..., n)$ is much larger than the correlation length ξ, then $F(12 \cdots l, l + 1,..., n)$ factorizes into $F(12 \cdots l)F(l + 1,..., n)$. Due to this product property we can introduce in the standard way cluster functions $C(12 \cdots l)$, which vanish for separated configurations. Hence we have

$$\int \cdots \int_{v_c} d\mathbf{r}_1 \cdots d\mathbf{r}_l\, C(12 \cdots l) \equiv \langle aa \cdots a\rangle_c = O(v_c) \tag{A.6}$$

The cluster expansion is given by the relations

$$\begin{aligned} F(12) &= C(12) \\ F(123) &= C(123) \\ F(1234) &= C(12)C(34) + C(13)C(24) + C(14)C(23) + C(1234) \qquad \text{(A.7)} \\ F(12345) &= \textstyle\sum C(12)C(345) + C(12345) \\ &\vdots \end{aligned}$$

where $\sum$ indicates a sum over partitions. Notice that cluster functions for isolated particles do not occur in (A.7) since $C(1) = F(1) = 0$. For the integrated quantities in (A.4) one has an identical set of relations in which $F(12 \cdots n)$ is replaced by $\langle a_1 a_2 \cdots a_n \rangle$ and $C(12 \cdots n)$ by $\langle a_1 a_2 \cdots a_n \rangle_c$, the last quantity being of $O(v_c)$.

Using (A.7) and $\langle aa \rangle = v_c \mathbf{1}$, we can write (A.3) as

$$\langle a \rangle_J = v_c \bar{B} + \tfrac{1}{2} \langle aaa \rangle : \overline{\bar{B}\bar{B}} + \cdots \tag{A.8a}$$

$$\langle aa - \langle aa \rangle \rangle_J = \langle aaa \rangle \cdot \bar{B} + v_c{}^2 \overline{\bar{B}\bar{B}} + \tfrac{1}{2} \langle aaaa \rangle_c : \overline{\bar{B}\bar{B}} + \cdots \tag{A.8b}$$

It is clear now that the last term in (A.8b) is of relative order v_c^{-1} and can be neglected. However, to give a consistent solution of (A.8) we have to know the relative volume dependence of all terms on the right- and left-hand sides of these equations. Only then can we decide whether we should keep, say, $\langle aa \rangle = O(v_c)$, compared to $v_c{}^2 \overline{\bar{B}\bar{B}}$. The volume dependence of $\langle a \rangle_J$ and $\langle aa \rangle_J$ is not clear a priori. We need, however, only

$$\overline{\overline{B \cdots B}} \equiv \int dJ f_{\text{eq}}(J) J \overline{B \cdots B} \tag{A.9}$$

From the relations (A.8) and (A.9) and $\int dJ f_{\text{eq}}(J) J \langle a \cdots a \rangle_J = \langle \hat{j} a \cdots a \rangle$ we deduce at once

$$\langle \hat{j} a \rangle = v_c \bar{\bar{B}} + \tfrac{1}{2} \langle aaa \rangle : \overline{\overline{\bar{B}\bar{B}}} + \cdots \tag{A.10a}$$

$$\langle \hat{j} [aa - \langle aa \rangle] \rangle = \langle aaa \rangle \cdot \bar{\bar{B}} + v_c{}^2 \overline{\overline{\bar{B}\bar{B}}} + \tfrac{1}{2} \langle aaaa \rangle_c : \overline{\overline{\bar{B}\bar{B}}} + \cdots \tag{A.10b}$$

The averages $\langle \hat{j} a \rangle$ vanish identically for the currents entering in the correlation functions. If we want to cover in our discussion also currents with components parallel to a, then $\langle \hat{j} a \rangle = O(v_c)$ typically. Under the assumption that the higher order terms in (A.10) can be neglected, Eq. (A.10b) implies that $\overline{\overline{\bar{B}\bar{B}}}$ is at most of $O(v_c^{-1})$, since $\langle \hat{j}(aa - \langle aa \rangle) \rangle$ is $O(v_c)$. Consequently, we have from (A.10a) that $\bar{\bar{B}} = O(1)$. In our case, with $\langle \hat{j} a \rangle = 0$; B vanishes.

Now, it is clear that the last term in (A.10a) and (A.10b) is of relative order v_c^{-1} and can be neglected, so that

$$\overline{\overline{\bar{B}\bar{B}}} = v_c^{-2} \langle \hat{j}(aa - v_c^{-1} \langle aaa \rangle \cdot a - \langle aa \rangle) \rangle \tag{A.11}$$

Combination of (A.9) and (A.11) yields Eq. (99) in the text.

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