Hydrodynamics and Fluctuations Outside of Local Equilibrium: Driven Diffusive Systems

Gregory L. Eyink

Department of Mathematics

Building No. 89

University of Arizona

Tucson, AZ 85721

Joel L. Lebowitz

Departments of Mathematics and Physics

Rutgers University

Hill Center-Busch Campus

New Brunswick, NJ 08903

Herbert Spohn

Theoretische Physik

Ludwig-Maximilians-Universität

Theresienstr. 37, 80333

München, Germany

Abstract

We derive hydrodynamic equations for systems not in local thermodynamic equilibrium, that is, where the local stationary measures are "non-Gibbsian" and do not satisfy detailed balance with respect to the microscopic dynamics. As a main example we consider the *driven diffusive systems*(DDS), such as electrical conductors in an applied field with diffusion of charge carriers. In such systems, the hydrodynamical description is provided by a nonlinear drift-diffusion equation, which we derive by a microscopic method of *nonequilibrium distributions.* The formal derivation yields a Green-Kubo formula for the bulk diffusion matrix and microscopic prescriptions for the drift velocity and "nonequilibrium entropy" as functions of charge density. Properties of the hydrodynamical equations are established, including an "H-theorem" on increase of the thermodynamic potential, or "entropy," describing approach to the homogeneous steady-state. The results are shown to be consistent with the derivation of the linearized hydrodynamics for DDS by the Kadanoff-Martin correlation-function method and with rigorous results for particular models.

We discuss also the internal noise in such systems, which we show to be governed by a generalized *fluctuation-dissipation relation*(FDR), whose validity is not restricted to thermal equilibrium or to time-reversible systems. In the case of DDS, the FDR yields a version of a relation proposed some time ago by Price between the covariance matrix of electrical current noise and the bulk diffusion matrix of charge density. Our derivation of the hydrodynamical laws is in a form—the so-called "Onsager force-flux form" —which allows us to exploit the FDR to construct the Langevin description of the fluctuations. In particular, we show that the probability of large fluctuations in the hydrodynamical histories is governed by a version of the Onsager "principle of least dissipation," which estimates the probability of fluctuations in terms of the Ohmic dissipation required to produce them and provides a variational characterization of the most probable behavior as that associated to least (excess) dissipation.

Finally, we consider the relation of long-range spatial correlations in the steady-state of the DDS and the validity of ordinary hydrodynamical laws. We discuss also briefly the application of the general methods of this paper to other cases, such as reaction-diffusion systems or magnetohydrodynamics of plasmas.

TABLE OF CONTENTS

6. Appendices

1 Introduction

(1.1) Description of the Program

This paper describes an approach to the derivation of hydrodynamical equations— including also the fluctuations around the deterministic behavior due to internal "molecular" noise—for microscopic interacting particle systems. The method is quite general but particular attention is given to cases where the system is not locally in thermodynamic equilibrium.

By "local equilibrium" we mean here that the system may be divided into cells small on the macroscopic scale but each containing a large number of molecules whose statistics are described by the equilibrium distributions (classical or quantum) of Gibbs. Many nonequilibrium phenomena commonly encountered are in this class, including ordinary hydrodynamics of fluids, heat conduction, Fickian diffusion of solutes, etc. Nonequilibrium situations of this kind generally occur either through the relaxation of imposed initial inhomogeneities of hydrodynamic densities or through the imposition of steady nonuniform thermodynamic potentials on the boundaries. Therefore, even if very large overall departures of the system from equilibrium occur, the small local regions are well-described to lowest order in gradients by a Gibbs distribution with parameters slowly varying across the system. Thermohydrodynamic fluid flow in Bénard cells is, for the convective regime with a large temperature difference, a standard example which is globally far from equilibrium.

Nevertheless, there is another large class of nonequilibrium systems, e.g. those locally driven by external fields, which can have statistics in small cells strongly deviating from those of Gibbs. In such cases, there may still be a hydrodynamic behavior associated to *locally stationary states* with slowly varying parameters, in which homogeneous, stationary measures of the driven dynamics, generally non-Gibbsian, play the role of equilibrium states. Examples of this sort include charge diffusion in semiconductors under applied electric fields, chemical reaction-diffusion processes with a local supply of reagents, magnetohydrodynamics of driven plasmas, etc. A crucial feature of these systems is that the local stationary measures are *non-*

reversible, i.e. they do not satisfy detailed balance with respect to the dynamics. Far less is known for these systems than for the local equilibrium ones. For example, there is no generally accepted prescription for the noise strength in a Langevin equation to describe the fluctuations around such a state, such as the fluctuation-dissipation relation of equilibrium systems.

The method we use here to derive the hydrodynamic laws is a formalism based upon *nonequilibrium distributions* which has been pioneered and applied by a number of authors, particularly H. Mori [1], D. N. Zubarev [2], and J. A. McLennan [3] among others. We may mention also the work of Ya. G. Sinai [4], who made such techniques the basis of an attempt to rigorously derive the Euler hydrodynamic equations of one-dimensional Hamiltonian particle systems. One of us (G.L.E.) has elsewhere reviewed this method for local-equilibrium states of classical Hamiltonian or quantum dynamics with conservation laws [5]. The objective there was to discuss the mathematical foundations of the method and to clarify the conditions required for a hydrodynamic description to be valid: the separation of scales, dynamical properties of ergodic type, etc. Here let us just say that the non-equilibrium distribution itself is a rigorous formula of Gibbs exponential type for a time-evolved local equilibrium distribution, a kind of "Girsanov formula" for the density with respect to a homogeneous reference measure. It is a suitable object for subsequent formal expansion in the small ratio-of-scales parameter ϵ, which yields the constitutive laws for fluxes and the hydrodynamic equations.

We shall discuss in this work the extension of this approach to systems without local equilibrium. In particular, we will discuss the new features that appear with local stationary measures of non-Gibbsian type and attempt to formulate explicitly the assumptions under which the derivations are obtained. Our method is constructive, since we introduce basic quantities that appear in the hydrodynamical laws by formally exact microscopic expressions which are calculable (in principle) analytically and (in practice) by molecular dynamics. These quantities are: (i) a *nonequilibrium entropy*, or a "fundamental equation" for the entropy as function of the hydrodynamic densities, expressed as a suitable large-volume limit; (ii) *drift* (or, *convection*) *velocities* as stationary averages of microscopic currents; and (iii) *Onsager relaxation* *coefficients* or transport coefficients, given by Green-Kubo formulae in terms of the microscopic dynamics.

¿From these microscopic quantities we construct also the statistical theories of fluctuations about the deterministic hydrodynamic behavior. It is possible to develop a nonequilibrium distribution method based upon the microscopic probability conservation for the empirical distributions (a "level-2" approach) adequate to derive a hydrodynamic Fokker-Planck equation for the fluctuations. [1] This has been done some time ago for local-equilibrium systems by Zubarev and Morozov [7], applying a general method of Zwanzig [8]. However, we shall follow here a more economical procedure. In fact, it is an important principle of statistical physics, emphasized by Einstein [9] and Onsager [10], that fluctuations are determined by hydrodynamics, so that a derivation of the latter also suffices to prescribe the distribution law of fluctuations. In the present context this connection is given by a *(generalized) fluctuation-dissipation relation* which we derive. For the small fluctuations described by a linear Langevin equation, the stationary distribution must be calculated to an approximation including correlations in the steady-state. For large deviations an "Onsager-Machlup action" functional is constructed which gives the probability of spontaneous fluctuations of the hydrodynamic histories, and which yields automatically a variational principle for the most probable behavior, a version of Onsager's "least-dissipation principle."

(1.2) Driven Diffusive Systems

As a main example of the formalism we discuss in the text the *driven diffusive systems* (DDS), and only consider other concrete areas of application in the conclusion section. The DDS have a long history in statistical physics, going back at least to the 1951 work of Wannier

[1] The "empirical distribution" in this method is just the delta-distribution $\hat{f}[\rho] = \prod_{\mathbf{r}} \delta(\rho(\mathbf{r}) - \hat{\rho}(\mathbf{r}))$, supported on the density function of the n-body system, $\hat{\rho}(\mathbf{r}) = \sum_{i=1}^{n} \delta(\mathbf{r} - \mathbf{r}_i)$. The latter is the "empirical density," or level-1 object. It is easy to check that the empirical distribution evolved under the microscopic dynamics satisfies a continuity equation of the form $\partial_t \hat{f}_t[\rho] + \int d\mathbf{r} \frac{\delta}{\delta\rho(\mathbf{r})} \hat{J}_t[\mathbf{r}, \rho] = 0$. When this is averaged with respect to a suitable ensemble of initial conditions, the Fokker-Planck equation results.

based upon a Boltzmann transport equation [11, 12]. Studying gaseous ionic conductors in a strong electric field, Wannier observed that inhomogeneities of charge density imposed upon the current-carrying state slowly spread about their drift displacement. He showed that the diffusion concept is still applicable in the nonequilibrium system and was able to calculate the diffusion tensor for the driven state. There are a broad class of physical situations in nature which exhibit a similar behavior. Certainly the technologically most important example are the inhomogeneous semiconductor devices, such as the well-known P-N diode, which constitute still an active field of research in condensed matter physics, electrical engineering and applied mathematics : see [13] for a nice, up-to-date account. Other physical examples include electrolyte solutions, certain solid electrolytes, and magnetohydrodynamics of plasmas.

Most of the past work on these systems has started from a kinetic equation description. This should be adequate at low density of charge carriers but will fail for higher concentrations. More importantly, beginning with the kinetic Boltzmann level omits consideration of fundamental issues of statistical physics for such systems. Therefore, we study here instead a fully microscopic system, a lattice-gas with Kawasaki-type stochastic dynamics, which was introduced earlier [14] as a model of fast ionic conductors. This so-called *driven lattice gas* or *DLG model* of a driven-diffusive system has several advantages for our purposes. First, its simple dynamical rules are ideal for computer realization and have allowed large-scale simulations to be performed, revealing a wealth of unexpected phenomena. For a review of these and a survey of present theoretical understanding, see the recent article of Schmittmann and Zia [15]. In addition, there are for particular simple cases of DLG models rigorous results with which to compare our theory, e.g. see [16] and our Appendix III. Such checks are important because the methods we use are formal and the physics is largely unexplored, so that corroboration by rigorous results is very reassuring when it is available. There are many deficiencies of the DLG models as a realistic portrayal of nature [13] and one may expect technical problems in extending our treatment to real systems with long-range Coulombic interactions. We believe that the difficulties already present at this stage justify their removal until a later time.

The DLG models were defined in the paper of Katz et al. (KLS) [14] and discussed systematically in Chapter 4 of [20]. Here we just recall that particles in the model hop on a simple hypercubic lattice $\mathbf{Z}^d$ in d dimensions with a hard-core exclusion condition (at most one particle per site). The particles are subjected to driving by an external electric field $\mathbf{E}$ and confined to a box $\Lambda \subset \mathbf{Z}^d$ with periodic boundary conditions. The stochastic hopping is "thermally activated" by a heat reservoir at inverse temperature $\beta = 1/k_B T$, which absorbs the Ohmic power produced by the field. More mathematically, the state space of the system is the set of all possible particle configurations $\eta = \{\eta_{\mathbf{x}} : \mathbf{x} \in \Lambda\}$, where $\eta_{\mathbf{x}}$ is the site occupation variable

$$\eta_{\mathbf{x}} = \begin{cases} 1 & \text{if } \mathbf{x} \in \Lambda \text{ is occupied,} \\ 0 & \text{if } \mathbf{x} \in \Lambda \text{ is empty.} \end{cases} \tag{1.1}$$

The dynamics is prescribed by the exchange rates $c_E(\mathbf{x}, \mathbf{y}, \eta) \geq 0$ between pairs of sites $\mathbf{x}, \mathbf{y} \in \Lambda$. Letting $\eta^{\mathbf{xy}}$ denote the configuration with occupations at sites $\mathbf{x}$ and $\mathbf{y}$ interchanged, the time evolution of a probability distribution $P_t(\eta)$ is given by the master equation

$$\begin{aligned} \frac{d}{dt} P_t(\eta) &= \frac{1}{2} \sum_{\mathbf{x},\mathbf{y} \in \Lambda} \{ c_E(\mathbf{x}, \mathbf{y}, \eta^{\mathbf{xy}}) P_t(\eta^{\mathbf{xy}}) - c_E(\mathbf{x}, \mathbf{y}, \eta) P_t(\eta) \} \\ &\equiv L^*_{E,\Lambda} P_t(\eta). \end{aligned} \tag{1.2}$$

The exchange rates are assumed, for simplicity, to be only nearest neighbor, i.e. $c_E(\mathbf{x}, \mathbf{y}, \eta) = 0$ for $|\mathbf{x} - \mathbf{y}| > 1$. It is also useful to assume that $c_E(\mathbf{x}, \mathbf{y}, \eta) > 0$ for $|\mathbf{x} - \mathbf{y}| = 1$ and $\eta_{\mathbf{x}} \neq \eta_{\mathbf{y}}$ to avoid degeneracies. Another natural condition is space homogeneity, guaranteed by invariance of the rates under translation, i.e. $c_E(\mathbf{x}+\mathbf{a}, \mathbf{y}+\mathbf{a}, \tau_{\mathbf{a}}\eta) = c_E(\mathbf{x}, \mathbf{y}, \eta)$ where $(\tau_{\mathbf{a}}\eta)_{\mathbf{x}} = \eta_{\mathbf{x}-\mathbf{a} (\mathrm{mod}\, \Lambda)}$ is the periodically shifted configuration. Finally, and most importantly, we impose *local detailed balance* with respect to a lattice-gas Hamiltonian $H(\eta)$ including the work done by the electric field $\mathbf{E}$ in the jump. This has the form

$$c_E(\mathbf{x}, \mathbf{y}, \eta) = c_E(\mathbf{x}, \mathbf{y}, \eta^{\mathbf{xy}}) e^{-\beta[H(\eta^{\mathbf{xy}}) - H(\eta)]} e^{-q\beta \mathbf{E}\cdot(\mathbf{x}-\mathbf{y})(\eta_{\mathbf{x}} - \eta_{\mathbf{y}})}, \tag{1.3}$$

where q is the particle charge and $H(\eta)$ is the Hamiltonian. For example, H could be chosen as the Ising model Hamiltonian $H(\eta) = \frac{-J}{2} \sum_{\mathbf{x},\mathbf{y} \in \Lambda, |\mathbf{x}-\mathbf{y}|=1} \eta_{\mathbf{x}} \eta_{\mathbf{y}}$. If $E = 0$, then this is the usual condition of detailed balance, which implies stationarity and reversibility of the Gibbs measure $P \propto e^{-\beta H}$ for the dynamics. For any value of field $\mathbf{E}$ and number of particles N the dynamics has a unique stationary measure $P_{E,N,\Lambda}$ in the periodic domain Λ. However, if $E \neq 0$, then the measure $P_{E,N,\Lambda}$ supports a finite mean current and is non-reversible.

In this work we shall consider the hydrodynamical description of the relaxation of an inhomogeneous but slowly-varying distribution of charge $\rho(\epsilon\mathbf{x})$ in the large domain $\epsilon^{-1}\Lambda$, for $\epsilon \to 0$. This is equivalent to the situation with a smooth distribution of charge $\rho(\mathbf{r})$ in the fixed domain Λ as the lattice spacing $\epsilon \to 0$. We shall derive a diffusion equation with drift to describe the relaxation of this charge density on a macroscopic time scale $\tau \sim \epsilon^{-1}t$, of the form

$$\partial_\tau \rho(\mathbf{r},\tau) = -\boldsymbol{\nabla} \cdot [\hat{\mathbf{j}}(\rho(\mathbf{r},\tau)) - \epsilon \mathbf{D}(\rho(\mathbf{r},\tau)) \cdot \boldsymbol{\nabla}\rho(\mathbf{r},\tau)], \tag{1.4}$$

in which the drift current is given at low fields by Ohm's law as $\hat{\mathbf{j}}(\rho) = \boldsymbol{\sigma}(\rho)\cdot\mathbf{E}$. It differs primarily from the "drift-diffusion equation" of semiconductor physics [13] in containing just one sign of charge and in lacking the terms describing recombination and generation of electron-hole pairs. Microscopic expressions for the quantities appearing in the equation will be one output of our derivation, as well as results for its solutions such as an "H-theorem," etc. It will be a chief concern of this paper to stress the statistical and dynamical conditions underlying the validity of the equation. Such properties are commonly assumed in the distribution function derivations of hydrodynamics but ordinarily only implicitly. This is particularly important for DDS since the conditions may even be violated there!

A second main problem addressed in our work is the theory of fluctuations for these same systems. It might be expected from analogy with other hydrodynamic systems that the fluctuations in the charge density will be described by adding to the conservation law a "noisy current," as

$$\partial_\tau \rho(\mathbf{r},\tau) = -\boldsymbol{\nabla} \cdot [\hat{\mathbf{j}}(\rho(\mathbf{r},\tau)) - \epsilon \mathbf{D}(\rho(\mathbf{r},\tau)) \cdot \boldsymbol{\nabla}\rho(\mathbf{r},\tau) + \epsilon^{d/2}\mathbf{j}'(\mathbf{r},\tau)]. \tag{1.5}$$

In local equilibrium systems such fluctuating hydrodynamics has been long used, with the stochastic current $\mathbf{j}'$ distributed according to a "fluctuation-dissipation relation." We shall show that even for irreversible systems such as the DDS there is a generalized fluctuation-dissipation relation. Furthermore, our derivation of the hydrodynamics is in a form (so-called *force-flux form*) which allows us to exploit the FDR to write down directly the Langevin equation for fluctuations.

Although the DLG models are our primary example, our *methods* are not at all restricted to that context and there is little difficulty in extending them to microscopic particle systems governed by classical Hamiltonian or quantum dynamics. In fact, Zubarev and his collaborators have for some years employed similar methods to derive results for physical nonequilibrium systems, including those without local equilibrium. For example, see the early work of Kalashnikov on hot electron kinetics in semiconductors [18]. At least in one simple *deterministic* model of DDS type—the Lorentz gas with applied field and "Gaussian" thermostat—the program we outline can be carried through in a mathematically rigorous way to derive Ohm's law of charge transport [19].

(1.3) Outline of the Work

The remainder of this paper is organized as follows:

In Section 2 we give our derivation of the hydrodynamical law. We start with some preliminaries on the thermodynamics of the stationary non-Gibbsian states of the dynamics. Thereafter, we derive the nonequilibrium distribution formula, which is an exact and rigorously valid expression for a time-evolved local-equilibrium measure. Using this formula as the starting point, the constitutive laws for fluxes and the hydrodynamical equations are derived by formal expansions in the separation of scales parameter ϵ. The perturbation expansions are based upon an asymptotic method à la Chapman-Enskog, exploiting the "normal form" of the nonequilibrium distribution. Mathematical rigor is lost at this stage and various approximations, e.g. neglect of memory effects, are made which lack justification and may even prove false for certain applications. However, we find the results to agree with the recent rigorous theorems of Esposito et al. [16] and Landim et al. [17] for a simple example, the *asymmetric simple exclusion process* (ASEP) [16]. We conclude this section by discussing linear transport near $E = 0$

including consequences of reversibility, the celebrated *Onsager reciprocal relations*.

In Section 3 we turn our attention to the theory of internal noise for these systems at the linear level, i.e. for small fluctuations about the steady-state. We derive the fluctuation-dissipation relation and emphasize the important fact that it is a *macroscopic relation* independent of many details of the particle system, such as microscopic reversibility. In particular, we obtain in this way a form of the *generalized Einstein relation* for DDS proposed in 1965 by Price [21], which relates the covariance of microscopic current fluctuations and the bulk diffusion tensor of charge inhomogeneities. Calculating the steady-state accurately to a Gaussian approximation, correctly incorporating the steady-state two-point spatial correlations, we can use it to write down the linear Langevin equation for small fluctuations. An alternative approach to deriving the linearized hydrodynamics can be based upon this theory—the "correlation function method" of Kadanoff and Martin [22],[23]. This method was already employed in the 1984 paper of KLS and we show that the results are consistent with those of the present approach. However, we also show that the phenomenon of long-range power-law decay of static correlations in the DLG models poses some severe difficulties for the derivation of hydrodynamical laws in these systems.

Section 4 extends some of these results to the case of large fluctuations described by a nonlinear Fokker-Planck equation. In this context there is an FDR, due in a general form to Graham [24], which allows us likewise to write down directly the Fokker-Planck equation for the distribution of the fluctuating density field. It will hold under the same assumptions which justify the hydrodynamic law itself. In the limit $\epsilon \to 0$ the probability of large fluctuations are exponentially small and a precise asymptotic formula for the decay exponent can be derived by steepest descent from a path-integral solution of the Fokker-Planck equation. This is also a procedure pioneered by Graham [25]. The result has the form of a nonlinear Onsager-Machlup action. Its physical interpretation is as an *excess dissipation function* and it leads to a variational characterization of the most probable behavior as the state of least excess dissipation.

Our concluding remarks are in Section 5 where we consider generalizations and limitations of the formalism. Some material, subsidiary to the main discussion, is collected in various Appendices in Section 6.

2 Nonequilibrium Distributions and Hydrodynamical Laws

(2.1) Nonequilibrium Thermodynamics of Driven Steady-States

As a necessary preliminary to the discussion of hydrodynamics and fluctuations, we must first address a different issue, the nonequilibrium thermodynamics of the DLG models, which involves the large-volume limit $\Lambda \to \infty$ of its stationary, translation-invariant measures. Thermodynamics plays a crucial role in the distribution function method, since generally in this method the hydrodynamic density n is spatially-modulated by the imposition of a local chemical potential λ. It is therefore required to specify a functional relationship, $\lambda = \lambda(n)$ of the chemical potential to produce a given density n. The traditional nonequilibrium thermodynamic description of electrical conducting systems postulates a local "free energy function" $f(\beta, n)$, in terms of which an "electrochemical potential" may be defined by the isothermal derivative:

$$\mu = \left(\frac{\partial f}{\partial n}\right)_\beta . \tag{2.1}$$

See Landau and Lifshitz [27], Sections 25-26. Such a description is appropriate for a system in contact with a heat bath at inverse temperature β. Alternatively, a "nonequilibrium entropy" density $s(u, n)$ may be employed which is a function of internal energy u, as in DeGroot and Mazur [28], Ch.XIII,3d, and Callen [29], Ch. 17. These functions are presumed to have also the relation to microscopic fluctuations proposed by Einstein for thermal equilibrium [9]. Namely, the probability to observe the density n in a large volume Λ should be proportional to $\sim \exp[|\Lambda| \cdot s(u, n)/k_B]$, with k_B the Boltzmann constant; see [28, 29]. This aspect of the thermodynamic functions will play an important role in our discussion of fluctuations in the DDS.

However, except for $E = 0$ or for special choices of the rates in $d = 1$, the stationary measures of the DLG models are not Gibbs measures $\propto e^{-\beta H}$ with respect to the *a priori* Hamiltonian H appearing in the definition of the dynamics. (It is an open question, discussed in Appendix I, whether they are also not Gibbs for any other Hamiltonian, or, more technically, not Gibbs probability measures for any summable potential [26].) Therefore, standard proofs of the infinite-volume limit and of the validity of thermodynamic formalism for Gibbs measures do not obviously apply. Furthermore, although the stationary measures of the DLG for fixed density are expected to be space-ergodic, they have been observed numerically to have a very slow power-law decay of correlations for all temperatures and densities when $E \neq 0$, rather than the exponential clustering exhibited in Gibbs measures away from critical points [30]. This means that much of the standard lore of equilibrium thermodynamics has no sound mathematical foundation for driven steady-states. The present section will review what little is known concerning the stationary measures of the DLG in infinite volume, and, in particular, will attempt to systematize the standard hypotheses on thermoelectric systems [27, 28, 29] in the context of the DLG models. All of our discussion will be confined to the high-temperature region of those models. As for the Ising-type lattice gas at $E = 0$, a second-order transition appears to occur at some critical T_c, below which there is a two-phase coexistence region for "liquid" and "gas" phases with different densities. In this work, we stay well away from the critical point and coexistence regions.

For finite Λ, the basic facts about stationary measures are quite straightforward to establish. Because the number of particles N is conserved, there is a stationary distribution satisfying

$$L^*_{E,\Lambda} P_{E,N,\Lambda} = 0, \tag{2.2}$$

for each $N = 0, 1, ..., |\Lambda|$. Since the jump rates are non-degenerate, these are unique, and the theory of finite-state Markov chains ensures an exponential approach to stationarity. $P_{E,N,\Lambda}$ must inherit the symmetries of the dynamics; for example, it is invariant under (periodic) translations:

$$P_{E,N,\Lambda}(\tau_{\mathbf{a}}\eta) = P_{E,N,\Lambda}(\eta). \tag{2.3}$$

The zero-field measure $P_{0,N,\Lambda}$ is the canonical equilibrium state but no explicit expression is generally available for $P_{E,N,\Lambda}$, which is defined only implicitly by Eq.(2.2). The states with

$E \neq 0$, will support a nonvanishing number current of charge carriers

$$\mathbf{j}(\mathbf{E}) = \langle \mathbf{j}_E(\mathbf{x}) \rangle_{E,N,\Lambda} \neq 0, \tag{2.4}$$

where the mth vector component of the current,

$$j_{E,m}(\mathbf{x}, \eta) = c_E(\mathbf{x}, \mathbf{x} + \hat{\mathbf{e}}_m, \eta)(\eta_{\mathbf{x}} - \eta_{\mathbf{x}+\hat{\mathbf{e}}_m}), \tag{2.5}$$

is the expected jump rate from $\mathbf{x}$ to $\mathbf{x} + \hat{\mathbf{e}}_m$ conditioned on being in the configuration η.

However, very little is rigorously known about the stationary states of the DLG dynamics for the limit $\Lambda \uparrow \mathbf{Z}^d$. Therefore, we shall proceed by making plausible hypotheses. It seems reasonable to believe that a (weak) limit exists,

$$P_{E,n} = w - \lim_{\Lambda \uparrow \mathbf{Z}^d} P_{E,N,\Lambda}, \tag{2.6}$$

when the density $n_\Lambda = N/|\Lambda|$ is selected so that

$$\lim_{\Lambda \uparrow \mathbf{Z}^d} n_\Lambda = n. \tag{2.7}$$

Existence of limits along suitable subsequences follows from a compactness argument, but one believes that the limit is, in fact, independent of the choice of subsequence. The limit point $P_{E,n}$ is a translation-invariant probability measure on the infinite-volume configuration space $\Omega = \{0,1\}^{\mathbf{Z}^d}$. It is also stationary under the infinite-volume version of the DLG dynamics, i.e. it satisfies

$$\langle L_E f \rangle_{E,n} = 0, \tag{2.8}$$

for any local function $f(\eta)$ depending upon a finite number of occupation numbers with

$$L_E f(\eta) \equiv \frac{1}{2} \sum_{\mathbf{x},\mathbf{y} \in \mathbf{Z}^d} c_E(\mathbf{x}, \mathbf{y}, \eta)[f(\eta^{\mathbf{xy}}) - f(\eta)]. \tag{2.9}$$

Furthermore, the measures $P_{E,n}$ will presumably be the unique stationary, homogeneous measures of the dynamics for each fixed density n.

The fundamental hypothesis we will need in our later development is the following: for any subset $\Lambda \subset \mathbf{Z}^d$, define the *empirical density* $n_\Lambda(\eta)$ as

$$\begin{aligned} n_\Lambda(\eta) &= \frac{N_\Lambda(\eta)}{|\Lambda|} \\ &= \frac{1}{|\Lambda|}\sum_{\mathbf{x}\in\Lambda} \eta_{\mathbf{x}}. \end{aligned} \tag{2.10}$$

Our basic assumption is then that

Hypothesis 1 *There exists, for a chosen "reference density" $n^* \in [0,1]$ (say, $n^* = \frac{1}{2}$), a concave function $s_E : [0,1] \to [-\infty, 0]$ such that, for any of the intervals $J = [a,b], (a,b], [a,b), (a,b) \subset [0,1]$,*

$$\lim_{\Lambda\uparrow\mathbf{Z}^d} \frac{1}{|\Lambda|} \log P_{E,n^*}\{n_\Lambda \in J\} = \sup_{n\in J} s_E(n). \tag{2.11}$$

Mathematically, this is an assumption of *large deviations* type [32]. Our fundamental hypothesis has not been rigorously established up until now for the stationary measures P_{E,n^*} of the DLG dynamics with $E \neq 0$. Such a result has been proved for invariant measures of non-conservative, so-called "attractive" interacting particle systems on Ω, for which an FKG property is satisfied [31]. The result means, heuristically, that

$$P_{E,n^*}\{n_\Lambda \approx n\} \sim \exp[|\Lambda| \cdot s_E(n)], \tag{2.12}$$

and the function s_E plays the same role as the entropy function in the Einstein formula for equilibrium fluctuations [9]. For this reason, we refer to $s_E(n)$ as the *nonequilibrium entropy*, or to $s = s_E(n)$ as the *fundamental equation* in the language of Callen [29]. It would be more appropriate to use the notation $s_E(n,\beta)$, but we shall not generally make the temperature-dependence explicit. The physical interpretation will be discussed further below.

It is a consequence of the Hypothesis 1 that the limit

$$\lim_{\Lambda\uparrow\mathbf{Z}^D} \frac{1}{|\Lambda|} \log\langle \exp(\lambda N_\Lambda)\rangle_{E,n^*} = p_E(\lambda), \tag{2.13}$$

exists for all real λ, and that

$$p_E(\lambda) = \sup_{n\in[0,1]} (\lambda n + s_E(n)). \tag{2.14}$$

This follows from Varadhan's theorem on asymptotics of integrals (e.g. see Section II.7 of [32]). In particular, the "pressure function" $p_E(\lambda)$ is convex in λ.Because $s_E(n)$ was assumed to be concave, it follows that it is conjugate to p_E in the sense that

$$s_E(n) = \inf_{\lambda \in \mathbf{R}} (p_E(\lambda) - \lambda n), \tag{2.15}$$

and, furthermore, the derivative

$$\lambda_E(n) = -s'_E(n) \tag{2.16}$$

exists except possibly at a countable set of points in $[0,1]$. For β small enough (or, for temperature high enough), the function s_E is expected to be strictly concave as a function of density. Therefore, the derivative $n_E(\lambda) = p'_E(\lambda)$ will exist for every $\lambda \in \mathbf{R}$.

The latter fact has an important consequence. Let $P^*_{E,\Lambda}$ be the marginal measure induced by P_{E,n^*} on $\Omega_\Lambda = \{0,1\}^\Lambda$. Then, we may define an "exponential family" of measures $P^*_{E,\Lambda,\lambda}$ on Ω_Λ by

$$P^*_{E,\Lambda,\lambda}(\eta) = \frac{1}{Z_{E,\Lambda}(\lambda)} \exp[\lambda N_\Lambda(\eta)] P^*_{E,\Lambda}(\eta), \tag{2.17}$$

with

$$Z_{E,\Lambda}(\lambda) = \langle \exp(\lambda N_\Lambda) \rangle_{E,n^*}. \tag{2.18}$$

Because of the differentiability of p_E, it can be shown that, for every $\epsilon > 0$,

$$\lim_{\Lambda \uparrow \mathbf{Z}^d} P^*_{E,\Lambda,\lambda}\{|n_\Lambda - n_E(\lambda)| < \epsilon\} = 1. \tag{2.19}$$

(For example, see Lemma VII.4.2 in [32].) In other words, the empirical density $n_\Lambda(\eta)$ distributed with respect to the measure $P^*_{E,\Lambda,\lambda}$ takes on the value nearly equal to $n_E(\lambda)$ with overwhelming probability in a large region Λ. Under our assumptions it is possible to show even a little more, namely, that the large-deviations hypothesis made for the single reference density n^* in fact implies the same property for the exponential modifications:

$$\lim_{\Lambda \uparrow \mathbf{Z}^d} \frac{1}{|\Lambda|} \log P^*_{E,\Lambda,\lambda}\{n_\Lambda \in J\} = \sup_{n \in J} s_E(n|\lambda), \tag{2.20}$$

with

$$s_E(n|\lambda) = s_E(n) + \lambda n - p_E(\lambda). \tag{2.21}$$

For example, see Theorem 3 in [31]. This implies that for any choice of reference density n, the Hypothesis 1 should hold for the measure $P_{E,n}$ also with "entropy function" $s_E(\cdot|\lambda_E(n))$.

The main point we wish to stress here is that, assuming only existence and concavity of s_E, it is possible to change the density to any desired value n by making the exponential modification with the chemical potential $\lambda = -s'_E(n)$ of the reference measure. In particular, there is no need to assume the stationary measure to be Gibbsian for this procedure to be valid. Unfortunately, while the use of the chemical potential as in Eq.(2.17) to change the density from n^* to n appears reasonable, it is not necessarily the "correct" method. One can argue that, since the number $N_\Lambda(\eta)$ of particles in a large volume Λ is conserved up to flux across the boundary, the members of the exponential family in Eq.(2.17) will be nearly stationary for a long time interval. In fact, it is to be expected that

$$P_{E,n_E(\lambda)} = w - \lim_{\Lambda\uparrow\mathbf{Z}^d} P^*_{E,\Lambda,\lambda}. \tag{2.22}$$

so that the (presumably unique) stationary measures $P_{E,n}$ with other densities $n \neq n^*$ ought to be recoverable from the infinite-volume limits of $P^*_{E,\Lambda,\lambda}$ with $\lambda = -s'_E(n)$. In the general context there is little we can say on this point, but for the specific case of the DLG models we can go a little further. It is shown in Appendix II that, under the assumption that *one* of the stationary measures of the DLG model is canonical Gibbs with a suitable Hamiltonian, then any of its ergodic, invariant measures is Gibbs for the same Hamiltonian and for a certain choice of chemical potential. The proof depends upon the extension of a result of Künsch [33] for stochastic spin-flip dynamics to our conservative models (due to A. Asselah), which states that if any translation-invariant, stationary measure of the DLG is canonical Gibbs with some Hamiltonian then all of them are canonical Gibbs with the same Hamiltonian. Of course, this does not settle the issue even for the DLG, since the invariant measures may well not be Gibbs. However, it does give some firmer dynamical grounds to our procedure. In general, our method must be judged solely on the basis of its results.

The concavity part of our hypothesis may be regarded as a kind of "stability assumption" on the stationary measures $P_{E,n}$. For example, it implies that $\chi_E^{-1} = \partial\lambda_E/\partial n = -d^2 s_E/dn^2 > 0$. Therefore, an increase in the "potential" λ is required to raise the density n. Also, if the fluctuation formula analogous to Eq.(2.12) is considered for $P_{E,n}$—which follows from the analogue of Eq.(2.20)—then in a quadratic approximation

$$P_{E,n}\{n_\Lambda \approx n'\} \sim \exp[-|\Lambda| \cdot \chi_E(n)(n'-n)^2/2], \tag{2.23}$$

and the probabilities of spontaneous fluctuations of the density away from n are damped out. Concavity of the entropy also implies heuristically a stability property in the coupling of systems in two large regions Λ_1, Λ_2 both distributed according to the measure $P_{E,n}$. The two systems are imagined to be coupled at a common boundary in an arbitrary way permitting free exchange of particles. If the volume-fractions $\lambda_1 = |\Lambda_1|/|\Lambda|, \lambda_2 = |\Lambda_2|/|\Lambda|$ are fixed and it is assumed that correlations of the subsystems are an ignorable boundary effect in the limit $|\Lambda| \to \infty$, then

$$\begin{aligned} P(n_\Lambda \approx n) &\sim \sup_{\{n_1,n_2:\lambda_1 n_1+\lambda_2 n_2=n\}} \{P(n_{\Lambda_1}\approx n_1)P(n_{\Lambda_2}\approx n_2)\} \\ &\sim \exp\left\{|\Lambda| \sup_{\{n_1,n_2:\lambda_1 n_1+\lambda_2 n_2=n\}} [\lambda_1 s_E(n_1)+\lambda_2 s_E(n_2)]\right\} \\ &\sim \exp[|\Lambda| s_E(n)], \end{aligned} \tag{2.24}$$

where the assumption of concavity was used in the last line. In other words, the distribution of density fluctuations is unchanged for the compound system and is something like an asymptotically "stable law" in the sense of limit theorems of probability. We believe the concavity is likely to hold for the systems we consider, or in general for steady-states with such stability.

To conclude this section, let us make a few explanatory remarks concerning the physical interpretation of the various quantities introduced above. Since the lattice-gas dynamics is at a fixed temperature, the appropriate thermodynamic potential is actually the *free-energy function* $f_E(n,\beta)$, with units of energy per volume, related to our "entropy" as

$$s_E(n,\beta) = -\beta f_E(n,\beta). \tag{2.25}$$

For notational convenience, however, we have preferred to work with the "entropy" s_E with units of inverse volume. Our parameter λ is likewise related to the usual electrochemical potential μ as $\lambda = \beta\mu$. In Section 4.3 of this paper we shall show that f has a simple operational significance as the minimum energy dissipated by external fields required to change the density from its reference value n^* to a new value. We have chosen to work with number density n, but we could just as well have used the charge density $\rho = q \cdot n$. In that case total charge Q_Λ would replace particle number N_Λ in the preceding arguments. The quantity $\varphi = \mu/q$ can then be interpreted as the "electric potential" of the system. If the need arises, we may denote the "number current" previously introduced as $\mathbf{j}^N$, and the "electric current" $q \cdot \mathbf{j}^N$ as $\mathbf{j}^Q$.

(2.2) The Nonequilibrium Distribution Formula

The problem we wish to study is the relaxation of an inhomogeneous density distribution $n_0(\mathbf{r})$ imposed upon the homogeneous measure P_{E,n^*}. A basic condition necessary for validity of a hydrodynamical description is the *separation of scales* between length- and time-scales associated to the microscopic conservative dynamics and the length- and time-scales over which the macroscopic density variations occur. We always denote by ϵ the small ratio of micro/macro length-scales, which is here just $\epsilon =$(lattice-spacing)/(gradient-length). Thus, we wish to consider states in which the density variation in lattice units is $n_0(\epsilon\mathbf{x})$. From the discussion in the previous section, one guesses that an appropriate way to modify the reference measure $P_{E,N^*,\Lambda}$ is by adding a local chemical potential term. More precisely, we define a *(canonical) local stationary measure* in the scaled domain $\Lambda_\epsilon = (\epsilon^{-1}\Lambda) \cap \mathbf{Z}^d$ as

$$P_\epsilon^{ls}(\eta) = \frac{1}{Z_\epsilon^{ls}} \exp\left(\sum_{\mathbf{x}\in\Lambda_\epsilon} \lambda_0(\epsilon\mathbf{x})\eta_{\mathbf{x}}\right) P_{E,N_\epsilon^*,\Lambda_\epsilon}(\eta), \tag{2.26}$$

with

$$Z_\epsilon^{ls} = \sum_{\eta\in\Omega_{\Lambda_\epsilon}} \exp\left(\sum_{\mathbf{x}\in\Lambda_\epsilon} \lambda_0(\epsilon\mathbf{x})\eta_{\mathbf{x}}\right) P_{E,N_\epsilon^*,\Lambda_\epsilon}(\eta). \tag{2.27}$$

Here $\lambda_0(\mathbf{r})$ is the smooth chemical potential field given by the "local prescription"

$$\lambda_0(\mathbf{r}) = -s'(n_0(\mathbf{r})). \tag{2.28}$$

(We drop E-subscripts in this section when no confusion will result.) Also, $N_\epsilon^* = [\![n^* \cdot \Lambda_\epsilon]\!]$, where $[\![\cdot]\!]$ denotes the integer part. For simplicity in the following expressions, we shall often abbreviate $P_{E,N_\epsilon^*,\Lambda_\epsilon} = P_\epsilon^*$. We consider always periodic b.c. on Λ.

The "nonequilibrium distribution formula" is an exact expression for the time-evolution of the initial measure P_ϵ^{ls} under the DLG dynamics. It can be written in the "weak" form for averages, as:

$$\begin{aligned}
\langle e^{\epsilon^{-1}\tau L} f\rangle_\epsilon^{ls} &= \langle f\rangle_\epsilon^{ls,\tau} \\
&-\epsilon\int_0^{\epsilon^{-1}\tau} ds \sum_{\mathbf{x}\in\Lambda_\epsilon} \Big(\partial_m^+\lambda(\epsilon\mathbf{x},\tau-\epsilon s)\langle(\Delta_{\mathbf{x},\mathbf{x}+\hat{\mathbf{e}}_m} e^{sL} f) I_{\epsilon,m}^{\tau-\epsilon s}(\mathbf{x})\rangle_\epsilon^{ls,\tau-\epsilon s} \\
&\qquad + \partial_\tau\lambda(\epsilon\mathbf{x},\tau-\epsilon s)\langle(e^{sL} f)(\eta_{\mathbf{x}} - \langle\eta_{\mathbf{x}}\rangle_\epsilon^{ls,\tau-\epsilon s})\rangle_\epsilon^{ls,\tau-\epsilon s}\Big). \qquad (2.29)
\end{aligned}$$

We must explain our various notations. The parameter τ is a "macroscopic time," associated to a microscopic time $\epsilon^{-1}\tau$ which goes to infinity as $\epsilon \to 0$.. The formula itself is valid without any need for ϵ to be small (e.g. it is true with $\epsilon = 1$), but we wish to consider the evolution of the density distribution slowly varying on the spatial scale $\sim \epsilon^{-1}$. A change of order 1 in the density occurs in a time $\sim \epsilon^{-1}$ through the transport of particles over distances $\sim \epsilon^{-1}$ by the local drift velocity. The formula depends upon a space-time chemical potential field $\lambda(\mathbf{r},\tau)$ in the macroscopic variables which—we emphasize—is at this stage *arbitrary* except for the initial condition $\lambda(\mathbf{r},0) = \lambda_0(\mathbf{r})$ and some reasonable smoothness. The expectation $\langle\cdot\rangle_\epsilon^{ls,\tau}$ is with respect to the local stationary distribution for the profile $\lambda(\cdot,\tau)$. For any choice of a chemical potential profile, $\mathbf{I}_\epsilon$ is a "modified current" defined by

$$\begin{aligned}
I_{\epsilon,m}(\mathbf{x},\eta) &\equiv c(\mathbf{x},\mathbf{x}+\hat{\mathbf{e}}_m,\eta)\left[\frac{\exp\left((\lambda(\epsilon(\mathbf{x}+\hat{\mathbf{e}}_m)) - \lambda(\epsilon\mathbf{x}))(\eta_{\mathbf{x}} - \eta_{\mathbf{x}+\hat{\mathbf{e}}_m})\right) - 1}{\lambda(\epsilon(\mathbf{x}+\hat{\mathbf{e}}_m)) - \lambda(\epsilon\mathbf{x})}\right] \\
&= j_m(\mathbf{x},\eta) + O(\epsilon). \qquad (2.30)
\end{aligned}$$

The symbol ∂_m^+ denotes the operation of forward difference, $(\partial_m^+\lambda)(\mathbf{r}) \equiv \frac{\lambda(\mathbf{r}+\epsilon\hat{\mathbf{e}}_m)-\lambda(\mathbf{r})}{\epsilon}$, and $\Delta_{\mathbf{xy}}$ is the "exchange operator," $(\Delta_{\mathbf{xy}})f(\eta) \equiv f(\eta^{\mathbf{xy}})$. We use the summation convention for the repeated index m (except in Eq.(2.30)) and also below for other repeated Latin indices. The basic advantage of the "nonequilibrium distribution formula" is that it separates the true time-evolved distribution into a leading part of order ~ 1, which is given by a canonical local stationary distribution with a reference profile $n(\cdot,\tau)$, and a correction term of order $\sim \epsilon$ involving microscopic space-time correlations.

The proof of Eq.(2.29) is straightforward, based upon a simple trick using the fundamental theorem of calculus. In fact, it is easy to see that

$$\begin{aligned}\langle e^{\epsilon^{-1}\tau L}f\rangle_\epsilon^{ls} &= \frac{1}{Z_\epsilon^{ls,\tau}}\sum_\eta P_\epsilon^*(\eta)\exp\left(\sum_{\mathbf{x}\in\Lambda_\epsilon}\lambda(\epsilon\mathbf{x},\tau)\eta_{\mathbf{x}}\right)f(\eta)\\ &\quad+\sum_\eta\int_0^{\epsilon^{-1}\tau}ds\,\frac{d}{ds}\left[\frac{\exp\left(\sum_{\mathbf{x}\in\Lambda_\epsilon}\lambda(\epsilon\mathbf{x},\tau-\epsilon s)\eta_{\mathbf{x}}\right)}{Z_\epsilon^{ls,\tau-\epsilon s}}(e^{sL}f)(\eta)\right]P_\epsilon^*(\eta)\end{aligned}\tag{2.31}$$

where $Z_\epsilon^{ls,\tau}$ is as defined in Eq.(2.27) but with $\lambda_0(\epsilon\mathbf{x})$ replaced by $\lambda(\epsilon\mathbf{x},\tau)$. Carrying out the differentiation inside the integral then gives

$$\begin{aligned}\langle e^{\epsilon^{-1}\tau L}f\rangle_\epsilon^{ls} &= \langle f\rangle_\epsilon^{ls,\tau}\\ &\quad+\sum_\eta\int_0^{\epsilon^{-1}\tau}ds\,\frac{1}{Z_\epsilon^{ls,\tau-\epsilon s}}\Big[(Le^{sL}f)(\eta)\\ &\qquad-\epsilon\sum_{\mathbf{x}\in\Lambda_\epsilon}\partial_\tau\lambda(\epsilon\mathbf{x},\tau-\epsilon s)(\eta_{\mathbf{x}}-\langle\eta_{\mathbf{x}}\rangle_\epsilon^{ls,\tau-\epsilon s})\times(e^{sL}f)(\eta)\Big]\\ &\qquad\times\exp\left(\sum_{\mathbf{x}\in\Lambda_\epsilon}\lambda(\epsilon\mathbf{x},\tau-\epsilon s)\eta_{\mathbf{x}}\right)P_\epsilon^*(\eta).\end{aligned}\tag{2.32}$$

In the first term in the square brackets one obtains the factor $(L^*P_\epsilon^{ls,\tau-\epsilon s})(\eta)$ by taking the adjoint L^* of the operator L with respect to counting measure, which is given by Eq.(1.2). For any local stationary distribution this factor is easily calculated to be

$$(L^*P_\epsilon^{ls})(\eta)=-\epsilon\sum_{\mathbf{x}\in\Lambda_\epsilon}\partial_m^+\lambda(\epsilon\mathbf{x})\Delta_{\mathbf{x},\mathbf{x}+\hat{\mathbf{e}}_m}\left[I_{\epsilon,m}(\mathbf{x},\eta)P_\epsilon^{ls}(\eta)\right],\tag{2.33}$$

when the stationarity condition $(L^*P_\epsilon^*)(\eta)=0$ is used. If this result is now substituted into Eq.(2.32), the originally claimed formula is obtained.

Another useful form of the expression can be established by noting that, for any local stationary distribution,

$$\sum_{\mathbf{x}\in\Lambda_\epsilon}\partial_m^+\lambda(\epsilon\mathbf{x})\langle I_{\epsilon,m}(\mathbf{x})\rangle_\epsilon^{ls}=0\tag{2.34}$$

as an exact identity. This will also turn out to be very important in the discussion of hydrodynamics in the next subsection. It is easily proved by summing equation Eq.(2.33) over $\eta \in \Omega_{\Lambda_\epsilon}$. As a consequence of this identity, the entire distribution formula can be expressed in terms of truncated expectations $\langle fg\rangle^\top = \langle fg\rangle - \langle f\rangle\langle g\rangle$, as

$$\begin{aligned}\langle e^{\epsilon^{-1}\tau L} f\rangle_\epsilon^{ls} &= \langle f\rangle_\epsilon^{ls,\tau} \\ &-\epsilon \int_0^{\epsilon^{-1}\tau} ds \sum_{\mathbf{x}\in\Lambda_\epsilon} \Big(\partial_m^+\lambda(\epsilon\mathbf{x},\tau-\epsilon s)\langle(\Delta_{\mathbf{x},\mathbf{x}+\hat{\mathbf{e}}_m} e^{sL} f) I_{\epsilon,m}^{\tau-\epsilon s}(\mathbf{x})\rangle_\epsilon^{ls,\tau-\epsilon s,\top} \\ &\qquad + \partial_\tau\lambda(\epsilon\mathbf{x},\tau-\epsilon s)\langle(e^{sL}f)\eta_{\mathbf{x}}\rangle_\epsilon^{ls,\tau-\epsilon s,\top}\Big) + o(\epsilon). \end{aligned} \tag{2.35}$$

The $o(\epsilon)$ error term arises from the fact that $\langle \Delta_{\mathbf{x},\mathbf{x}+\hat{\mathbf{e}}_m} e^{sL} f\rangle_\epsilon^{ls,\tau-\epsilon s}$ becomes independent of $\mathbf{x}$ only in the limit $\epsilon \to 0$. This new form is very crucial in taking that limit.

Until this last point we have not made any simplifications based upon small ϵ and the results are formally exact. We have also not made any particular choice of reference profile $\lambda(\cdot,\tau)$, except for the required initial condition. However, we wish now to make an evaluation of the formula which should be asymptotically exact for $\epsilon \to 0$. In particular, we wish to approximate the expectation $\langle e^{\epsilon^{-1}\tau L}\tau_{[\![\epsilon^{-1}\mathbf{r}]\!]} f\rangle_\epsilon^{ls}$, when f is a local function supported in a neighborhood of the origin shifted to the lattice point $[\![\epsilon^{-1}\mathbf{r}]\!]$. The first term in the desired approximation ought to be the "local steady-state" part, in which the average is with respect to the homogeneous stationary measure at density $n(\mathbf{r},\tau)$ and the second term is a correction. Of course, to have a chance of getting a small correction of order $\sim \epsilon$ it is necessary to choose the correct hydrodynamic profile in the leading term of order ~ 1, which should be determined by the solution of the "Euler-level" hydrodynamic equation. The resulting formula, which characterizes the behavior of the time-evolved measure in the neighborhood of the space-time point $(\mathbf{r},\tau)$ is:

$$\begin{aligned}\langle e^{\epsilon^{-1}\tau L}\tau_{[\![\epsilon^{-1}\mathbf{r}]\!]} f\rangle_\epsilon^{ls} &= \langle f\rangle_{n(\mathbf{r},\tau)} \\ &+\epsilon\partial_m\lambda(\mathbf{r},\tau)\sum_{\mathbf{x}} x_m\langle\eta_{\mathbf{x}} f\rangle_{n(\mathbf{r},\tau)}^\top \\ &-\epsilon\partial_m\lambda(\mathbf{r},\tau)\int_0^\infty dt \sum_{\mathbf{x}\in\mathbf{Z}^d}\Big[\langle(\Delta_{\mathbf{x},\mathbf{x}+\hat{\mathbf{e}}_m} e^{tL} f) j_m(\mathbf{x})\rangle_{n(\mathbf{r},\tau)}^\top \\ &\qquad - \chi((n(\mathbf{r},\tau))\hat{\jmath}_m'(n(\mathbf{r},\tau))\,\hat{f}'(n(\mathbf{r},\tau))\Big] + o(\epsilon). \end{aligned} \tag{2.36}$$

We have introduced a new notation

$$\hat{f}(n) \equiv \langle f \rangle_n, \tag{2.37}$$

where the expectation is with respect to the infinite-volume stationary measure at density n and $\hat{f}'$ denotes derivative with respect to n. (We also use the same notation $\hat{f}(\lambda)$ for $\hat{f}(n(\lambda))$, hopefully without any confusion.) In Eq.(2.36) we have now made a special selection of the reference profile, as the solution of the initial-value problem for the first-order hyperbolic equation

$$\partial_\tau \lambda(\mathbf{r},\tau) = -\hat{\jmath}'(n(\lambda(\mathbf{r},\tau))) \cdot \nabla\lambda(\mathbf{r},\tau). \tag{2.38}$$

As we discuss in the following subsection, this is indeed the "Euler" hydrodynamic equation of the system, correct to describe the evolution of the density for microscopic times $\sim \epsilon^{-1}$. We shall see below that it is this choice of the profile which produces the term proportional to $\hat{\jmath}'(n)$ in Eq.(2.36). A formal argument, given in Appendix 3 of [14], shows that this is precisely the subtraction necessary for the convergence of the time-integral.

The derivation of the asymptotic formula Eq.(2.36) is no longer rigorous, but is based upon plausible formal reasoning. The first two terms come from an evaluation of the local stationary expectation in Eq.(2.35). In fact, if one approximates

$$\lambda(\epsilon\mathbf{x},\tau) = \lambda(\mathbf{r},\tau) + \epsilon\boldsymbol{\nabla}\lambda(\mathbf{r},\tau)\cdot(\mathbf{x}-\epsilon^{-1}\mathbf{r}) + O(\epsilon^2) \tag{2.39}$$

for $\epsilon\mathbf{x} \approx \mathbf{r}$ and expands out the exponent in the canonical form, one obtains

$$\langle \tau_{[\![\epsilon^{-1}\mathbf{r}]\!]} f \rangle_\epsilon^{ls,\tau} = \langle f \rangle_{n(\mathbf{r},\tau)} + \epsilon\partial_m\lambda(\mathbf{r},\tau)\sum_{\mathbf{x}} x_m \langle \eta_{\mathbf{x}} f \rangle_{n(\mathbf{r},\tau)}^{\top} + o(\epsilon). \tag{2.40}$$

To evaluate the time-integral term, one first substitutes Eqs.(2.30) and (2.38), to obtain for it

$$\begin{aligned}\epsilon\int_0^{\epsilon^{-1}\tau} ds \sum_{\mathbf{x}\in\Lambda_\epsilon} \partial_m\lambda(\epsilon\mathbf{x},\tau-\epsilon s)\Big[&\langle \Delta_{\mathbf{x},\mathbf{x}+\hat{\mathbf{e}}_m} e^{sL}\tau_{[\![\epsilon^{-1}\mathbf{r}]\!]} f \cdot j_m(\mathbf{x})\rangle_\epsilon^{ls,\tau-\epsilon s,\top} \\ &- \hat{\jmath}'_m(n(\epsilon\mathbf{x},\tau-\epsilon s))\langle e^{sL}\tau_{[\![\epsilon^{-1}\mathbf{r}]\!]} f\cdot\eta_{\mathbf{x}}\rangle_\epsilon^{ls,\tau-\epsilon s,\top}\Big] + o(\epsilon).\end{aligned} \tag{2.41}$$

If the truncated correlations decay rapidly enough on the microscopic scale, then the time-integral and space-sum in the above expression get their main contributions from the regions

$s \approx 0$ and $\epsilon\mathbf{x} \approx \mathbf{r}$. In that case, the reference profile may be evaluated everywhere to leading order by its value at space-time point $(\mathbf{r},\tau)$ and the integral and sum may be extended to infinity. In this way, the previous expression is estimated as

$$\epsilon\partial_m\lambda(\mathbf{r},\tau)\int_0^\infty ds\left[\sum_{\mathbf{x}\in\mathbf{Z}^d}\langle\Delta_{\mathbf{x},\mathbf{x}+\hat{\mathbf{e}}_m}e^{sL}f\cdot j_m(\mathbf{x})\rangle^\top_{n(\mathbf{r},\tau)}\right.$$
$$\left.-\,\hat{\jmath}'_m(n(\mathbf{r},\tau))\sum_{\mathbf{x}\in\mathbf{Z}^d}\langle e^{sL}f\cdot\eta_{\mathbf{x}}\rangle^\top_{n(\mathbf{r},\tau)}\right]+o(\epsilon). \tag{2.42}$$

However, appealing to the fact that the state $\langle\cdot\rangle_n$ may be obtained by adding the chemical potential term $\exp[\lambda\sum_{\mathbf{x}}\eta_{\mathbf{x}}]$ to the reference measure in finite volume, one finds by a formal exchange of limits that

$$\begin{aligned}\hat{f}'(n) &= \frac{1}{\chi(n)}\hat{f}'(\lambda)\\ &= \frac{1}{\chi(n)}\sum_{\mathbf{x}}\langle f\eta_{\mathbf{x}}\rangle^\top_n.\end{aligned} \tag{2.43}$$

Substituting this expression into the final term of (2.42), one checks that the claimed asymptotic formula Eq.(2.36) is the result. Although no careful justification was provided for the various steps in the previous argument, it is clear that the main requirement is a rapid decay of the truncated space-time correlation functions. This certainly restricts us to dimensions $d > 2$, since there are non-integrable long-time tails in dimensions $d = 1, 2$ (see Appendix III.) If the assumption is violated—for any reason —then a similar evaluation could still be made, but with a result nonlocal in the space-time profile $\lambda(\mathbf{r}',\tau')$ and involving an integral kernel defined in terms of truncated correlations. The assumption of fast decay has allowed us to "Markovianize" the expression.

To conclude this subsection, we will observe that the same types of formulae, like both the exact expression Eq.(2.29) and the approximate asymptotic expression Eq.(2.36), can be derived as well for classical or quantum microscopic dynamics. The exact expression can also be written as a formula for the density (or, Radon-Nikodým derivative) of the true time-evolved measure with respect to a reference local stationary measure. In the case of classical or quantum dynamics there is another version of this formula which can be proved by exploiting the Liouville theorem in which the space-time integral appears in an exponent [1, 2, 3, 23]. This density of exponential type is closer to the "Girsanov formula" for change of measure in stochastic analysis. In all cases, the exact expression for the time-evolved measure involves an integral over the past history of the system.

(2.3) Microscopic Derivation of Hydrodynamics

Hydrodynamic behavior in the systems we consider is due to the presence of microscopic conservation. In the DLG model there is *local conservation* of particle number, expressed as

$$\partial_t N(\mathbf{x},t) + \nabla_m^- J_m(\mathbf{x},t) = 0. \tag{2.44}$$

Here $N(\mathbf{x},t)$ is the occupation number at point $\mathbf{x}$ at time t in a particular realization of the dynamics, $J_m(\mathbf{x},t)$ is the instantaneous flux of particles from site $\mathbf{x}$ to site $\mathbf{x}+\hat{\mathbf{e}}_m$ at time t for the same stochastic trajectory, and $(\nabla_m^- b)(\mathbf{x}) = b(\mathbf{x}) - b(\mathbf{x}-\hat{\mathbf{e}}_m)$ is the backward difference operator. The expectation of this relation at $t=0$, conditional on being in configuration η, is

$$L\eta_{\mathbf{x}} + \nabla_m^- j_m(\mathbf{x},\eta) = 0 \tag{2.45}$$

(which is quite easy to verify also by direct calculation). These two relations are the expression of the fact that particles are neither created nor destroyed in the DLG dynamics but simply hop to neighboring sites.

The derivation of the hydrodynamic law, at least for ensemble averaged densities, is considerably simplified by the existence of the microscopic conservation laws. In fact, let us define the mean number-density for the time-evolved measure, as

$$\bar{n}_\epsilon(\mathbf{r},\tau) \equiv \langle e^{\epsilon^{-1}\tau L}\eta_{[\![\epsilon^{-1}\mathbf{r}]\!]}\rangle_\epsilon^{ls} \tag{2.46}$$

and the mean current density as

$$\bar{\mathrm{J}}_\epsilon(\mathbf{r},\tau) \equiv \langle e^{\epsilon^{-1}\tau L}\mathbf{j}([\![\epsilon^{-1}\mathbf{r}]\!])\rangle_\epsilon^{ls}. \tag{2.47}$$

Then, using the conservation relation Eq.(2.45), it follows that

$$\partial_\tau \bar{n}_\epsilon(\mathbf{r},\tau) + \partial_m^- \bar{\mathrm{J}}_{\epsilon,m}(\mathbf{r},\tau) = 0. \tag{2.48}$$

Therefore, the "balance equation" for the mean density follows straightforwardly. At this point we have not even made any use of the fact that the averages are with respect to a "local stationary" measure, and the above equation holds quite generally.

However, we have seen also in the preceding subsection that the time-evolved measure can be separated into two contributions, the leading-order term having the canonical local stationary form and a correction term of order $\sim \epsilon$. The local stationary part must, for consistency, have a reference profile which is the same as $\bar{n}_\epsilon(\cdot, \tau)$, up to possible deviations of order $\sim \epsilon$. This means that an equation for the mean density can be derived by evaluating the mean current correctly up to that same order and substituting into the balance Eq.(2.48). The required result for the mean current is

$$\bar{\jmath}_{\epsilon,m}(\mathbf{r}, \tau) = \hat{\jmath}_m(\bar{n}_\epsilon(\mathbf{r}, \tau)) + O(\epsilon). \tag{2.49}$$

This "constitutive law" closes the balance equation in terms of the density at that order, yielding the following first-order hyperbolic equation

$$\partial_\tau \bar{n}(\mathbf{r}, \tau) + \partial_m \hat{\jmath}_m(\bar{n}(\mathbf{r}, \tau)) = 0. \tag{2.50}$$

It is easy to see that this Eq.(2.50) is the same as Eq.(2.38), and is the "Euler hydrodynamic equation" for the DLG model. It is expected to give a correct description of the evolution in the initial density distribution over microscopic times of order ϵ^{-1} up to corrections $\sim \epsilon$:

$$\bar{n}_\epsilon(\mathbf{r}, \tau) = n(\mathbf{r}, \tau) + O(\epsilon). \tag{2.51}$$

The equation does not involve ϵ, so that its solution is likewise independent of ϵ and has been denoted simply as $n(\mathbf{r}, \tau)$.

An important fact concerning the Euler hydrodynamics can be deduced from the exact relation Eq.(2.34) in the previous section. Consider that equation for any chosen reference profile $\lambda(\cdot)$. Recalling that $I_{\epsilon,m}(\mathbf{x}, \eta) = j_m(\mathbf{x}, \eta) + O(\epsilon)$, substituting into Eq.(2.34), multiplying by ϵ^d and taking the limit $\epsilon \to 0$ gives

$$\int_\Lambda d\mathbf{r}\ \partial_m \lambda(\mathbf{r}) \hat{\jmath}_m(n(\mathbf{r})) = 0, \tag{2.52}$$

whenever the reference profile λ has at least one continuous derivative. This result can be given a direct physical interpretation by defining the following "local entropy functional"

$$S(n) \equiv \int_\Lambda d\mathbf{r}\ s(n(\mathbf{r})), \tag{2.53}$$

in terms of density profiles $n(\cdot)$ over Λ. Then, noting that

$$\lambda(\mathbf{r}) = -\frac{\delta S}{\delta n(\mathbf{r})}, \tag{2.54}$$

a simple integration by parts gives

$$\int_\Lambda d\mathbf{r}\ \frac{\delta S}{\delta n(\mathbf{r})}\partial_m \hat{\jmath}_m(n(\mathbf{r})) = 0. \tag{2.55}$$

Therefore, this relation just states that $dS/dt = 0$ instantaneously for the Euler evolution, which is therefore *conservative* of the entropy S. [2] However, it should be noted that this result assumed the differentiability of the chemical potential profile $\lambda(\cdot)$ (or of the density profile $n(\cdot)$). Even if this were true for the initial density $n_0(\cdot)$, it need not be true for the solution $n(\cdot,\tau)$ of the Euler equations. Since the function $\hat{\jmath}(n)$ is nonlinear in general, the characteristic velocities $\mathbf{c}(n) = \hat{\jmath}'(n)$ depend upon the density and shock-type singularities may appear after a finite time if characteristics intersect. Thereafter, the equations would have to be interpreted in a suitable "weak" sense and the entropy conservation law for the Euler dynamics would break down.

[2] For the situation we are considering here with a single conserved quantity, this is actually a trivial result. In fact, for *any* differentiable function $h(n)$, one can define the associated flux $\mathbf{j}^h(n) = \int_{n^*}^{n} d\bar{n}\, h'(\bar{n})\hat{\jmath}'(\bar{n})$, so that a smooth solution of the Euler equation satisfies $\partial_\tau h(n(\mathbf{r},\tau)) + \partial_m j^h_m(n(\mathbf{r},\tau)) = 0$. Therefore any "local functional" $H(n) = \int_\Lambda d\mathbf{r}\ h(n(\mathbf{r}))$ is conserved. However, our result holds also with an arbitrary number of conserved densities $\boldsymbol{\rho} = (\rho_1, ..., \rho_r)$ and then the existence of a conserved, concave entropy function $s(\boldsymbol{\rho})$ is a nontrivial and important fact. Indeed, it is not hard to show that there is an associated current function $\mathbf{j}^s(\boldsymbol{\rho})$ so that a local conservation holds as above, i.e. $(s, \mathbf{j}^s)$ are a convex Lax entropy pair for the drift-diffusion equation Eq.(2.61). Existence of such an entropy pair is generally true of the PDE's which arise in statistical mechanics, as discussed elsewhere [36].

The Euler equations, while adequate in certain regimes, will not be sufficient for long times. For example, they cannot describe the relaxation of the initial density $n_0(\mathbf{r})$ to a final homogeneous state $n_\infty(\mathbf{r})$ of constant density $\bar{n}_0 = \frac{1}{|\Lambda|}\int_\Lambda d\mathbf{r}\; n_0(\mathbf{r})$: if the solution stays smooth, then S will keep its initial value $S(n_0)$ forever and never even begin to approach the expected final value $S(n_\infty) = |\Lambda| \cdot s(\bar{n}_0) > S(n_0)$. (The inequality follows by strict concavity of s.) In fact, production of entropy and the relaxation to the final stationary state will occur on a longer time scale $\sim \epsilon^{-2}$ in microscopic units due to an additional *diffusive* contribution to the current of order $\sim \epsilon$. To derive the corresponding "Navier-Stokes hydrodynamic equation," therefore, the mean current in Eq.(2.48) must be evaluated to that order.

The evaluation may be accomplished using the general formula Eq.(2.36) for the order $\sim \epsilon$ correction to the local stationary state. Observe that this formula is valid if the correct mean density $\bar{n}_\epsilon$ is used for the reference density in the local stationary state rather than the solution of the Euler equation, because $\partial_\tau \lambda(\bar{n}_\epsilon(\mathbf{r},\tau)) = -\hat{\mathrm{j}}'(\bar{n}_\epsilon(\mathbf{r},\tau))\cdot\nabla\lambda(\bar{n}_\epsilon(\mathbf{r},\tau)) + O(\epsilon)$ and the changes to the formula Eq.(2.36) are $O(\epsilon^2)$. Therefore, it is easy to calculate the "constitutive law" at the next to leading order, as

$$\bar{\mathrm{j}}_{\epsilon,m}(\mathbf{r},\tau) = \hat{\mathrm{j}}_m(\bar{n}_\epsilon(\mathbf{r},\tau)) - \epsilon L_{ml}(\bar{n}_\epsilon(\mathbf{r},\tau))\partial_l\lambda(\bar{n}_\epsilon(\mathbf{r},\tau)) + o(\epsilon), \tag{2.56}$$

with

$$L_{ml}(n) = -\sum_{\mathbf{x}} x_l \langle \eta_{\mathbf{x}} j_m\rangle_n^\top + \int_0^\infty dt \left[\sum_{\mathbf{x}\in\mathbf{Z}^d} \langle \Delta_{\mathbf{x},\mathbf{x}+\hat{\mathbf{e}}_l} e^{tL} j_m(\mathbf{0}) \cdot j_l(\mathbf{x})\rangle_n^\top - \chi(n)\hat{\mathrm{j}}'_m(n)\hat{\mathrm{j}}'_l(n)\right], \tag{2.57}$$

or, equivalently,

$$\bar{\mathrm{j}}_{\epsilon,m}(\mathbf{r},\tau) = \hat{\mathrm{j}}_m(\bar{n}_\epsilon(\mathbf{r},\tau)) - \epsilon D_{ml}(\bar{n}_\epsilon(\mathbf{r},\tau))\partial_l\bar{n}_\epsilon(\mathbf{r},\tau) + o(\epsilon), \tag{2.58}$$

with

$$L_{ml}(n) = D_{ml}(n)\chi(n). \tag{2.59}$$

Since $\mathbf{D}$ is the coefficient of the contribution to the particle flux linearly proportional to the density gradient, it may be identified as the *(bulk) diffusion matrix*, whereas $\mathbf{L}$ is the so-called

Onsager coefficient. The first version of the constitutive law is said to be in the *(Onsager) force-flux form*, because $\mathbf{X} = -\nabla\lambda$ is the "thermodynamic force" conjugate to the "flux" $\mathbf{j}$. It is interesting to note that if we discuss charge density ρ rather than number density n, then the constitutive law for the electric current is obtained as

$$\bar{\mathrm{j}}^{Q}_{\epsilon,m}(\mathbf{r},\tau) = \hat{\mathrm{j}}^{Q}_{m}(\bar{\rho}_\epsilon(\mathbf{r},\tau)) - \epsilon\Sigma_{ml}(\bar{\rho}_\epsilon(\mathbf{r},\tau))\partial_l\bar{\varphi}_\epsilon(\mathbf{r},\tau) + o(\epsilon), \tag{2.60}$$

with $\mathbf{\Sigma} = \beta q^2\mathbf{L}$. The latter is a kind of "conductivity matrix" for the electric field $\mathbf{E} = -\nabla\bar{\varphi}$ due to the inhomogeneous "electrochemical potential." (See Section 2.5 for further discussion.)

Employing the order $\sim \epsilon$ constitutive law in the local balance Eq.(2.48) then yields the "Navier-Stokes equation," in the form

$$\partial_\tau n_\epsilon(\mathbf{r},\tau) = -c_m(n_\epsilon(\mathbf{r},\tau))\partial_m n_\epsilon(\mathbf{r},\tau) + \epsilon\partial_m\left[D_{ml}(n_\epsilon(\mathbf{r},\tau))\cdot\partial_l n_\epsilon(\mathbf{r},\tau)\right]. \tag{2.61}$$

This is precisely a *driven diffusion equation*, where $\mathbf{c}$ represents the drift and $\mathbf{D}$ the diffusion. [3] It is also easy to write down the equation in "Onsager form" using the Onsager matrix rather than the diffusion matrix. This equation contains the small parameter ϵ and its solution, which depends now upon ϵ, may be denoted as $n_\epsilon(\mathbf{r},\tau)$. (Physicists may not be accustomed to writing the small parameter explicitly in this equation, but elementary kinetic theory estimates show that transport coefficients like D are proportional to the mean-free-path in physical systems and, therefore, $\sim \epsilon$ when lengths are measured in macroscopic units.) It is expected that the

[3] A peculiarity of the single-component case is that only the *symmetric part* of the diffusion matrix $\mathbf{D}$ enters into the hydrodynamic equations. Indeed, the diffusive term can be written as, $D_{ml}(n)\partial_m\partial_l n + D'_{ml}(n)(\partial_m n)(\partial_l n)$, which makes explicit that only the symmetric part enters. This is responsible for a number of special features that appear here. In particular, if the drift current happens to vanish— as it does in some known cases (Appendix III)—then time-reversibility is restored at the macroscopic level, although it is broken microscopically. This will become clear from our discussions in Sections 2.4 and 4.1, which imply that the Graham-Haken "potential conditions" are satisfied here. It is not true for multi-component DDS, or more generally, and we shall make no use of this fact in our discussions. Note that even for the single-component case an unambiguous definition of the full diffusion tensor, antisymmetric as well as symmetric part, is provided by the current response to a local chemical potential gradient: see Eq.(2.58) and Section 2.5

solution n_ϵ will be accurate to describe the mean density $\bar{n}_\epsilon$ not only for times $\sim \epsilon^{-2}$ but even uniformly in time, in the sense that

$$\sup_{\mathbf{r}\in\Lambda, 0<\tau<\infty} |n_\epsilon(\mathbf{r},\tau) - \bar{n}_\epsilon(\mathbf{r},\tau)| = o(\epsilon), \tag{2.62}$$

as long as the solution stays smooth (i.e. no density gradients of order ϵ^{-1} develop) and the basic assumption of a separation of scales remains valid. However, if the Euler equations develop shocks, then the shock-width for the Navier-Stokes equation will be of order $\sim \epsilon$ and the scale-separation assumption will break down. In that case, the approximation of the Navier-Stokes density to the true density in the region of the shock may be poor and an alternative description may be necessary. On the other hand, the Navier-Stokes solution should undergo a smooth approach to the final homogeneous stationary state. Entropy is no longer conserved, but instead

$$\frac{d}{d\tau} S(n_\epsilon(\tau)) = \epsilon \int_\Lambda d\mathbf{r}\ L_{ml}(n_\epsilon(\mathbf{r},\tau)) \cdot \partial_m \lambda_\epsilon(\mathbf{r},\tau) \partial_l \lambda_\epsilon(\mathbf{r},\tau). \tag{2.63}$$

Observe that only the symmetric part $\mathbf{L}^s = \frac{1}{2}(\mathbf{L}+\mathbf{L}^\top)$ contributes in this expression. We shall, in fact, show in the next section that $\mathbf{L}^s$ is a *positive-definite matrix*, so that the entropy must *increase*, $dS/d\tau \geq 0$, and $dS/d\tau = 0$ only for a homogeneous state with $\partial_m \lambda \equiv 0$. Among all of the density profiles with the same mean density $\frac{1}{|\Lambda|}\int_\Lambda d\mathbf{r}\ n(\mathbf{r}) = \bar{n}_0$, the homogeneous state $n_\infty(\mathbf{r}) = \bar{n}_0$ is the unique, absolute maximum of S when $s(n)$ is strictly concave. Therefore, $S(n)$ is a *Lyapunov functional* for the Navier-Stokes dynamics, which guarantees the approach to the final stationary state n_∞.

The derivation we have given of the hydrodynamical equations is essentially an analogue for microscopic systems of the *Chapman-Enskog expansion* in the Boltzmann equation context, a fact which was previously observed by Zubarev [2] for local-equilibrium systems. Chapman-Enskog expansion was also the method employed by Wannier in his original discussion of DDS at the Boltzmann level [12]. The "nonequilibrium distribution" may be regarded as an explicit solution of the master equation, Eq.(1.2), in so-called *normal form*. That is, it is a solution which is a functional of the conserved density field $n_\epsilon(\mathbf{r},\tau)$ through the chemical potential histories $\lambda_\epsilon(\mathbf{r},\tau)$ used in its construction. The basic perturbation method employed for the constitutive relations is one in which the fluxes are assumed to have an asymptotic expansion in the *explicit* ϵ-dependence (at least when truncated at order ϵ), but the *implicit* ϵ-dependence via the solutions n_ϵ themselves is not expanded. This is exactly the procedure used in the Chapman-Enskog method for the Boltzmann equation, which yields at zeroeth order the Euler equations, at first order the Navier-Stokes equations, and at second order the so-called "Burnett equations." We have not attempted to carry out the expansion in the DLG model to second-order, since, for most microscopic systems in $d = 3$, the resulting expressions for the Burnett-order transport coefficients will diverge as a consequence of "long-time tails" [3], [35].

We note at this point that using Hypothesis 1 we have not only succeeded to derive the hydrodynamic laws, but also we have developed microscopic expressions for all of the quantities involved: the entropy function or "fundamental equation," the Eulerian fluxes, and the Onsager relaxation coefficients (see Eqs.(2.11),(2.49),(2.57)). In a more realistic context, therefore, the methods developed here should be useful in molecular dynamics calculation of nonequilibrium transport behavior. The exact expressions may also be useful for theoretical evaluation by approximation methods, such as low-density expansions. We should emphasize that, although our methods are formal, our results agree with available rigorous results for the infinite-temperature ($\beta = 0$) case of the DLG, the so called Assymmetric Simple Exclusion Process, or ASEP (Appendix III). We shall show furthermore in Section 3.3 that the results are consistent with those of another formal method, the so-called "correlation-function approach."

A final comment here concerns the probability sense of our derivation. We have only given a (heuristic) derivation of the hydrodynamical laws for the ensemble-averages. In fact, the equations should hold in a much stronger sense, i.e. they should be true for empirical densities in individual realizations with probability going to one as $\epsilon \to 0$ (hydrodynamic law of large numbers). We shall discuss a refinement of this statement, the hydrodynamic large-deviations principle, in Section 4. No mathematical derivation of these probabilistic statements will be attempted, even at a formal level. (However, this could be done by the "level-2" method of

Zwanzig [8]). When we speak of "derivation" we mean it in the sense of Physics as "guessing the correct answer."

(2.4) Reversibility and Onsager Reciprocity

At this point we shall consider the subject of *time-reversal* for the stochastic DLG dynamics, and, in particular, the consequences for the hydrodynamics of time-reversal invariance of the stationary measures when $E = 0$. The meaning of time-reversal for stochastic dynamics is essentially the same as for classical or quantum dynamics but may be somewhat less well-known to physicists. Therefore, we shall start by briefly reviewing the subject. Basically it means that a history of the system which is *typical* of the stationary state will appear the same when run forward or backward in time. Another way of saying it is that transitions from a set $A \subset \Omega$ to a set $B \subset \Omega$ occur, in the stationary state, with the same frequency as transitions from B to A. On the more formal level, let $\{N(\mathbf{x},t)\}$ as before denote the ensemble of particle histories composed of realizations of the stationary Markov process obtained by using the stationary distribution P_n as the initial distribution on Ω and $P_t(B|\eta) = (e^{tL}\chi_B)(\eta)$ as the Markov transition probability from configuration η into subset $B \subset \Omega$ (χ_B is the characteristic function of the set B.) Then, the "time-reversed process" N^r is, very naturally, defined by

$$N^r(\cdot,t) = N(\cdot,-t). \tag{2.64}$$

This is also a stationary Markov process, with single-time distribution P_n. The generator L^r can be obtained from the definition $\langle f \cdot e^{tL} g\rangle_n = \mathsf{E}_n[f(N(0))g(N(t))]$ and $\mathsf{E}_n[f(N^r(0))g(N^r(t))] = \mathsf{E}_n[f(N(0))g(N(-t))] = \mathsf{E}_n[f(N(t))g(N(0))]$. We use the symbol $\mathsf{E}_n(\cdot)$ to denote the average in path-space over ensembles of histories, as opposed to the single-time average $\langle\cdot\rangle_n$ with distribution P_n. Therefore,

$$\langle f \cdot e^{L^r t} g\rangle_n = \langle e^{Lt} f \cdot g\rangle_n, \tag{2.65}$$

and we conclude that L^r is the adjoint $L^\dagger$ of L with respect to P_n. A point worth emphasizing is that—unlike the adjoint L^* with respect to counting measure—the adjoint $L^\dagger$ requires knowledge of the stationary measure P_n for its calculation.

It follows from these general results that the Kawasaki dynamics we consider have a time-reversal of the same type. For example, it is also conservative

$$\partial_t N^r(\mathbf{x},t) + \nabla_m^- J_m^r(\mathbf{x},t) = 0, \tag{2.66}$$

with local current $J_m^r(\mathbf{x},t) = -J_m(\mathbf{x},-t)$. In a finite volume Λ it is easy to show also that the time-reversed dynamics is a Markov jump process with rates given by

$$\frac{c^r(\mathbf{x},\mathbf{y},\eta^{\mathbf{xy}})}{c(\mathbf{x},\mathbf{y},\eta)} = \frac{P_n(\eta)}{P_n(\eta^{\mathbf{xy}})}. \tag{2.67}$$

Indeed, it is direct to check, with Eq.(2.67) as a definition, that $\langle f \cdot Lg\rangle_n - \langle L^r f \cdot g\rangle_n = \langle L(fg)\rangle_n = 0$. It is helpful here to note the identity

$$\langle f \cdot c^r(\mathbf{x},\mathbf{y})\rangle_n = \langle \Delta_{\mathbf{xy}} f \cdot c(\mathbf{x},\mathbf{y})\rangle_n. \tag{2.68}$$

The result Eq.(2.67) has also an extension to infinite-volume, if the righthand side is expressed in terms of local conditional distributions [33].

The process is said to be *reversible* if $N^r = N$ in distribution. This definition has a number of mathematically equivalent formulations. Obviously, one equivalent statement is that $L^\dagger = L$, i.e. the generator of the process should be self-adjoint with respect to measure P_n. A more intuitively appealing formulation follows from Eq.(2.67) when $c^r = c$, namely:

$$c(\mathbf{x},\mathbf{y},\eta^{\mathbf{xy}})P_n(\eta^{\mathbf{xy}}) = c(\mathbf{x},\mathbf{y},\eta)P_n(\eta), \tag{2.69}$$

which is the usual condition for *detailed balance* of the rates with respect to P_n. The DLG dynamics are not reversible if $E \neq 0$. In fact, applying the relation Eq.(2.68) for $f = \eta_{\mathbf{x}} - \eta_{\mathbf{y}}$ gives

$$\langle j_m^r(\mathbf{x})\rangle_n = -\langle j_m(\mathbf{x})\rangle_n \tag{2.70}$$

as a general relation and $\langle j_m\rangle_n = 0$ in a reversible case. (Of course, it may be true even in a non-reversible case that $\langle j_m\rangle_n = 0$: see Appendix III for rigorous results on such examples.) However, the DLG measures at finite E support a nonvanishing mean current. Of course, for

$E = 0$ the DLG dynamics is reversible with respect to the Gibbs measures with Hamiltonian H by construction (see Eq.(1.3)).

An important consequence of micro-reversibility for hydrodynamics was derived by Onsager in 1931, the famous *Onsager reciprocity* (OR) for the transport coefficients: see [10], usually referred to as RRIP I,II, and also the later elaboration with his student Machlup [36]. To explain the reciprocal relations in a wide context (e.g. see [37]), we consider a general hydrodynamic law for conserved densities $\rho_\alpha, \alpha = 1, ..., k$ in the "Onsager form":

$$\partial_t \rho_\alpha(\mathbf{r}, t) = -\partial_m \hat{\jmath}_{m\alpha}(\rho(\mathbf{r}, t)) + \partial_m \left[L_{m\alpha,l\beta}(\rho(\mathbf{r}, t)) \cdot \partial_l \lambda_\beta(\mathbf{r}, t)\right]. \tag{2.71}$$

(For simplicity of notation we set $\epsilon = 1$ here.) As with our particular example, the "chemical potentials" λ_α are conjugately related to the conserved densities through a "local entropy functional" S as, $\lambda_\alpha(\mathbf{r}) = -\delta S(\rho)/\delta\rho_\alpha(\mathbf{r})$. The entropy S is conserved by the Eulerian flux $\hat{\jmath}_{m\alpha}(\rho)$. If the time-reversal parity of the density ρ_α is $\epsilon_\alpha = \pm$, then Onsager reciprocity states that

$$\epsilon_\alpha \epsilon_\beta L_{m\alpha,l\beta}(\epsilon \cdot \rho) = L_{l\beta,m\alpha}(\rho). \tag{2.72}$$

One can also add to this the relation

$$\epsilon_\alpha \hat{\jmath}_{m\alpha}(\epsilon \cdot \rho) = -\hat{\jmath}_{m\alpha}(\rho) \tag{2.73}$$

for time-reversal of the Eulerian flux. Notice that this relation implies that the Euler part must vanish if all time-parities are even, $\epsilon_\alpha \equiv 1$. Onsager's explanation of these very general phenomenological relations as a consequence of time-reversibility of the microscopic dynamics is still considered one of the great achievements of nonequilibrium statistical physics. We shall demonstrate here that these relations are valid for the expressions we have derived from the microscopic DLG dynamics when $E = 0$ and micro-reversibility holds.

It should be pointed out that Onsager's original derivation, unlike ours below, did not use any microscopic expressions for the transport coefficients. In fact, the key hypothesis—Eq.(1.2) in RRIP II—can be reinterpreted in the Langevin formulation of Onsager and Machlup as stochastic reversibility at the level of a macroscopic thermodynamic description. In other words, with a suitable interpretation, Onsager's derivation did not even use micro-reversibility in any essential way except to motivate the assumption of *macro-reversibility* of the Langevin fluctuating hydrodynamic description. This means that it is actually somewhat more general than our derivation, because it applies also to systems without micro-reversibility but where detailed balance is restored on the macroscopic level (e.g. Rayleigh-Benárd cells near onset of convection and lasers near the critical inversion [38]). Therefore, Onsager reciprocity ought to be considered a result of macroscopic thermodynamics proper.

In our derivation of the OR for the DLG model transport coefficients, it is useful to consider a generalization of those relations which applies even when $E \neq 0$. Such a *generalized Onsager reciprocity* (GOR) was considered recently by Dufty and Rubí [39] and gives a formal extension of OR to systems without detailed balance (see also [40]). GOR relates quantities for the forward and reverse dynamics, as:

$$\epsilon_\alpha \epsilon_\beta L^r_{m\alpha,l\beta}(\epsilon \cdot \rho) = L_{l\beta,m\alpha}(\rho), \tag{2.74}$$

and

$$\epsilon_\alpha \hat{\jmath}^r_{m\alpha}(\epsilon \cdot \rho) = -\hat{\jmath}_{m\alpha}(\rho). \tag{2.75}$$

Unlike the original OR relation, it does not relate physically measurable quantities, since it is impossible in practice to realize the time-reversed dynamics in the laboratory. Nevertheless, they will turn out to be formally useful to us in our discussion of the DLG model in order to establish the equivalence of the Kadanoff-Martin and nonequilibrium distribution function methods for deriving hydrodynamics. Here the GOR relations take the simple form

$$L^r_{ml}(\rho) = L_{lm}(\rho), \tag{2.76}$$

and

$$\hat{\jmath}^r_m(\rho) = -\hat{\jmath}_m(\rho), \tag{2.77}$$

since there is only one component ($k = 1$) and the time-parity of the number density is even. We verify now that these relations hold for our microscopic expressions. The second relation, in fact, follows directly from the definitions and Eq.(2.70). For the main GOR, Eq.(2.76), it is helpful to distinguish two contributions to $\mathbf{L}$, a "static" term

$$L_{ml}^{\mathrm{stat}} = -\sum_{\mathbf{x}} x_l \langle \eta_{\mathbf{x}} j_m(\mathbf{0})\rangle_n^{\top}, \tag{2.78}$$

and a "dynamic" one

$$L_{ml}^{\mathrm{dyn}} = \int_0^\infty dt \left[\sum_{\mathbf{x}\in\mathbf{Z}^d} \langle \Delta_{\mathbf{x},\mathbf{x}+\hat{\mathbf{e}}_l} e^{tL} j_m(\mathbf{0}) \cdot j_l(\mathbf{x})\rangle_n^{\top} - \chi(n)\hat{\jmath}_m'(n)\hat{\jmath}_l'(n) \right]. \tag{2.79}$$

We verify the GOR for each separately.

For the static contribution we use the following alternative form:

$$L_{ml}^{\mathrm{stat}} = -\lim_{\Lambda\uparrow\mathbf{Z}^d} \frac{1}{|\Lambda|} \sum_{\mathbf{x},\mathbf{y}\in\Lambda} x_m y_l \langle L\eta_{\mathbf{x}} \cdot \eta_{\mathbf{y}}\rangle_{\Lambda,n}^{\top}. \tag{2.80}$$

In fact,

$$\begin{aligned} L_{ml}^{\mathrm{stat}} &= -\lim_{\Lambda\uparrow\mathbf{Z}^d} \sum_{\mathbf{y}\in\Lambda} y_l \langle j_m(\mathbf{0})\eta_{\mathbf{y}}\rangle_{\Lambda,n}^{\top} \\ &= -\lim_{\Lambda\uparrow\mathbf{Z}^d} \frac{1}{|\Lambda|} \sum_{\mathbf{x},\mathbf{y}\in\Lambda} (y_l - x_l)\langle j_m(\mathbf{x})\eta_{\mathbf{y}}\rangle_{\Lambda,n}^{\top} \\ &= \lim_{\Lambda\uparrow\mathbf{Z}^d} \frac{1}{|\Lambda|} \sum_{\mathbf{x},\mathbf{y}\in\Lambda} x_m y_l \langle (\nabla_k^- j_k)(\mathbf{x})\eta_{\mathbf{y}}\rangle_{\Lambda,n}^{\top}. \end{aligned} \tag{2.81}$$

In the last line, a contribution from the x_l-term in the previous line was neglected since it may be shown by using particle conservation, Eq.(2.45), that it is a surface term, which vanishes in the limit if correlations decay rapidly enough. Finally, Eq.(2.80) follows by applying again particle conservation. However, the new expression for $\mathbf{L}^{\mathrm{stat}}$ makes GOR manifest, since

$$L_{ml}^{\mathrm{stat},r} = -\lim_{\Lambda\uparrow\mathbf{Z}^d} \frac{1}{|\Lambda|} \sum_{\mathbf{x},\mathbf{y}\in\Lambda} x_m y_l \langle L^r\eta_{\mathbf{x}} \cdot \eta_{\mathbf{y}}\rangle_{\Lambda,n}^{\top}. \tag{2.82}$$

Therefore, $L_{ml}^{\mathrm{stat},r} = L_{lm}^{\mathrm{stat}}$ follows using $L^r = L^\dagger$.

For the dynamic contribution we also utilise an alternative form, using the identity Eq.(2.68) with $f = e^{tL} j_m(\mathbf{0}) \cdot (\eta_{\mathbf{x}} - \eta_{\mathbf{x}+\hat{\mathbf{e}}_l})$, to rewrite it as:

$$L_{ml}^{\mathrm{dyn}} = -\int_0^\infty dt \left[\sum_{\mathbf{x}\in\mathbf{Z}^d} \langle e^{tL} j_m(\mathbf{0}) \cdot j_l^r(\mathbf{x})\rangle_n^{\top} - \chi(n)\hat{\jmath}_m'(n)\hat{\jmath}_l^{r,\prime}(n) \right]. \tag{2.83}$$

The relation Eq.(2.77) was also used. However, this expression can be directly verified to satisfy GOR, since

$$\begin{aligned} L_{ml}^{\mathrm{dyn},r} &= -\int_0^\infty dt \left[\sum_{\mathbf{x}\in\mathbf{Z}^d} \langle e^{tL^r} j_m^r(\mathbf{0}) \cdot j_l(\mathbf{x})\rangle_n^\top - \chi(n)\hat{\jmath}_m^{r,\prime}(n)\hat{\jmath}_l^{\prime}(n) \right] \\ &= -\int_0^\infty dt \left[\sum_{\mathbf{x}\in\mathbf{Z}^d} \langle j_m^r(-\mathbf{x}) \cdot e^{tL} j_l(\mathbf{0})\rangle_n^\top - \chi(n)\hat{\jmath}_m^{r,\prime}(n)\hat{\jmath}_l^{\prime}(n) \right] \\ &= -\int_0^\infty dt \left[\sum_{\mathbf{x}\in\mathbf{Z}^d} \langle e^{tL} j_l(\mathbf{0}) \cdot j_m^r(\mathbf{x})\rangle_n^\top - \chi(n)\hat{\jmath}_l^{\prime}(n)\hat{\jmath}_m^{r,\prime}(n) \right] \\ &= L_{lm}^{\mathrm{dyn}}, \end{aligned} \tag{2.84}$$

as required. As for the static part, the main property used here was $L^r = L^\dagger$, in the second line. This concludes the verification that GOR is satisfied.

If we make a decomposition of $\mathbf{L}$ into even and odd parts, as:

$$\mathbf{L}^e = \frac{1}{2}(\mathbf{L} + \mathbf{L}^r) \qquad \mathbf{L}^o = \frac{1}{2}(\mathbf{L} - \mathbf{L}^r), \tag{2.85}$$

then the GOR states that the even and symmetric parts of $\mathbf{L}$ coincide and, likewise, so do the odd and antisymmetric parts:

$$\mathbf{L}^e = \mathbf{L}^s \qquad \mathbf{L}^o = \mathbf{L}^a. \tag{2.86}$$

Specializing to the case when $E = 0$, we recover the traditional OR in the DLG model. In that case $\hat{\jmath}_m(n) = 0$, so the "Euler term" of the dynamics disappears. For reversible lattice gases, the entire density change occurs on the "diffusive" time scale $\sim \epsilon^{-2}$ in microscopic units, and, by making the time scaling of this type, the factor of ϵ in front of the diffusion term is removed. Notice also that $\mathbf{L}^o = 0$, and only the even terms remain. OR here states simply that these remaining terms are symmetric,

$$\mathbf{L} = \mathbf{L}^s. \tag{2.87}$$

(2.5) Linear Response and the Einstein Relation

Locally the lattice gas sets up a diffusive flux in response to a small density gradient. As a consistency check, in linear response we should find a coefficient of proportionality which is identical to the one from the nonequilibrium distribution method. The formal manipulations in this calculation are essentially identical but the intuitive content is perhaps clearer. Let us prepare then, at $t=0$, a state with a small negative density gradient by setting

$$\mu_0(\eta) = Z(\lambda')^{-1} P_n(\eta) \exp[-\sum_{\mathbf{x}} \lambda' x_l \eta_{\mathbf{x}}] \tag{2.88}$$

The response in the average excess current at the origin at time t is given by

$$Z(\lambda')^{-1} \langle (e^{tL}(j_m(\mathbf{0}) - \langle j_m(\mathbf{0})\rangle_n)) \exp[-\sum_{\mathbf{x}} \lambda' x_l \eta_{\mathbf{x}}]\rangle_n, \tag{2.89}$$

with $\langle\cdot\rangle_n$ the stationary state at density n. To first order in λ', Eq.(2.89) equals

$$\begin{aligned}\Delta_l j_m(t) = \lambda'\{&-\sum_{\mathbf{x}} x_l \langle j_m(\mathbf{0})\eta_{\mathbf{x}}\rangle_n^{\top} \\ &-\int_0^t ds[\sum_{\mathbf{x}} \langle (e^{sL} j_m(\mathbf{0})) j_l^r(\mathbf{x})\rangle_n^{\top} - \chi(n)\hat{\jmath}'_m(n)\hat{\jmath}'_l(n)] - t\chi(n)\hat{\jmath}'_m(n)\hat{\jmath}'_l(n)\}.\end{aligned} \tag{2.90}$$

Here we differentiated the exponential and, as before, once iterated the time evolution operator $e^{tL} = 1 + \int_0^t ds\ e^{L(t-s)}L$.

Initially the average density equals $n - \lambda'\chi(n)x_l$ for small λ'. According to the Euler equations Eq.(2.50), linearized at n since λ' is small, the density at the origin at time t is $n + \lambda' t\chi(n)\hat{\jmath}'_l(n)$. This induces a change in the current at the origin as $-\hat{\jmath}'_m(n)\lambda' t\chi(n)\hat{\jmath}'_l(n)$. Subtracting out this term yields

$$\lim_{t\to\infty} \frac{1}{\lambda'} \Delta_l j_m(t) - t\chi(n)\hat{\jmath}'_m(n)\hat{\jmath}'_l(n) = L_{ml}(n) \tag{2.91}$$

in agreement with Eq.(2.57). Note that we have obtained the full, in general nonsymmetric, Onsager matrix.

Besides the response to a density gradient, it is of interest physically to consider also the response to "mechanical" perturbations, i.e. to small changes in the dynamics. In our case the driving field is singled out. The response of the average current to it defines the conductivity matrix, $\sigma_{ml}(n, \mathbf{E})$, at field strength $\mathbf{E}$. At $E=0$ the local detailed balance condition (3) implies the Einstein relation

$$\boldsymbol{\sigma}(n, E=0) = \frac{q^2}{k_B T}\mathbf{L}(n, E=0). \tag{2.92}$$

In fact, this relation does not depend on how the driving field is incorporated into the rates as long as (3) is satisfied: see [20], Section II. 2.5. For $E \neq 0$, this will no longer be the case. One fairly natural choice is

$$c_E(\mathbf{x}, \mathbf{y}, \eta) = c_0(\mathbf{x}, \mathbf{y}, \eta) \exp[-\frac{1}{2} q\beta \mathbf{E} \cdot (\mathbf{x} - \mathbf{y})(\eta_{\mathbf{x}} - \eta_{\mathbf{y}}]) \tag{2.93}$$

with c_0 the equlibrium rates satisfying

$$c_0(\mathbf{x}, \mathbf{y}, \eta) = c_0(\mathbf{x}, \mathbf{y}, \eta^{\mathbf{xy}}) \exp[-\beta(H(\eta^{\mathbf{xy}}) - H(\eta))]. \tag{2.94}$$

Then (3) holds and, taking into account that $j_m(\mathbf{0})$ also depends on $\mathbf{E}$,

$$\begin{aligned} \sigma_{ml}(n, E) &= \frac{\partial}{\partial E_l} \langle j_m(\mathbf{0}) \rangle_n \\ &= q^2 \beta \left(\frac{1}{2} \delta_{ml} \langle c(\mathbf{0}, \hat{\mathbf{e}}_m) \rangle_n - \int_0^\infty dt \sum_{\mathbf{x}} \langle \frac{1}{2} (j_l(\mathbf{x}) + j_l^r(\mathbf{x})) e^{tL} j_m(\mathbf{0}) \rangle_n \right). \end{aligned} \tag{2.95}$$

We note that $\langle j_l(\mathbf{x}) + j_l^r(\mathbf{x}) \rangle_n = 0$ and no subtraction in (2.95) is needed. At $E = 0$, since $j_l = j_l^r$, (2.95) reduces to the Einstein relation (2.92), but such an identity will not hold, in general.

If the electric field is represented by the gradient of an electrostatic potential φ, as $\mathbf{E} = -\boldsymbol{\nabla}\varphi$, then as long as the field is weak enough for linear response to be valid, there is a useful combination of electrostatic and chemical potentials. (Note that the gradient representation by a single-valued potential is not possible for a constant E with our periodic b.c., but we have in mind more physically realistic situations.) According to the Einstein relation Eq.(2.92), the constants of proportionality in the linear response to $-\boldsymbol{\nabla}\varphi$ and $-\frac{1}{q}\boldsymbol{\nabla}\mu$ are identical. In that case, it is useful to consider a combined *electrochemical potential* $\varphi + \frac{1}{q}\mu$; cf. Landau and Lifshitz, [27], Section 25. However, it should be clear that this is only possible in the weak-field regime, and that the response to chemical potential gradient and the differential response to the electric field in the strong field case are generally distinct.

3 Current Noise and the Fluctuation-Dissipation Relation

(3.1) A Review of Standard Theory

We now turn to a somewhat different subject, the theory of fluctuations about the hydrodynamic behavior whose description was derived in the previous section. While the predictions of the hydrodynamic equations, like Eq.(2.61), are observed with a probability going to one as $\epsilon \to 0$, there will also be small, random corrections $\sim \epsilon^{d/2}$. That is, if the "fluctuation variable"

$$X^{\epsilon}(A,\tau) = \frac{1}{\epsilon^{d/2}}\left[\epsilon^{d}\sum_{\epsilon\mathbf{x}\in A} e^{\epsilon^{-1}\tau L}\eta_{\mathbf{x}} - \int_A d\mathbf{r}\ \bar{n}_{\epsilon}(\mathbf{r},\tau)\right], \tag{3.1}$$

is considered for any macroscopic time τ and region $A \subset \Lambda$, then it is expected that its distribution will converge to a Gaussian for $\epsilon \to 0$. Observe that the normalization $\sim \epsilon^{-d/2}$ of the sum corresponds to a standard central limit theorem scaling, since the size of the region A in microscopic units grows as $\sim \epsilon^{-d}$. The basic problem of fluctuation theory is to derive the form of the limiting distribution laws. A physical theory for this in the case of the Navier-Stokes system was proposed by Landau and Lifshitz in 1959 [41], the so-called "fluctuating hydrodynamics," which was an application to simple fluids of general principles set out in the work of Onsager and Machlup [36]. Extended to the more general context of hydrodynamics in "Onsager form," their hypothesis was that $X^{\epsilon}_{\alpha}(A,\tau)$ is approximated in the limit $\epsilon \to 0$ by $\int_A d\mathbf{r}\ \xi^{\epsilon}_{\alpha}(\mathbf{r},\tau)$, where $\xi^{\epsilon}_{\alpha}(\mathbf{r},\tau)$ is the solution of the stochastic PDE (Langevin equation)

$$\partial_{\tau}\xi^{\epsilon}_{\alpha}(\mathbf{r},\tau) = -\mathcal{H}_{\alpha\beta}(\bar{\rho}(\mathbf{r},t))\xi^{\epsilon}_{\beta}(\mathbf{r},\tau) - \boldsymbol{\nabla}\cdot\mathbf{j}'_{\alpha}(\mathbf{r},\tau), \tag{3.2}$$

with "linearized hydrodynamic operator" given by $\mathcal{H}^{\epsilon}_{\alpha\beta}(\bar{\rho}(\mathbf{r},t))f(\mathbf{r}) = \partial_m[H^{\epsilon}_{m,\alpha\beta}(\bar{\rho}(\mathbf{r},t))f(\mathbf{r})]$ and

$$H^{\epsilon}_{m,\alpha\beta}(\rho) = \frac{\partial\hat{\jmath}_{m\alpha}}{\partial\rho_{\beta}}(\rho) - \epsilon L_{m\alpha,l\gamma}(\rho)\frac{\partial\lambda_{\gamma}}{\partial\rho_{\beta}}(\rho)\partial_l. \tag{3.3}$$

The $j'_{m\alpha}(\mathbf{r},\tau)$ are "random fluxes," which were taken to be Gaussian fields with zero-mean. In other words, the fluctuations were assumed to be governed by the hydrodynamic equations linearized about the deterministic solution $\bar{\rho}_{\alpha}(\mathbf{r},\tau)$, to which were added additional stochastic currents representing effects of molecular noise. To complete the theory, a prescription was given also for the covariance of the random fluxes, as

$$\langle j'_{m\alpha}(\mathbf{r},\tau)j'_{l\beta}(\mathbf{r}',\tau')\rangle = 2k_B \cdot \epsilon L^s_{m\alpha,l\beta}(\bar{\rho}(\mathbf{r},\tau))\delta(\mathbf{r}-\mathbf{r}')\delta(\tau-\tau'), \tag{3.4}$$

where k_B is Boltzmann's constant and $\mathbf{L}^s$ is the symmetric part of the Onsager matrix. This is a particular example of a *fluctuation-dissipation relation*, since it relates the covariance of fluctuating currents to the dissipative Onsager coefficients.

There are actually a number of distinct—but related—general types of FDR's. A quite recent monograph of Stratonovich [42] gives a detailed discussion of linear and nonlinear fluctuation-dissipation theorems with a wide variety of applications. This work makes a classification of FDR's into 1st, 2nd, and 3rd types. We shall briefly discuss here the various types following Stratonovich, although his terminology for FDR's of 1st and 2nd kind is exactly the *opposite* of that in the Western literature. Note: *we follow here Stratonovich' classification!* Although there is some weight of tradition against this usage, we find it supported by the internal logic of the subject.

In Stratonovich' classification, the FDR of 1st type is a relation between noise characteristics and dissipation elements, e.g. the noise covariance and the (Onsager) relaxation coefficient at linear level such as is given in Eq.(3.4). The Einstein relation (ER) [43] can be considered a prototype of such a relation. In fact, if the conductivity $\boldsymbol{\sigma}$ is considered to be the linear coefficient in the Langevin dynamics

$$\dot{x}_i(t) = -\sigma_{ij}\frac{\partial U}{\partial x_j}(x(t)) + \eta_i(t) \tag{3.5}$$

and $\mathbf{D}$ is taken to be the noise-strength parameter

$$\langle \eta_i(t)\eta_j(0)\rangle = 2D_{ij}\delta(t) \tag{3.6}$$

then

$$P(\mathbf{x}) \sim \exp[-\beta U(\mathbf{x})] \tag{3.7}$$

is the stationary measure if and only if

$$\boldsymbol{\sigma} = \beta \mathbf{D}. \tag{3.8}$$

This is a typical FDR of 1st type, and it is the type which is most important for this work.

The FDR of 2nd type according to Stratonovich is a relation between time-correlation functions and (dissipative) response functions to an external perturbation. The Callen-Welton relation [44]was a first case of 2nd-type FDR for quantum systems. In our previous example, the linear FDR of 2nd type is

$$\beta\langle \dot{x}_i(t)x_j(0)\rangle = -\langle \frac{\delta x_i(t)}{\delta E_j(0)}\rangle, \quad t > 0 \tag{3.9}$$

for expectations in the stationary state, when the external field $\mathbf{E}(t)$ is coupled into the Langevin equation as:

$$\dot{x}_i(t) = \sigma_{ij}\left[-\frac{\partial U}{\partial x_j}(x(t)) + E_j(t)\right] + \eta_i(t). \tag{3.10}$$

The ER is also closely related to the FDR of 2nd type, if one now considers $\boldsymbol{\sigma}$ as a response to an external field and $\mathbf{D}$ as defined in terms of time-correlations. The 2nd-type FDR is actually slightly more general than the ER. In fact, one can define a conductivity response kernel $\sigma_{ij}(t) = \langle \delta v_i(t)/\delta E_j(0)\rangle$, giving the delayed current response, with $\mathbf{v}(t) \equiv \dot{\mathbf{x}}(t)$. Taking a time-derivative of the FDR, one obtains the relation

$$\beta\langle v_i(t)v_j(0)\rangle = \sigma_{ij}(t). \tag{3.11}$$

In the Fourier representation this becomes

$$\sigma_{ij}(\omega) = \beta\int_0^\infty dte^{-i\omega t}\langle v_i(t)v_j(0)\rangle, \tag{3.12}$$

which gives the full frequency response. It is not hard to show that Eq.(3.12) yields $\sigma_{ij}(\omega = 0) = \beta D_{ij}$.

It is a general feature that the FDR of 2nd type implies the FDR of 1st type (e.g. [42], Section 5.5.3), as one sees here by taking $\omega = 0$. To obtain the FDR of second type requires an appropriate special coupling of the external field, which can always be done ([42], Section 5.5.1, also [24] and the following section). The relation between FDR's of 1st and 2nd types is intuitive since the roles of fluctuational and (appropriate) external forces are essentially interchangeable. Stratonovich also defines another set of relations, FDR's of 3rd type, as those existing between noise characteristics and response functions, e.g. random force covariance and linear response function. The Nyquist relation [45] in electrical circuits between the power spectrum of the noisy EMF (electromotive force) and the frequency-dependent impedance is a prototypical example. We will not consider this subject here.

It is our purpose here to consider the extension of the Landau-Lifshitz method to the systems without local equilibrium and to develop corresponding prescriptions for the random fluxes, such as current noise in electrical conductors. It seems to be still not generally appreciated that there is a *generalized fluctuation-dissipation relation* which applies to such systems. In fact, the theory is rather scattered throughout the literature, and we shall develop the subject *ab initio* here. The presentation we give below is therefore not precisely the same as that we have found anywhere in the literature of the subject, but it borrows from a number of others. An important early work was that of Price [21], who proposed a charge diffusion-current noise relation in semiconductors and plasmas, which we shall discuss in the next subsection. The classic work of Fox and Uhlenbeck [46] also derived the generalized FDR in the Langevin equation context in a way that extends directly to systems without local equilibrium (although they did not consider that extension). A paper of Tomita and Tomita [47] made an important decomposition of the systematic flux in the linear Langevin equation which is relevant to the FDR, since it corresponds to a separation into "conservative" and "dissipative" terms. Our perspective on this has been strongly influenced by the work of Graham in 1977 [24], who generalized this decomposition to general nonlinear Langevin equations. In fact, we believe that Graham's paper is an important contribution to the subject which has been unduly neglected, perhaps because of the abstract style of presentation. The last section of this paper will discuss that work extensively and hopefully will make its physical relevance more clear. A number of review articles and books have also been useful to us. The paper of Gantsevich et al. [48], contains a wealth of information on the theory of fluctuations in nonequilibrium electron gases, developed from the point of view of Boltzmann transport equations and slanted toward the Soviet work on semiconductors. A more general review article is that of Tremblay [49], which discusses these developments and parallel work in the West up to a decade ago. The general philosophy of Stratonovich in [42] is also very close to and has influenced ours.

(3.2) The Generalized FDR in Linear Theory

The FDR's of 1st and 2nd type are established quite easily in the context of a general linear Langevin equation, as we now discuss. In particular, we wish to make clear in our derivation that the FDR is independent of any near-equilibrium assumption. We consider, as in the original paper of Onsager-Machlup, a discrete set of variables $\boldsymbol{\alpha} = \{\alpha_i | i = 1, ..., p\}$. It is quite easy to extend the theory to spatially extended systems as above by considering the index i to stand for $(\mathbf{r}, m, \alpha)$. The variables evolve according to

$$\dot{\alpha}_i = -H_{ij}\alpha_j + \eta_i, \tag{3.13}$$

a linear relaxational equation, $\mathbf{H}^s \geq 0$, with Gaussian white-noise force

$$\langle \eta_i(t)\eta_j(t')\rangle = 2k_B Q_{ij}\delta(t-t'). \tag{3.14}$$

The stationary probability distribution of $\boldsymbol{\alpha}$ is then also Gaussian:

$$P(\boldsymbol{\alpha}) \propto \exp\left[-\frac{1}{2k_B}(\mathbf{g}^{-1})_{ij}\alpha_i\alpha_j\right], \tag{3.15}$$

where $k_B \cdot \mathbf{g}$ is the steady-state covariance

$$\langle \alpha_i \alpha_j \rangle = k_B g_{ij}. \tag{3.16}$$

To obtain $\mathbf{g}$ ones solves the Langevin equation as

$$\boldsymbol{\alpha}(t) = e^{-\mathbf{H}t}\boldsymbol{\alpha}_0 + \int_0^t ds\ e^{-\mathbf{H}(t-s)}\boldsymbol{\eta}(s), \tag{3.17}$$

and then calculates $\langle \alpha_i(t)\alpha_j(t)\rangle$ in the limit $t \to +\infty$. This yields

$$\mathbf{g} = 2\int_0^\infty dt\; e^{-\mathbf{H}t}\mathbf{Q}e^{-\mathbf{H}^\top t}. \tag{3.18}$$

As a direct consequence, we obtain

$$\mathbf{H}\mathbf{g} + \mathbf{g}\mathbf{H}^\top = 2\mathbf{Q}. \tag{3.19}$$

It is this relation which is the FDR of first type and arises as a simple "stationarity" or "balance" condition. The above general derivation goes back to [46].

However, the Eq.(3.19) does not give the FDR in its most familiar form, which involves "dissipative Onsager coefficients" appearing in a "force-flux form" of the equations. Even without the near-equilibrium assumption, we may *define* an "entropy" associated to the steady-state distribution $P(\boldsymbol{\alpha})$ as $s(\boldsymbol{\alpha}) = k_B \cdot \log P(\boldsymbol{\alpha})$, or

$$s(\boldsymbol{\alpha}) = s_0 - \frac{1}{2}(\mathbf{g}^{-1})_{ij}\alpha_i\alpha_j. \tag{3.20}$$

We can also introduce a "force"

$$X_i = -\frac{\partial s}{\partial \alpha_i}. \tag{3.21}$$

The Langevin equation is then expressed in "force-flux form," as:

$$\dot{\alpha}_i = -L_{ij}X_j + \eta_i, \tag{3.22}$$

with $\mathbf{L} = \mathbf{H}\mathbf{g}$, which are the "Onsager coefficients." In terms of these the Eq.(3.19) becomes simply

$$2\mathbf{Q} = \mathbf{L} + \mathbf{L}^\top. \tag{3.23}$$

It just means that the noise strength Q_{ij} is the symmetric part of L_{ij},

$$\mathbf{Q} = \mathbf{L}^s, \tag{3.24}$$

which is the standard linear FDR of 1st type. Its derivation is completely straightforward. The distinction of the general case from the near-equilibrium one is that, for equilibrium systems, the "entropy" is the ordinary thermodynamic equilibrium entropy which has been much studied and is known for many systems by a theoretical or empirical determination of the fundamental equation. For nonequilibrium systems, such as the DDS, the quantity $s(\boldsymbol{\alpha})$, or $\mathbf{g}$, is not readily available. Nevertheless, the relationship between the variables $\mathbf{Q}, \mathbf{H}$, and $\mathbf{g}$ remains valid.

The terminology "fluctuation-dissipation" is explained in the present context, by considering the decomposition of $\mathbf{L}$ into its symmetric part $\mathbf{L}^s$ and it anti-symmetric part denoted $\mathbf{L}^a$,

$$L_{ij} = L^a_{ij} + L^s_{ij}. \tag{3.25}$$

Correspondingly, one can define a "conservative flux"

$$r_i = -L^a_{ij} X_j, \tag{3.26}$$

and a "dissipative flux"

$$d_i = -L^s_{ij} X_j. \tag{3.27}$$

Since $\partial s / \partial \alpha_i = -X_i$ and $\partial X_j / \partial \alpha_i = \left(\mathbf{g}^{-1}\right)_{ij}$, the r_i automatically satisfy

$$\frac{\partial s}{\partial \alpha_i} r_i(\alpha) = 0 \tag{3.28}$$

from symmetry of $X_i X_j$, and

$$\frac{\partial r_i}{\partial \alpha_i} = 0 \tag{3.29}$$

from symmetry of g_{ij}. The first of these implies that the evolution under r_i indeed conserves the "entropy" s, while the second is a Liouville theorem. Hence, the entire evolution of the "entropy" under the systematic (non-random) evolution arises from d_i, and is calculated as

$$\frac{d}{dt} s(\alpha) = L^s_{ij} X_i X_j \geq 0. \tag{3.30}$$

The positivity follows because $\mathbf{L}^s$ is, by the FDR, a covariance matrix, which is necessarily positive-definite. Therefore, the general decomposition of the linear Langevin equation

$$\dot{\alpha}_i = r_i + d_i + \eta_i \tag{3.31}$$

is achieved into parts "conservative" and "dissipative" for the "entropy" of the stationary state. The same decomposition was made by Tomita and Tomita [47] from a different motivation, discussed below.

Note that one can also show easily in this context the FDR of 2nd type. The principle is to add the external force $\mathbf{F}$ in the equation so that the stationary measure is changed by the addition of a linear term $\mathbf{F}\cdot\boldsymbol{\alpha}$ in the exponent. It is not hard to show that for this the driven equation must be taken to be

$$\begin{aligned}\dot{\alpha}_i &= -L_{ij}(X_j - F_j) + \eta_i \\ &= -H_{ij}\alpha_j + L_{ij}F_j + \eta_i.\end{aligned} \tag{3.32}$$

Then the response function $G_{ij}(t) = \delta\alpha_i(t)/\delta F_j(0)$ (which is now deterministic) satisfies

$$\dot{G}_{ij}(t) = -H_{ik}G_{kj}(t) + L_{ij}\delta(t), \tag{3.33}$$

whose solution is

$$G_{ij}(t) = \left[e^{-\mathbf{H}t}\mathbf{L}\right]_{ij}\theta(t). \tag{3.34}$$

On the other hand, the linear regression solution to the Langevin equation is

$$\langle\alpha_i(t)\alpha_j(0)\rangle = k_B\left[e^{-\mathbf{H}|t|}\mathbf{g}\right]_{ij}\theta(t) + k_B\left[\mathbf{g}e^{-\mathbf{H}^\top|t|}\right]_{ij}\theta(-t). \tag{3.35}$$

The FDR of 2nd type

$$\langle\dot{\alpha}_i(t)\alpha_j(0)\rangle = -k_B\left[G_{ij}(t) - G_{ji}(-t)\right], \tag{3.36}$$

follows automatically.

It is important to note that the FDR's all follow (rather simply!) from the assumption of a general description in terms of a random process described by a Langevin stochastic equation. No condition of time-reversibility or detailed balance is required. In fact, they are results of pure theory of random processes and involve "no physics." Actually, the physics enters in from the validity of the description by a Gaussian random process with some stable stationary state. This imposes some nontrivial requirement on the microscopic dynamics (Appendix IV).

Although the FDR as such does not require detailed balance, an important simplification occurs in that case. In fact, suppose that the variables α_i have time parities ϵ_i, so that the Onsager coefficient transforms under time-reversal as

$$L^r_{ij} = \epsilon_i \epsilon_j L_{ij}. \tag{3.37}$$

We are considering a generalized time-reversal transformation in which the histories $\{\alpha_i(t)\} \to \{\alpha^r_i(t) \equiv \epsilon_i \alpha_i(-t)\}$, with $\epsilon_i = \pm$. If detailed balance holds and the OR relations are therefore satisfied, then

$$L^r_{ij} = L_{ji}, \tag{3.38}$$

so that the decomposition of $\mathbf{L}$ into parts even and odd under time-reversal coincides with the decomposition into a symmetric, or "dissipative," part and an anti-symmetric, or "conservative," part: $\mathbf{L}^o = \mathbf{L}^a, \mathbf{L}^e = \mathbf{L}^s$. Equivalently, the "conservative" and "dissipative" fluxes are, respectively, odd and even under time-reversal,

$$r_i(\epsilon \cdot \boldsymbol{\alpha}) = -\epsilon_i r_i(\boldsymbol{\alpha}), \qquad d_i(\epsilon \cdot \boldsymbol{\alpha}) = +\epsilon_i d_i(\boldsymbol{\alpha}), \tag{3.39}$$

and they can be distinguished *a priori* on that basis. Without the reversibility condition, this decomposition into "conservative" and "dissipative" terms requires knowledge of the stationary measure, as above.

In this connection, we would like to make a few remarks on the interesting work of Tomita & Tomita (TT) [47]. They made as well the decomposition of the Onsager matrix $\mathbf{L}$ into its symmetric part and its antisymmetric part. They referred to the latter as the "irreversible circulation of fluctuation" and connected its nonvanishing with the violation of OR, interpreted as the symmetry of $\mathbf{L}$. However, it seems to us that TT's interpretation of their results is somewhat misleading. The presence or absence of the anti-symmetric part $\mathbf{L}^a$ is not related to microscopic reversibility in general, as they suggest. In fact, the interpretation of symmetry of $\mathbf{L}$ as Onsager symmetry is also wrong in general. It seems to be the source of a lot of confusion that the "dissipative" Onsager coefficient is *always* the symmetric part $\mathbf{L}^s$, but this has nothing to do with time-reversal or Onsager reciprocity. Second, it is not in general implied by time-reversibility that $\mathbf{L}^a = \mathbf{0}$, i.e. that $\mathbf{L} = \mathbf{L}^s$. This is only the case if all the parities $\epsilon_i = 1$. While this was, in fact, true for the systems TT considered, it need not always be the case. An obvious example, already pointed out in this context in the classic work of Fox and Uhlenbeck [46], is the simple fluid described by the compressible Navier-Stokes equations which has a non-vanishing Euler part. The possibility to have such a term here arises from the -1 parity of the velocity field, or momentum density. It is only for the cases with all parities +1, as for the DDS example, that the presence of the Euler term, or "irreversible circulation," can be attributed to violation of detailed balance.

(3.3) The Correlation Function Approach to Hydrodynamics

One of the main assumptions in the theory of fluctuations, reviewed in the previous section, is that the small spontaneous fluctuations from the stationary mean will decay according to the linearized hydrodynamic law governing macroscopic disturbances. This idea goes back to the original work of Onsager in 1931 and is known as the *Onsager regression hypothesis.* This assumption can be made the basis of a microscopic method for deriving hydrodynamics and calculating transport coefficients, the so-called "correlation-function method" which was pioneered in work of Kadanoff and Martin [22] (see also [5]). In fact, if $\Delta\alpha_i(t) \equiv \alpha_i(t) - \alpha_i(0)$, then it follows from the linear regression solution Eq.(3.35) that

$$\langle \Delta\alpha_i(t)\alpha_j(0)\rangle = k_B \left[e^{-\mathbf{H}t}\mathbf{g}\right]_{ij} - k_B \cdot g_{ij} \tag{3.40}$$

for $t > 0$. Hence

$$\begin{aligned} \lim_{t\to 0} \frac{1}{t}\langle \Delta\alpha_i(t)\alpha_j(0)\rangle &= -k_B \left[\mathbf{H}\mathbf{g}\right]_{ij} \\ &= -k_B L_{ij}. \end{aligned} \tag{3.41}$$

The Onsager matrix L_{ij} can thus be calculated from the correlation function $\langle \Delta\alpha_i(t)\alpha_j(0)\rangle$, determined by another method, e.g. a microscopic calculation. When using the latter, then the limit $t \to \infty$ should be considered instead, since the time in the Langevin equation must be considered a "macroscopic time." The limit of "macroscopic time" $\tau \to 0$ must match onto the limit of "microscopic time" $t \to \infty$. Although this method was originally applied by Kadanoff and Martin to hydrodynamics for near-equilibrium fluids, it should be quite general since its basis is just the regression hypothesis. In fact, the method was used already in the original paper of KLS to derive the linearized hydrodynamics of their lattice DDS model. We shall reconsider the calculation here because it provides an important check on the "nonequilibrium distribution method" developed in the first section. Also, the original discussion of KLS contained a few minor mistakes which this work corrects.

To begin, let us write down the form of the driven diffusion equation derived previously, Eq.(2.61), linearized about the homogeneous state of density n :

$$\partial_t \xi(\mathbf{r},t) = -c_m(n)\partial_m \xi(\mathbf{r},t) + D^s_{ml}(n)\partial_m\partial_l \xi(\mathbf{r},t). \tag{3.42}$$

(Again we set $\epsilon = 1$ for simplicity.) Note that the linearized equation contains only the *symmetric part* of the diffusion matrix (a feature special to the 1-component case.) The equation can also be Fourier transformed in space, to give

$$\partial_t \hat{\xi}(\mathbf{k},t) = i\mathbf{c}\cdot\mathbf{k}\hat{\xi}(\mathbf{k},t) - (\mathbf{k}\cdot\mathbf{D}\cdot\mathbf{k})\hat{\xi}(\mathbf{k},t), \tag{3.43}$$

where $\mathbf{c} = \mathbf{c}(n), \mathbf{D} = \mathbf{D}^s(n)$. This is often more convenient since the different wavenumber components are uncoupled. If the fluctuation theory of the previous subsection is applied, then we should add to this equation a noisy current $\hat{\mathbf{J}}'$, as

$$\partial_t \hat{\xi}(\mathbf{k},t) = i\mathbf{c}\cdot\mathbf{k}\hat{\xi}(\mathbf{k},t) - (\mathbf{k}\cdot\mathbf{D}\cdot\mathbf{k})\hat{\xi}(\mathbf{k},t) - i\mathbf{k}\cdot\hat{\mathbf{J}}'(\mathbf{k},t), \tag{3.44}$$

with

$$\langle \hat{J}'_i(\mathbf{k},t)\hat{J}'_j(\mathbf{k}',t')\rangle = 2R_{ij}(\mathbf{k})\delta(\mathbf{k}-\mathbf{k}')\delta(t-t'). \tag{3.45}$$

Then, from the regression solution, Eq.(3.35), the "structure function"

$$S(\mathbf{k},t) = \int d\mathbf{r}\; e^{i\mathbf{k}\cdot\mathbf{r}}\langle \xi(\mathbf{r},t)\xi(\mathbf{0},0)\rangle, \tag{3.46}$$

takes the value for $t > 0$

$$S(\mathbf{k}, t) = S(\mathbf{k}) \exp[i(\mathbf{c} \cdot \mathbf{k})t - (\mathbf{k} \cdot \mathbf{D} \cdot \mathbf{k})t]. \tag{3.47}$$

Here, $S(\mathbf{k})$ is the static value of the structure function, or stationary covariance, which from the FDR Eq.(3.24), is given as

$$S(\mathbf{k}) = \frac{\mathbf{k} \cdot \mathbf{R}(\mathbf{k}) \cdot \mathbf{k}}{\mathbf{k} \cdot \mathbf{D} \cdot \mathbf{k}}. \tag{3.48}$$

It is then easy to calculate that

$$\left.\frac{\partial}{\partial k_m} S(\mathbf{k}, t)\right|_{\mathbf{k}=\mathbf{0}} = \left(\frac{\partial}{\partial k_m} S\right)(\mathbf{0}) - i c_m t S(\mathbf{0}). \tag{3.49}$$

In fact, it follows from the evenness in $\mathbf{r}$ of the density-density correlation that $S(\mathbf{k})$ is even in $\mathbf{k}$, so that

$$\left(\frac{\partial}{\partial k_m} S\right)(\mathbf{0}) = 0. \tag{3.50}$$

In the same way, it is easy to check that

$$\left.\frac{\partial^2}{\partial k_m \partial k_l} S(\mathbf{k}, t)\right|_{\mathbf{k}=\mathbf{0}} = \left(\frac{\partial^2}{\partial k_m \partial k_l} S\right)(\mathbf{0}) - 2D_{ml} t S(\mathbf{0}) - c_m c_l t^2 S(\mathbf{0}). \tag{3.51}$$

Here we have assumed that $S(\mathbf{k})$ is twice-differentiable at $\mathbf{k} = \mathbf{0}$, an assumption that will be critically re-examined in the following subsection. If we now go back to the microscopic model and identify $S(\mathbf{0}) = \chi(n)$, then these results suggest that $\mathbf{c}, \mathbf{D}^s$ should be determined from the microscopic dynamics as

$$c_m(n) = \lim_{t \to \infty} \frac{1}{t} \left(\frac{1}{\chi(n)} \sum_{\mathbf{x}} x_m \langle e^{tL} \eta_{\mathbf{x}} \eta_{\mathbf{0}} \rangle_n^\top \right), \tag{3.52}$$

and

$$D^s_{ml}(n) = \lim_{t \to \infty} \frac{1}{2t} \left(\frac{1}{\chi(n)} \sum_{\mathbf{x}} x_m x_l \langle e^{tL} \eta_{\mathbf{x}} \eta_{\mathbf{0}} \rangle_n^\top - c_m(n) c_l(n) t^2 \right). \tag{3.53}$$

The righthand sides of these equalities were calculated in a formal (non-rigorous) way by KLS in Appendix 3 of [14]. The result for $\mathbf{c}$ was

$$c_m(n) = \hat{\jmath}'_m(n), \tag{3.54}$$

and the result for $\mathbf{D}^s$ was

$$D^s_{ml}(n) = \frac{1}{2}\int_{-\infty}^{+\infty} dt\ \left[\frac{1}{\chi(n)}\sum_{\mathbf{x}} \mathsf{E}_n(J_m(\mathbf{x},t)J_l(\mathbf{0},0))_n^\top - \hat{\jmath}'_m(n)\hat{\jmath}'_l(n)\right], \tag{3.55}$$

where $\mathbf{J}(\mathbf{x},t)$ is the instantaneous particle current introduced in Eq.(2.44).

Note that the result for $\mathbf{c}$ is exactly that found by the distribution function method in the previous section. To compare the results for $\mathbf{D}$ the current-current correlation must be evaluated. This was done in Appendix 3 of [14], but the result there contained an error (it does not give a symmetric result for D^s_{lm}.) A correct calculation (Appendix V) gives

$$\begin{aligned} \mathsf{E}_n(J_m(\mathbf{x},t)J_l(\mathbf{y},s)) &= \langle c(\mathbf{0},\hat{\mathbf{e}}_m)\rangle_n \delta_{ml}\delta_{\mathbf{xy}}\delta(t-s) \\ &\quad -\theta(t-s)\langle e^{L(t-s)} j_m(\mathbf{x})\cdot j^r_l(\mathbf{y})\rangle_n \\ &\quad -\theta(s-t)\langle e^{L(s-t)} j_l(\mathbf{y})\cdot j^r_m(\mathbf{x})\rangle_n . \end{aligned} \tag{3.56}$$

Substitution into Eq.(3.55) gives

$$\begin{aligned} D^s_{ml}(n) &= \frac{1}{2\chi(n)}\langle c(\mathbf{0},\hat{\mathbf{e}}_m)\rangle_n \delta_{ml} \\ &- \int_0^\infty dt\ \left[\frac{1}{2\chi(n)}\sum_{\mathbf{x}}\left(\langle e^{tL} j_m(\mathbf{x})\cdot j^r_l(\mathbf{0})\rangle_n^\top + \langle e^{tL} j_l(\mathbf{x})\cdot j^r_m(\mathbf{0})\rangle_n^\top\right) - \hat{\jmath}'_m(n)\hat{\jmath}^{r,\prime}_l(n)\right] \end{aligned} \tag{3.57}$$

Comparison with Eq. (2.83) shows that the "dynamic term" above agrees with the result of the distribution function calculation for the symmetric part of the diffusion matrix. To verify that the "static terms" are also the same we note that, by GOR, the symmetric and even parts must coincide. Using this fact, the symmetric part from the distribution method can be written as

$$D^{\mathrm{stat},s}_{ml} = -\frac{1}{2\chi(n)}\sum_{\mathbf{x}} x_l\langle \eta_{\mathbf{x}}(j_m + j^r_m)\rangle_n^\top . \tag{3.58}$$

However, this can be explicitly calculated using the identity Eq.(2.68), with $f(\eta) = \eta_{\mathbf{z}}(\eta_{\mathbf{x}} - \eta_{\mathbf{y}})$, and the result is

$$\begin{aligned} D^{\mathrm{stat},s}_{ml} &= -\frac{1}{2\chi(n)}\sum_{\mathbf{x}} x_l\langle (\eta_{\mathbf{x}} - \eta^{\mathbf{0},\hat{\mathbf{e}}_m}_{\mathbf{x}}) j_m)\rangle_n^\top \\ &= -\frac{1}{2\chi(n)}\delta_{lm}\langle (\eta_{\hat{\mathbf{e}}_m} - \eta_{\mathbf{0}}) j_m)\rangle_n \end{aligned}$$

$$= \frac{1}{2\chi(n)}\delta_{lm}\langle c(\mathbf{0},\hat{\mathbf{e}}_m)\rangle_n. \tag{3.59}$$

(Observe that the result is indeed symmetric, as required by GOR.) Comparing with Eq.(3.57), we see that the "static" term there also coincides with the symmetric part from the distribution function calculation. This is an important consistency check on both methods. However, the correlation function method in the 1-component case is unable to determine the anti-symmetric part of the diffusion matrix (as defined by the current response).

To complete this subsection, we shall now show that the matrix $\mathbf{D}^s(n)$ is positive-definite and (since $\chi(n) > 0$) so also is $\mathbf{L}^s(n)$, as claimed in the previous section. This is important since it guarantees that the "entropy" $S(n)$ is a Lyapounov functional for the nonlinear dynamics, monotonically increasing in time to its maximum value. The positivity is a consequence of the fact that the expression Eq.(3.55) for the symmetric diffusion is, in fact, a standard *Green-Kubo formula*, as we now demonstrate. For this we need to make some definitions. For local random variables $A(\mathbf{x})$ in the probability space of the microscopic process $N(\mathbf{x},\cdot)$, we define an inner-product, as

$$\langle A|B\rangle_n = \sum_{\mathbf{x}} \mathsf{E}_n(A(\mathbf{x})B(\mathbf{0}))^\top. \tag{3.60}$$

The elements $A(\mathbf{x})$ subject to the finite-norm condition $\|A\|_n^2 = \langle A|A\rangle_n < \infty$ (defined modulo total gradients $A(\mathbf{x}) \sim A(\mathbf{x}) + \nabla\Phi(\mathbf{x})$, which have zero norm) form a Hilbert space. In the physics literature, this is known as the *Zwanzig-Mori space* (e.g. see [5]). With our normalization of the inner-product

$$\|N\|_n^2 = \chi(n), \tag{3.61}$$

by definition. Notice that

$$\begin{aligned} \langle N|J_m\rangle_n &= \sum_{\mathbf{x}} \mathsf{E}_n(N(\mathbf{x})J_m(\mathbf{0}))^\top \\ &= \sum_{\mathbf{x}} \langle \eta_{\mathbf{x}} j_m(\mathbf{0})\rangle_n^\top \\ &= \hat{\jmath}_m'(n)\chi(n), \end{aligned} \tag{3.62}$$

where the last line follows from Eq.(2.43) in the previous section. Thus, the Eulerian velocity

$c_m(n) = \langle N|J_m\rangle_n/\|N\|_n$ is the projection of the microscopic flux onto the space of conserved variables spanned by $\{N\}$. In fact, observe that with these definitions the formula Eq.(3.55) may be rewritten as

$$D^s_{ml}(n) = \frac{1}{2\chi(n)}\int_{-\infty}^{+\infty} dt\ \left[\langle J_m(t)|J_l\rangle_n - \frac{\langle J_m|N\rangle_n\langle N|J_l\rangle_n}{\langle N|N\rangle_n}\right]. \tag{3.63}$$

This is the standard form of the Green-Kubo formula in which the time-integrand is a current autocorrelation function in the Zwanzig-Mori space with the projection onto the conserved subspace subtracted. It is usual to define projection operators,

$$P = \frac{|N\rangle\langle N|}{\langle N|N\rangle}, \quad Q = \mathbf{1} - \frac{|N\rangle\langle N|}{\langle N|N\rangle}, \tag{3.64}$$

the so-called *Zwanzig-Mori projectors*, so that Eq.(3.63) can also be written as

$$D^s_{ml}(n) = \frac{1}{2\chi(n)}\int_{-\infty}^{+\infty} dt\ \langle J_m(t)|Q\cdot J_l\rangle_n. \tag{3.65}$$

This exhibits $\mathbf{D}^s$ in an explicitly positive-definite form, since it is the integrated autocorrelation of the “fast” component of the current, $I_m \equiv Q\cdot J_m$.

It is quite generally true that the Green-Kubo formulae for the dissipative Onsager coefficients can be regarded as *microscopic versions* of the FDR of 1st type, where now the relaxation coefficients are related to the covariance of microscopic phase space functions, the “fast” components of currents, and the averages are taken with respect to the steady-state measures on phase space. In the case of the DDS, this relation is essentially the same as one proposed some time ago by Price [21] which relates the covariance of electric current noise in semiconductors and plasmas to the symmetric part of the bulk charge diffusion matrix. (See also [48], Sections 3.2 and 3.5, for discussions of the experimental validity and usefulness of this relation in semiconductors.) The KLS derivation with the correlation function method has given this relation an exact microscopic basis.

¿From the discussion it is clear that the covariance of the “fast” component of microscopic currents should be identified with the covariance of noisy currents in the Langevin equation, Eq.(3.44). Therefore, we have now a complete set of prescriptions to calculate, in principle, the parameters of that equation from the microscopic dynamics. The drift velocity $\mathbf{c}$ and the (full) diffusion matrix $\mathbf{D}$ are given by the already presented microscopic expressions, $\mathbf{c} = \hat{\jmath}'$ and the Green-Kubo formula Eq.(2.57). The noise covariance $\mathbf{R}$ is then calculated from the FDR of Price's form:

$$\mathbf{R} = \mathbf{D}^s \chi. \tag{3.66}$$

The use of this relation requires that the non-equilibrium entropy $s(n)$ be known, in order to calculate $\chi(n) = -1/s''(n)$. Of course, it is quite generally true that the fluctuation-dissipation relation, Eq.(3.23), allows one to determine the Langevin noise covariance $\mathbf{Q}$ from the steady-state covariance $\mathbf{g}$, assuming that the macroscopic hydrodynamics (and the linearized operator $\mathbf{H}$) is known. When that is the strategy, some other means must be employed to find $\mathbf{g}$, such as the high-temperature series expansion of [30]. Of course, if one's only interest were the steady-state correlations themselves, then it would be an empty exercise to calculate $\mathbf{Q}$. However, knowledge of $\mathbf{Q}$ allows arbitrary multi-time correlations to be obtained. These may be of interest, e.g. to describe inelastic scattering of light by the steady-state charge density fluctuations.

(3.3) Steady-State Correlations and Hydrodynamics

If we compare the Price-type FDR Eq.(3.66) we derived from the correlation function method with the FDR at the Langevin level, Eq.(3.48),

$$\mathbf{k} \cdot \mathbf{R} \cdot \mathbf{k} = (\mathbf{k} \cdot \mathbf{D}^s \mathbf{k}) S(\mathbf{k}), \tag{3.67}$$

then we see that they are consistent if we require that the static covariance in the Langevin formalism be wavenumber independent, as

$$S(\mathbf{k}) = \chi. \tag{3.68}$$

This is known to be generally false! In fact, there is a problem even with the existence of the limit as $k \to 0$. It was observed in [50] from computer simulation of the DLG model that the decay of correlations are actually a slow power-law, like $\sim r^{-d}$. This type of behavior has been corroborated in stochastic lattice dynamics, like the DLG models, by independent calculations using high-temperature series expansions [30]. Also, the existence of the power-laws has been confirmed in more phenomenological Ginsburg-Landau models of DDS by field-theoretic RG calculations [51]. The generic behavior of the correlation in space seems to be

$$C(\mathbf{r}) \sim \frac{\sum_i b_i r_i^2}{r^{d+2}}, \tag{3.69}$$

corresponding to a behavior in wavenumber space

$$S(\mathbf{k}) \sim \frac{\sum_i b_i k_i^2}{k^2}. \tag{3.70}$$

Hence, the wavenumber independence predicted by the naive form of the Price FDR, Eq.(3.66), is contradicted by both simulations and microscopic calculations. This may be the proper point to remark that the simple proportionality proposed by Price is known also to be violated at the kinetic level of description due to correlations created by collisions: see Section 2.2 of [48]. It should be emphasized that there is no inconsistency of the long-range correlations and the Langevin FDR Eq.(3.67). In fact, as was pointed out in [30], the correlation behavior exactly like that in Eq.(3.70) is recovered from the Langevin FDR, Eq.(3.67), when the simple proportionality of $\mathbf{R}$ and $\mathbf{D}^s$ is abandoned. The crucial difference with the naive form of the Price FDR is the appearance of the dot products with $\mathbf{k}$, which allows $S(\mathbf{k})$ to be wavenumber dependent even with $\mathbf{D}, \mathbf{R}$ constant. This appearance of $\mathbf{k}$ is due to the fact that really only the *divergence* $\nabla \cdot \mathbf{J}'$ of the noisy current appears in the fluctuating hydrodynamic equations.

Since the conclusion of the correlation function argument in the previous section seems to be generally false, the question is raised which of the assumptions it employed might be erroneous. In addition to rather plausible assumptions—such as the Onsager regression hypothesis—there was also, as we have already emphasized, a rather strong analyticity assumption on the structure function $S(\mathbf{k}, t)$ at small wavenumber. This assumption is in contradiction with the observed long-range correlations, or their wavenumber version Eq.(3.70), which implies that the limit

$$\chi(n,\hat{\mathbf{k}}) \equiv \lim_{k\to 0} \sum_{\mathbf{x}} e^{i\mathbf{k}\cdot\mathbf{x}} \langle \eta_{\mathbf{x}} \eta_{\mathbf{0}} \rangle_n \tag{3.71}$$

will exist but depend upon the direction vector $\hat{\mathbf{k}}$. In other words, the structure function cannot be expected to be even continuous at $\mathbf{k} = \mathbf{0}$. Nevertheless, it is possible to repeat the KLS correlation function argument under these weaker assumptions and, in that case, exactly the weaker FDR Eq.(3.67) is re-obtained, fully consistent with the Langevin formalism and the microscopic results on long-range correlations. This is done in Appendix VI of this paper. In consequence, the correlation function derivation of the *linearized* hydrodynamics seems to be consistent with all other known results.

However, the derivation of the nonlinear hydrodynamics in Section 2 by the method of nonequilibrium distributions is in worse shape. One of its main assumptions was that there is a *unique* relation between the density n and chemical potential λ, as $\lambda = \lambda(n)$. Integrating the function $\chi^{-1}(n,\hat{\mathbf{k}})$ from Eq.(3.71) with respect to n, one would instead expect to have independent relations $\lambda(n,\hat{\mathbf{k}})$ for each direction vector $\hat{\mathbf{k}}$. The problem shows up already at the level of our fundamental large deviations hypothesis in Section 2.1. Indeed, simple examples show that when the static structure function is discontinuous at zero wavenumber, the large deviations hypothesis must undergo some substantial revision. Even if it remains true, the resulting function $s(n)$ may depend upon the exact way in which the sequences of volumes Λ are taken to infinity! This can already be seen to occur for a Gaussian random field of spins $\{\sigma_{\mathbf{x}} : \mathbf{x} \in \mathbf{Z}^d\}$ when the static spin structure function

$$S(\mathbf{k}) = \sum_{\mathbf{x}} e^{i\mathbf{k}\cdot\mathbf{x}} \langle \sigma_{\mathbf{x}} \sigma_{\mathbf{0}} \rangle \tag{3.72}$$

has the form of Eq.(3.70) (or is otherwise discontinuous at $\mathbf{k} = \mathbf{0}$.) In fact, in that case, the "smooth" version of the volume-average magnetization

$$m_\epsilon(\varphi) \equiv \epsilon^d \sum_{\mathbf{x}} \varphi(\epsilon\mathbf{x}) \sigma_{\mathbf{x}} \tag{3.73}$$

satisfies the LD hypothesis for every square-integrable test-function φ, [4] with the quadratic entropy function

$$s_\varphi(m) = \frac{m^2}{2K_\varphi}, \tag{3.77}$$

in which

$$K_\varphi = \frac{1}{(2\pi)^d} \int d^d\mathbf{k}\ |\hat{\varphi}(\mathbf{k})|^2 \chi(\hat{\mathbf{k}}). \tag{3.78}$$

Because of the $\hat{\mathbf{k}}$ dependence of χ, the result clearly depends upon the precise test function adopted. For instance, if a function $\varphi(\mathbf{x}) = \prod_{i=1}^d \frac{1}{2a_i}\chi_{[-a_i,a_i]}(x_i)$ were chosen, corresponding to the rectangular parallelopiped $[-a_1, a_1] \times \cdots \times [-a_d, a_d]$ scaled to infinity, then the function $s_\varphi(m)$ obtained would depend upon the aspect ratios of the box sides. This may seem an artificial mathematical example, but it is just the Gaussian model of a uniaxial ferromagnet/ferroelectric with dipolar interactions, which has appeared in studies of critical behavior of those systems [52]. It is rather well-known for dipolar systems that the thermodynamics may be shape-dependent (see [23]) and the structure-function singularity in that case has even been experimentally observed [53]. However, it is perhaps less surprising to encounter such phenomena in systems with long-range forces, rather than in the DLG models where all particle-interactions are short-ranged.

Since the nonequilibrium distribution function method used rather crucially the relation $\lambda(n)$ between chemical potential and density to guess the correct modification of the reference

[4]This can be proved most easily by calculating the free energy

$$f_{\varphi,\epsilon}(h) \equiv \epsilon^d \log\langle \exp\left(h\sum_{\mathbf{x}} \varphi(\epsilon\mathbf{x})\sigma_{\mathbf{x}}\right)\rangle, \tag{3.74}$$

to be

$$f_{\varphi,\epsilon}(h) = \frac{h^2}{2}\cdot\frac{1}{(2\pi)^d}\int_{[-\frac{\pi}{\epsilon},\frac{\pi}{\epsilon}]} d^d\mathbf{k}\ |\hat{\varphi}(\mathbf{k})|^2 S(\epsilon\mathbf{k}). \tag{3.75}$$

Since its limit exists

$$\begin{aligned} f_\varphi(h) &\equiv \lim_{\epsilon\to 0} f_{\varphi,\epsilon}(h) \\ &= \frac{1}{2}K_\varphi h^2, \end{aligned} \tag{3.76}$$

and is differentiable for all real h, Theorem II.6.1 of Ellis [32] applies.

measure to produce a given smooth density profile, it is unclear to us how to proceed when that relation breaks down. Therefore, it is not clear to us that the nonlinear drift-diffusion equation of the form of Eq.(2.61) even still holds in the general case! It should be stressed that the derivation of hydrodynamics in Section 2 was based upon the explicit assumption of fast decay of correlations, and its validity is not questioned in such cases. Situations with such "fast decay" do exist, such as the ASEP discussed in Appendix III, whose stationary measures are completely uncorrelated (product measures), and, more generally, DLG models of "gradient type" (see [14], Appendix 4). These even include cases where the measures are not Gibbsian [54, 55]. However, it is rather disconcerting that such situations seem, according to present evidence, to be *non-generic* and, in general, the long-range correlations and discontinuous susceptibility at zero-wavenumber must be expected. On the other hand, we know of no experimental evidence for these phenomena in physical examples of DDS, although they should, in principle, show up clearly in electromagnetic scattering from the homogeneous steady state. [5] We regard the reconciliation of the long-range correlations and the nonlinear hydrodynamics to be one of the main outstanding theoretical puzzles in the driven-diffusive systems.

4 Large Hydrodynamic Fluctuations

(4.1) A Generalized FDR for the Nonlinear Fokker-Planck Equation

In the previous section we considered the problem of constructing the proper Langevin model of the small ($O(\epsilon^{d/2})$) fluctuations about the hydrodynamic behavior governed by the driven diffusion equation Eq.(2.61). However, there are also very rare large fluctuations of

[5] Note that the k^{-4} Rayleigh peaks which have been observed [56] in the steady-states with temperature gradients have a different character. These latter systems are local equilibrium states and the physical space correlation is there $\sim r^{-(d-2)}$ but proportional to ϵ^2 (or the square of the temperature gradient.) Thus, while longer-range than those considered above, the correlations in those states are *weaker,* locally vanishing as $\epsilon \to 0$. It seems to be harder to realize the periodic geometry of the DDS than the steady states driven by boundary conditions, making experimental study difficult.

order O(1). For the description of these there are a number of formalisms in the literature: for example, nonlinear Fokker-Planck and Langevin equations, or nonlinear Onsager-Machlup action functionals. For a discussion of each of these and their interrelations see [7, 37, 57] and references therein. What we shall investigate here is the possibility of applying such methods also to the systems without local equilibrium, focusing on our main example of the DDS models. In general, the same types of dynamical and statistical assumptions enter into the derivation of the fluctuation theory as into the derivation of the hydrodynamical equations themselves. Therefore, we believe that the "level-2" nonequilibrium distribution method used by Zubarev and Morozov in [7] could be also successfully applied to such systems (at least in the "fast-decay case" discussed in the last section).

However, rather than carry through such a microscopic derivation here, we would like to point out a quicker route to the final result, which may be more generally useful. This method exploits the fact that hydrodynamical equations derived by the previous procedure are automatically in the "Onsager force-flux form." In that case it is possible to invoke a general fluctuation-dissipation relation of first type for nonlinear Fokker-Planck equations, which allows one to write down directly the fluctuation theory by inspection of the hydrodynamic equations without further calculation. It was R.Graham in his paper [24] who formulated the FDR's we find useful in this context and who, particularly, emphasized their validity without any restriction of time-reversibility, even at the macroscopic level. (We should point out that other nonlinear FDR's of a different character and with their own specific applications are discussed by Stratonovich in his monograph [42].) It may be that this work has not received sufficient attention because of its abstract style and its formal differential-geometric language. Therefore, we would like here to illustrate its usefulness in the very concrete setting of electrical current-carrying systems, where it will be shown to give a nonlinear generalization of Price's noise-diffusion relation. Furthermore, we shall present a simplified version of Graham's relation, which turns out to avoid the geometric complexities of his original work but to be perfectly adapted for application to spatially-extended, macroscopic systems. The rest of this subsection will be devoted to the first task of explaining the generalized nonlinear FDR's of Graham in our simplified version and then comparing them with his more general formulation.

The most arbitrary Fokker-Planck equation in the p variables $\{\alpha^i|i=1,...,p\}$ may be written in the form

$$\frac{\partial P}{\partial t}(\boldsymbol{\alpha},t) = -\frac{\partial}{\partial\alpha^i}\left[J^i(\boldsymbol{\alpha})P(\boldsymbol{\alpha},t)\right] + \frac{\partial^2}{\partial\alpha^i\partial\alpha^j}\left[Q^{ij}(\boldsymbol{\alpha})P(\boldsymbol{\alpha},t)\right], \tag{4.1}$$

where the sum over repeated indices always goes from 1 to p and we have chosen notations in close accord with our discussion of linear Langevin equations in section 3.1. The Fokker-Planck equation itself corresponds to the Ito stochastic differential equation

$$\dot{\alpha}^i = J^i(\boldsymbol{\alpha}) + g^i_\mu(\alpha)\eta^\mu, \tag{4.2}$$

in which $\eta^\mu(t)$ are white-noise forces with zero mean and covariance

$$\langle\eta^\mu(t)\eta^\nu(t')\rangle = 2\delta^{\mu\nu}\delta(t-t'), \tag{4.3}$$

and the functions $g^i_\mu(\boldsymbol{\alpha})$ are a set of multiplicative noise-strengths such that

$$Q^{ij}(\boldsymbol{\alpha}) = g^i_\mu(\boldsymbol{\alpha})g^j_\mu(\boldsymbol{\alpha}). \tag{4.4}$$

(Throughout this section we take $k_B \equiv 1$ for simplicity). All of the analysis we shall make here depends upon a crucial assumption on the Fokker-Planck diffusion matrix $\mathbf{Q}(\boldsymbol{\alpha})$ [6], namely, that it is "divergence-free" in the sense that

$$\frac{\partial Q^{ij}(\boldsymbol{\alpha})}{\partial\alpha^j} = 0. \tag{4.5}$$

As shown in the original paper of Graham [24], and as we shall discuss further below, results similar to those obtained here may be obtained without this assumption. However, although it is a rather special restriction in the class of general Fokker-Planck equations, we shall show

[6] Note that the Fokker-Planck equation is itself a driven diffusion equation, but describing flow of probability in the state space of the $\boldsymbol{\alpha}$'s rather than of particle number in physical space! No confusion should result when we use the standard terminology of "diffusion" for $\mathbf{Q}$ and "drift" for $\mathbf{J}$, qualified, if necessary, by the adjective "Fokker-Planck."

that the spatially-extended, conservative systems always satisfy this assumption automatically and that it provides a suitable basis for our applications.

Under this single assumption, Eq.(4.5), we now show that the systematic part of the Langevin dynamics, the Fokker-Planck drift vector J^i, may be decomposed into a "conservative" part r^i and a "dissipative" part d^i,

$$J^i(\boldsymbol{\alpha}) = r^i(\boldsymbol{\alpha}) + d^i(\boldsymbol{\alpha}), \tag{4.6}$$

in a way quite analogous to our decomposition for the linear Langevin dynamics. (Of course, corresponding to different choices of $\mathbf{Q}$ there will be different decompositions of $\mathbf{J}$!) As in the linear case, making the decomposition requires knowledge of the stationary measure $P_0(\boldsymbol{\alpha})$, or of the corresponding "entropy" defined by

$$S(\boldsymbol{\alpha}) \equiv \log P_0(\boldsymbol{\alpha}). \tag{4.7}$$

The key point of the argument is to use the "divergence-free" assumption Eq.(4.5) to rewrite the Fokker-Planck equation as

$$\frac{\partial P}{\partial t}(\boldsymbol{\alpha}, t) = -\frac{\partial}{\partial \alpha^i}\left[J^i(\boldsymbol{\alpha})P(\boldsymbol{\alpha}, t) - Q^{ij}(\boldsymbol{\alpha})\frac{\partial}{\partial \alpha^j}P(\boldsymbol{\alpha}, t)\right]. \tag{4.8}$$

We can then *define*

$$d^i(\boldsymbol{\alpha}) \equiv Q^{ij}(\boldsymbol{\alpha})\frac{\partial S(\alpha)}{\partial \alpha^j}, \tag{4.9}$$

and

$$r^i(\alpha) \equiv J^i(\boldsymbol{\alpha}) - d^i(\boldsymbol{\alpha}). \tag{4.10}$$

Substituting $P = e^S$ into the Eq.(4.8), we find that the $\mathbf{Q}$ diffusion term cancels with the $\mathbf{d}$ part of the drift, giving

$$\frac{\partial}{\partial \alpha^i}\left[r^i(\boldsymbol{\alpha})e^{S(\boldsymbol{\alpha})}\right] = 0. \tag{4.11}$$

This expresses the stationarity of $P_0 = e^S$ under the deterministic evolution by $\mathbf{r}$ alone.

We can further verify that the above decomposition corresponds to one into "conservative" and "dissipative" parts, and, indeed, puts $\mathbf{J}$ into the "Onsager form" with respect to the potential S. To see this, we identify $L_s^{ij} \equiv Q^{ij}$, which gives

$$d^i(\boldsymbol{\alpha}) = -L_s^{ij}(\boldsymbol{\alpha})X_j(\boldsymbol{\alpha}), \tag{4.12}$$

in terms of the "force"

$$X_i(\boldsymbol{\alpha}) \equiv -\frac{\partial S(\boldsymbol{\alpha})}{\partial \alpha^i}. \tag{4.13}$$

Furthermore, following an idea of Graham [24], we represent the divergence-free vector $r^i e^S$ as

$$r^i(\boldsymbol{\alpha})e^{S(\boldsymbol{\alpha})} \equiv \frac{\partial F^{ij}(\boldsymbol{\alpha})}{\partial \alpha^j}, \tag{4.14}$$

with F^{ij} antisymmetric, so that

$$r^i(\boldsymbol{\alpha}) = L_a^{ij}(\boldsymbol{\alpha})\frac{\partial S(\boldsymbol{\alpha})}{\partial \alpha^j} + \frac{\partial L_a^{ij}(\boldsymbol{\alpha})}{\partial \alpha^j}, \tag{4.15}$$

in terms of a new antisymmetric matrix $L_a^{ij} \equiv F^{ij}e^{-S}$. Together Eq.(4.12) and Eq.(4.15) give the representation of the full drift

$$J^i(\boldsymbol{\alpha}) = -L^{ij}(\boldsymbol{\alpha})X_j(\boldsymbol{\alpha}) + \frac{\partial L_a^{ij}}{\partial \alpha^j}, \tag{4.16}$$

with

$$L^{ij}(\boldsymbol{\alpha}) \equiv L_s^{ij}(\boldsymbol{\alpha}) + L_a^{ij}(\boldsymbol{\alpha}). \tag{4.17}$$

The latter is now naturally interpreted as the "Onsager coefficient" and Eq.(4.16) is in the usual "force-flux" form except for the additional divergence term $\partial L_a^{ij}/\partial \alpha^j$. In fact, for the applications we shall consider it will be generally true also that

$$\frac{\partial L_a^{ij}(\boldsymbol{\alpha})}{\partial \alpha^j} = 0, \tag{4.18}$$

for the same reasons which imply the divergencelessness of L_s^{ij} (see below). In that case, the standard Onsager form is obtained. Furthermore, it follows under the condition Eq.(4.18) that the $\mathbf{r}$ part of the drift enjoys the Liouville property, or divergence-free condition

$$\frac{\partial r^i(\boldsymbol{\alpha})}{\partial \alpha^i} = 0, \tag{4.19}$$

by a direct calculation. Therefore, $\mathbf{r}$ is indeed "conservative" in the sense that

$$\frac{\partial S(\boldsymbol{\alpha})}{\partial \alpha^i} r^i(\boldsymbol{\alpha}) = 0, \tag{4.20}$$

which follows from the combination of Eqs.(4.11) and (4.19). In that case, the entire change of S under the systematic evolution is due to $\mathbf{d}$, which gives

$$\begin{aligned} \frac{d}{dt} S(\boldsymbol{\alpha}) &= \frac{\partial S(\boldsymbol{\alpha})}{\partial \alpha^i} d^i(\boldsymbol{\alpha}) \\ &= L_s^{ij}(\boldsymbol{\alpha}) X_i(\boldsymbol{\alpha}) X_j(\boldsymbol{\alpha}) \\ &\geq 0. \end{aligned} \tag{4.21}$$

Therefore, $\mathbf{d}$ is truly the "dissipative" part.

¿From these considerations we can see that the identification

$$L_s^{ij}(\boldsymbol{\alpha}) \equiv Q^{ij}(\boldsymbol{\alpha}), \tag{4.22}$$

is in fact a nonlinear extension of the *FDR of first type*, of the sort established in the linear Langevin case in Section 3.1. It was first obtained (in a more general setting) by Graham in [24]. Before discussing his results, let us reformulate the relation in a way directly useful to us below. The starting point above was the Fokker-Planck equation itself. However, it is instead our goal to deduce that equation, having in hand the microscopic derivation of the hydrodynamic law in the Onsager form. Since the systematic part of the Langevin dynamics must be the deterministic hydrodynamics itself, we would like to have a way to guess the proper choice of the noise to add to it. For that purpose, we may reverse the previous considerations to derive a result expressed formally in the following

Proposition 1 *Consider the Fokker-Planck Eq.(4.1) with the divergence-free condition* $\partial Q^{ij}/\partial \alpha^j = 0$. *If* $J^i = r^i + d^i$ *with* $\frac{\partial}{\partial \alpha^i}\left[r^i e^S\right] = 0$ *and* $d^i = L_s^{ij}\frac{\partial S}{\partial \alpha^j}$, *then* $Q^{ij} = L_s^{ij}$ *implies that* $P_o = e^S$ *is stationary. Furthermore, when also* $\partial L_s^{ij}/\partial \alpha^j = 0$, *then* $P_0 = e^S$ *stationary and non-degenerate implies conversely that* $Q^{ij} = L_s^{ij}$.

Here "non-degeneracy" of P_0 just signifies that $\frac{\partial^2 P_0}{\partial\alpha^i\partial\alpha^j}$ is a nonsingular matrix for almost all $\boldsymbol{\alpha}$ in the state space. We leave the elementary proof of this proposition to the reader. We note only that its statement specifies—under the expressed conditions—the *unique* choice of the noise strength to obtain the stationary measure $P_0 = e^S$.

In his original work [24], Graham obtained the equivalent decomposition as that above in the setting of the most general Fokker-Planck equation, without the "divergence-free" condition Eq.(4.5). His strategy was to redefine the notion of "derivative" in order to make the condition always true! In fact, it is possible to take the Fokker-Planck diffusion $Q^{ij}(\boldsymbol{\alpha})$ as a contravariant "metric" tensor defining a Riemannian geometric structure in the state space of the $\boldsymbol{\alpha}$'s. In that case, one can take the associated definition of "connection" and "covariant derivative" so that the metric tensor itself has vanishing covariant derivative:

$$Q^{ij}_{;k} = 0, \tag{4.23}$$

expressing that "lengths" are invariant under the "parallel transport" in state space. (We follow the standard device of denoting the covariant derivative $D/D\alpha^k$ by $(\cdot)_{;k}$.) This stronger condition implies at once the zero covariant-divergence result $Q^{ij}_{;j} = 0$. In that case, it is not hard to put the Fokker-Planck equation into a completely covariant version

$$\partial_t P = -\left(J^i P - Q^{ij} P_{;j}\right)_{;i}, \tag{4.24}$$

identical in structure to Eq.(4.8) above. Note here that P and J^i *are not the same as those above*, but correspond also to covariant versions.

Having arrived at the equation (4.24), Graham then derived the same decomposition as ours above, with exactly the same argument, simply replacing ordinary derivatives with covariant derivatives everywhere. Note that Graham's decomposition is the full nonlinear generalization of that introduced by Tomita and Tomita [47] for linear Langevin dynamics. While Graham did not explicitly state that the identity between Q^{ij} and L^{ij}_s was a type of FDR nor even mention the relation to "Onsager form," he did make a special emphasis of the fact that the

FDR of second type extended to this general Fokker-Planck context without, in particular, any assumptions on time-reversibility. This required a special coupling of the external force, constructed according to the principle that the stationary measure in the presence of forces F_i should change by the exponential modification $\propto \exp[F_i\alpha^i]$ (see also [42], Section 5.5.1). The correct way to achieve this is by introducing the force into the Onsager form of J^i, as

$$J^i = -L^{ij}(X_j - F_j) + (L_a^{ij})_{;j}. \tag{4.25}$$

If one defines as usual the response function $G^{ij}(t) = \langle\frac{\delta\alpha^i(t)}{\delta F_j(0)}\rangle$ to this imposed force, then Graham showed that

$$\langle\dot{\alpha}^i(t)\alpha^j(0)\rangle = -\left[G^{ij}(t) - G^{ji}(-t)\right], \tag{4.26}$$

just as the usual FDR of second type. (In fact, we can obtain the same result also in our restricted context.) Observe that it is necessary in general to know the stationary meaure P_0 in order to make the appropriate coupling of F_i, since the decomposition of J^i into $r^i + d^i$ used S. As Graham observed, the only role of time-reversibility —when it is present—is that it allows this decomposition to be made *a priori* on the basis that r^i is odd and d^i even under time-reversal. These statements are equivalent to the well-known "potential conditions" of Graham and Haken for detailed balance [58], and they imply that the Onsager matrix $L^{ij}(\boldsymbol{\alpha})$ obeys the Onsager reciprocal relations $\epsilon_i\epsilon_j L^{ij}(\epsilon\cdot\boldsymbol{\alpha}) = L^{ji}(\boldsymbol{\alpha})$. [7]

(4.2) Fluctuating Equations for the Driven Diffusion Model

We now turn to the problem of constructing the nonlinear fluctuation theory corresponding to the drift-diffusion equation Eq.(2.61). As discussed above, we make no attempt here to derive it microscopically but simply assume it to exist and then use the previous results to restrict its form. To be more precise, we make the following:

Hypothesis 2 *There exists a Langevin description of the nonlinear fluctuations of the form*

$$\partial_\tau n = -\partial_m[\hat{\jmath}_m(n) - \epsilon D_{ml}(n)\partial_l n + \epsilon^{d/2} j'_m], \tag{4.27}$$

[7] Without the reversibility assumption, rather formal "generalized" OR and potential conditions can be formulated also in the nonlinear Fokker-Planck context. E.g. see Section VI.B of [40].

with the stationary measure

$$P_o^\epsilon[n] \propto \exp\left\{\epsilon^{-d} S[n]\right\}. \tag{4.28}$$

Let us say a few words to justify some particular aspects of the hypothesis. First, it is essential that the equation preserve the feature of local particle conservation, since that is a fundamental property of the microscopic dynamics. Therefore, the Langevin force ought to be added as an additional "noisy current" into the driven-diffusion equation (which must be recovered in the $\epsilon \to 0$ limit.) The basic assumption is that the correct stationary measure of the Langevin dynamics is that given in Eq.(4.28). It seems reasonable from the way in which $s(n)$ was defined in terms of the fluctuation probabilities for density n in the stationary states of the microscopic dynamics. Since $S[n] = \int_\Lambda d^d\mathbf{r}\ s(n(\mathbf{r}))$, the main assumption involved here is a "locality property" of static fluctuations in the extended system, which ought at least to be valid in the "fast-decay" case. From a physical point of view, the hydrodynamical density which appears in these equations is supposed to represent "coarse-grained values" obtained by averaging over "mesoscopic" cells of size intermediate between the scale of the particles and the system size. [8] For such variables there will be simultaneously nonlinear effects of the dynamics and strong stochastic influence of the molecular noise. Because of the coarse-graining procedure these hydrodynamic fields should contain no modes smaller than the cell-size, and it is simplest to represent them mathematically by Fourier series truncated at a wavenumber k_0 the inverse of the cell-diameter ℓ_0, as we do here, or alternatively by discretization on a lattice with grid-unit ℓ_0. Since the theory of stochastic PDE's is difficult, it is desirable even from a purely mathematical point of view to define the nonlinear Langevin dynamics instead as a stochastic ODE for a finite set of variables (Fourier modes or lattice field-variables). For a further discussion of the microscopic derivation of such equations by "coarse-graining" see Zubarev and Morozov, Section 2.1 of [7]. We shall also comment further at the end of this section upon the physical uses (and limitations) of our nonlinear fluctuation equations for the

[8] More formally, the cells are of linear dimension $\ell_0 \sim \epsilon^\gamma$ with some $0 < \gamma < 1$ in the limit $\epsilon \to 0$, with the macroscopic domain Λ fixed.

DDS.

The basic problem is to guess the form of the noisy currents in Eq.(4.27). We shall first present here the result, and afterward give its justification. The Langevin currents are assumed in the particular form

$$j'_m(\mathbf{r},\tau) = \sqrt{\epsilon} g_m^\sigma(n(\mathbf{r},\tau))\eta_\sigma(\mathbf{r},\tau), \tag{4.29}$$

$\sigma = 1, ..., d$, where η is a spacetime white-noise with covariance

$$\langle \eta_\sigma(\mathbf{r},\tau)\eta_\rho(\mathbf{r}',\tau')\rangle = 2\delta_{\sigma\rho}\delta^d(\mathbf{r}-\mathbf{r}')\delta(\tau-\tau'). \tag{4.30}$$

(Observe that $\delta(\mathbf{r}-\mathbf{r}')$ here denotes a "coarse-grained" delta functional, or $\Delta(\mathbf{r}-\mathbf{r}')$ in the notation of Zubarev and Morozov [7].) This choice of the noisy currents is not entirely necessary but does have some convenient features. For example, the discussion of Morozov in [57] implies that for this form the Ito and Stratonovich interpretations coincide. This allows us to use ordinary rules of calculus. With this choice of noise, our proposed Langevin equation then leads to the Fokker-Planck equation

$$\begin{aligned} \frac{\partial}{\partial\tau}P(n,\tau) &= -\int d^d\mathbf{r}\ \frac{\delta}{\delta n(\mathbf{r})}J(\mathbf{r};n)P(n,\tau) \\ &\quad +\epsilon^{d+1}\int d^d\mathbf{r}\int d^d\mathbf{r}'\ \frac{\delta^2}{\delta n(\mathbf{r})\delta n(\mathbf{r}')}Q(\mathbf{r},\mathbf{r}';n)P(n,\tau), \end{aligned} \tag{4.31}$$

where

$$J(\mathbf{r};n) = -\partial_m[\bar{\jmath}_m(n(\mathbf{r})) - \epsilon D_{ml}(n(\mathbf{r}))\partial_l n(\mathbf{r})], \tag{4.32}$$

and

$$Q(\mathbf{r},\mathbf{r}';n) = -\partial_m[Q_{ml}(n(\mathbf{r}))\partial_l\delta(\mathbf{r}-\mathbf{r}')], \tag{4.33}$$

with

$$Q_{ml}(n) = g_m^\sigma(n)g_l^\sigma(n). \tag{4.34}$$

Any other choice of the Langevin equation which leads to the same Fokker-Planck equation is stochastically equivalent and would suffice just as well. For example, other square roots of the operator Q could be employed in the noise term, such as the positive one (e.g. see Section 6 of

[37]), but it is more convenient to choose the local form above. The essential part of our choice is that the matrix $\mathbf{Q}$ must be taken as

$$Q_{lm}(n) = L^s_{lm}(n), \tag{4.35}$$

with $\mathbf{L}^s$ the Onsager matrix calculated in Section 2. This is given, for example, as $\chi(n)$ times $\mathbf{D}^s$ written as the Green-Kubo formula in Eq.(144). Therefore, the $\{g^\sigma_m\}$ are introduced here as an orthogonal set of eigenvectors of $\mathbf{Q}$ (normalized to have length given by the square root of the corresponding eigenvalues.)

The motivation for this prescription is that the systematic part of the equation, the driven diffusion dynamics, may be written in the form

$$J(\mathbf{r};n) = r(\mathbf{r};n) + d(\mathbf{r};n), \tag{4.36}$$

with

$$r(\mathbf{r};n) = -\mathbf{c}(n)\cdot\boldsymbol{\nabla}_{\mathbf{r}}n + \epsilon\int d^d\mathbf{r}'\ L_a(\mathbf{r},\mathbf{r}';n)\frac{\delta S(n)}{\delta n(\mathbf{r}')}, \tag{4.37}$$

and

$$d(\mathbf{r};n) = \epsilon\int d^d\mathbf{r}'\ L_s(\mathbf{r},\mathbf{r}';n)\frac{\delta S(n)}{\delta n(\mathbf{r}')}, \tag{4.38}$$

with

$$L(\mathbf{r},\mathbf{r}';n) = -\partial_m[L_{ml}(n(\mathbf{r}))\partial_l\delta(\mathbf{r}-\mathbf{r}')]. \tag{4.39}$$

(Actually, as already observed, the $\mathbf{L}_a$ term in (4.37) vanishes for the one-component system, but we shall not make use of this fact.) We shall show that the r term leaves invariant the proposed stationary measure:

$$\int d^d\mathbf{r}\ \frac{\delta}{\delta n(\mathbf{r})}\left(r(\mathbf{r};n)e^{\epsilon^{-d}S(n)}\right) = 0. \tag{4.40}$$

Furthermore, we shall show that the fundamental property of the noise covariance and the Onsager matrix (kernel) are satisfied:

$$\int d^d\mathbf{r}'\ \frac{\delta L(\mathbf{r},\mathbf{r}';n)}{\delta n(\mathbf{r}')} = \int d^d\mathbf{r}'\ \frac{\delta Q(\mathbf{r},\mathbf{r}';n)}{\delta n(\mathbf{r}')} = 0. \tag{4.41}$$

¿From these facts we can infer by our previous proposition that the chosen form of the noise will lead uniquely to our desired stationary measure. It will turn out, incidentally, that the factor ϵ^{-d} in $\exp\left(\epsilon^{-d}S\right)$ is fixed only by the requirement that

$$d(\mathbf{r};n) = \epsilon^{d+1}\int d^d\mathbf{r}'\ Q(\mathbf{r},\mathbf{r}';n)\cdot\epsilon^{-d}\frac{\delta S(n)}{\delta n(\mathbf{r}')}, \tag{4.42}$$

while all the other terms satisfy invariance with an arbitrary factor.

The fundamental property of the Onsager matrix, Eq.(4.41), was already demonstrated in the work of Zubarev and Morozov, Appendix E of [7]. In fact, it is a general property of kernels of the form in Eqs.(4.33),(4.39). For completeness, we will repeat the proof here. The argument uses the fact that the UV regularization procedure is space-reflection symmetric. By explicit calculation,

$$\frac{\delta L(\mathbf{r},\mathbf{r}';n)}{\delta n(\mathbf{r}'')} = -\partial_m[L'_{ml}(n(\mathbf{r}))\delta(\mathbf{r}-\mathbf{r}'')\partial_l\delta(\mathbf{r}-\mathbf{r}')]. \tag{4.43}$$

Setting $\mathbf{r}''=\mathbf{r}'$ in this expression gives then

$$\frac{\delta L(\mathbf{r},\mathbf{r}';n)}{\delta n(\mathbf{r}')} = -\partial_m[L'_{ml}(n(\mathbf{r}))\delta(\mathbf{r}-\mathbf{r}')]\cdot\partial_l\delta(\mathbf{0}). \tag{4.44}$$

However,

$$\begin{aligned}\boldsymbol{\nabla}\delta^d(\mathbf{0}) &= \frac{1}{|\Lambda|}\sum_{k<k_0}\mathbf{k}\\ &= \mathbf{0},\end{aligned} \tag{4.45}$$

so that the result follows. Notice that the property is basically a consequence of the locality of the hydrodynamical equation and the gradient constitutive relation. The same argument applies to Q and to general Onsager kernels for spatially-extended, conservative systems.

To establish the invariance condition Eq.(4.40) it is desirable to decompose $r = r_c + r_a$ where

$$r_c(\mathbf{r};n) = -\mathbf{c}(n)\cdot\boldsymbol{\nabla}_{\mathbf{r}}n, \tag{4.46}$$

and

$$r_a(\mathbf{r};n) = \epsilon\int d^d\mathbf{r}'\ L_a(\mathbf{r},\mathbf{r}';n)\frac{\delta S(n)}{\delta n(\mathbf{r}')}. \tag{4.47}$$

We show the invariance separately for each term. The conservation property

$$\int d^d\mathbf{r}\ \frac{\delta S(n)}{\delta n(\mathbf{r})} r_c(\mathbf{r};n) = 0, \tag{4.48}$$

was already shown in Section 2: see Eq.(2.55) there. Likewise, we can check that r_c satisfies the Liouville property

$$\int d^d\mathbf{r} \frac{\delta r_c(\mathbf{r};n)}{\delta n(\mathbf{r})} = 0. \tag{4.49}$$

when it is defined with a UV regularization by high-wavenumber cutoff. In fact, the argument applies to a general "Euler term" of the type of a local conservation-law:

$$r^\alpha(\mathbf{r};\rho) = -\boldsymbol{\nabla}\cdot\mathbf{j}^\alpha(\rho(\mathbf{r})). \tag{4.50}$$

It is then easy to see that

$$\sum_\alpha \frac{\delta r^\alpha(\mathbf{r})}{\delta\rho^\alpha(\mathbf{r})} = -\sum_\alpha \frac{\partial \mathbf{j}^\alpha}{\partial \rho^\alpha}(\rho(\mathbf{r}))\cdot\boldsymbol{\nabla}\delta^d(\mathbf{0}). \tag{4.51}$$

Since $\boldsymbol{\nabla}\delta^d(\mathbf{0}) = \mathbf{0}$ the Liouville property follows. The Liouville property Eq.(4.49) and the conservation property Eq.(4.48) together imply the invariance condition Eq.(4.40) for r_c. Finally, we note that

$$r_a(\mathbf{r};n) = \epsilon\int d^d\mathbf{r}'\ L_a(\mathbf{r},\mathbf{r}';n)\frac{\delta S(n)}{\delta n(\mathbf{r}')} + \epsilon^{d+1}\int d^d\mathbf{r}'\ \frac{\delta L_a(\mathbf{r},\mathbf{r}';n)}{\delta n(\mathbf{r}')}, \tag{4.52}$$

since the added term is zero by the property Eq.(4.41) of the Onsager kernel. Therefore, r_a satisfies also the invariance condition Eq.(4.40) since this last expression has exactly the general form Eq.(4.15) of a term with that property. This completes our verification of the conditions of the Proposition 1 for the DDS.

We may write out explicitly the final form of our conjectured Langevin equation, as

$$\partial_\tau n = -\partial_m[\bar{\jmath}_m(n) - \epsilon D_{ml}(n)\partial_l n + \epsilon^{(d+1)/2} g_m^\sigma(n)\eta_\sigma]. \tag{4.53}$$

As in the case of nonlinear fluctuations at thermal equilibrium studied by Zubarev and Morozov [7, 57], state-dependent or "multiplicative" noise is required to produce the correct stationary measure. The sense of validity of the equation should be that it yields asymptotically correct predictions for fluctuations of the hydrodynamic variables in the limit $\epsilon \to 0$, even for those which are $O(1)$, or macroscopic in size. It also contains the information on small fluctuations $O(\epsilon^{d/2})$. Indeed, if we consider its linearized form for solutions

$$n(\mathbf{r}, \tau) = n^* + \epsilon^{d/2}\xi(\mathbf{r}, \tau), \tag{4.54}$$

then we recover the linear Langevin equation, Eq.(3.44), proposed in subsection 3.2. In particular, the noise correlations in that case obey the (naive) Price relation Eq.(3.66), which the nonlinear FDR, when applied to the DDS, generalizes to large fluctuations.

Note, however, that Eq.(4.53) does *not* apply near the critical point of the DLG model, even approaching the transition from the high temperature side. The reason is that it is valid in the limit $\epsilon \to 0$ *with all other length-scales fixed.* This means that it can be applied near the critical point but only so long as the inequality $L_\triangle \gg \xi$ is satisfied by the macroscopic gradient-length $L_\triangle$ (over which hydrodynamic profiles sensibly change) and the correlation length ξ [9]. Therefore, Eq.(4.27) is not adequate to calculate critical scaling properties of the DDS, and should be distinguished from the "mesoscopic Langevin equation" discussed by Schmittmann and Zia [15], which is more in the spirit of the "time-dependent Ginzburg-Landau equations" of critical dynamics. However, the present equation will be adequate to account for the large fluctuation behavior in the high temperature regime. We now turn to this latter topic.

(4.3) Onsager-Machlup Lagrangian and Least Excess Dissipation

We now wish to consider, briefly, the theory of *hydrodynamic large deviations* for the DDS. This topic is reviewed in a general way by one of us (G.E.) [37], where the subject was developed for local equilibrium systems on the basis of reasonable hypotheses, motivated by rigorous results [59] and prior physical theories [36, 60]. In fact, it was conjectured at the very end of that work that the basic results should extend also to electrical-conducting systems. Here we shall make concrete that proposal.

[9]For the definition of the correlation length in this context see Section 2.2 of [15].

The basic goal is to deduce a formula for the probability of occurence of a "density-history" $\{n(\mathbf{r},\tau)\}$ *other than the solution of the hydrodynamic equation.* Of course, this probability is exceedingly small. The "large deviations" formula quantifies this statement. We shall just sketch the main lines of the derivation and refer mostly to the literature for details. The starting point is our proposed Fokker-Planck equation Eq.(4.31). This equation may be solved for the transition probability in the form of a path-integral, as

$$P(n,\tau|n_0)=\int_{\{n(0)=n_0,n(\tau)=n\}}\mathcal{D}n\ \exp\left[\frac{-1}{\epsilon^{d+1}}\int_0^\tau d\sigma\ L(n(\sigma),\dot{n}(\sigma))\right], \tag{4.55}$$

where the integration is over all histories with the specified initial and final conditions. The functional in the exponent, the so-called *Onsager-Machlup Lagrangian*, is of the form

$$\begin{aligned} L(n,\dot{n}) \;=\; & \frac{1}{4}\int_\Lambda d^d\mathbf{r}\int_\Lambda d^d\mathbf{r}' G(\mathbf{r},\mathbf{r}';n) \\ & \times\left[\dot{n}(\mathbf{r})+\boldsymbol{\nabla}\cdot\hat{\mathbf{j}}(n(\mathbf{r}))-\epsilon\boldsymbol{\nabla}\cdot(\mathbf{L}(n(\mathbf{r}))\cdot\boldsymbol{\nabla}\lambda(\mathbf{r}))\right] \\ & \times\left[\dot{n}(\mathbf{r}')+\boldsymbol{\nabla}'\cdot\hat{\mathbf{j}}(n(\mathbf{r}'))-\epsilon\boldsymbol{\nabla}'\cdot(\mathbf{L}(n(\mathbf{r}'))\cdot\boldsymbol{\nabla}'\lambda(\mathbf{r}'))\right] \\ & +O(\epsilon^{d+1}), \end{aligned} \tag{4.56}$$

evaluated just to leading order. $G(\mathbf{r},\mathbf{r}';n)$ is the kernel of the Green's operator, which is the inverse of the elliptic operator with the Onsager kernel $L(\mathbf{r},\mathbf{r}';n)$,

$$-\boldsymbol{\nabla}\cdot\left[\mathbf{L}(n(\mathbf{r}))\cdot\boldsymbol{\nabla}G(\mathbf{r},\mathbf{r}';n)\right]=\delta^d(\mathbf{r}-\mathbf{r}'), \tag{4.57}$$

specified here with periodic b.c. on the domain Λ. See papers of Graham [25, 61].

Particular sets of histories (cylinders) may be defined by a sequence of values at consecutive times: $n(\tau_1)=n_1,...,n(\tau_p)=n_p$. By the Markov property, their probability is given in the steady-state as

$$P\left(\{n(\tau_1)=n_1,...,n(\tau_p)=n_p\}\right)=\int_{\{n(\tau_1)=n_1,...,n(\tau_p)=n_p\}}\mathcal{D}n\ \exp\left[\frac{-1}{\epsilon^{d+1}}\int_{-\infty}^{+\infty}d\tau\ L(n(\tau),\dot{n}(\tau))\right]. \tag{4.58}$$

In the limit $\epsilon\to 0$ this integral may be evaluated by steepest descent, yielding

$$P\left(\{n(\tau_1)=n_1,...,n(\tau_p)=n_p\}\right)\sim\exp\left[-\epsilon^{-(d+1)}\min_{\{n(\tau_1)=n_1,...,n(\tau_p)=n_p\}}\int_{-\infty}^{+\infty}d\tau\ L(n(\tau),\dot{n}(\tau))\right]. \tag{4.59}$$

The minimization is over all histories satisfying the constraints. The formula, giving an exponentially small probability of the event as $\epsilon \to 0$, is a typical result of large-deviations type. The same result could be obtained by extending formally to our Langevin equation Eq.(4.53) the rigorous theorems of Freidlin and Wentzell for weak stochastic perturbations of ODE's [62]. (Those theorems do not literally apply. Although we deal for each finite ϵ with stochastic ODE's, the number of degrees of freedom also goes to infinity as $\epsilon \to 0$.)

The final result Eq.(4.59) has a simple physical interpretation which can be elucidated in terms of *driven hydrodynamic equations*

$$\partial_\tau n(\mathbf{r},\tau) = -\boldsymbol{\nabla}\cdot\left[\hat{\mathbf{j}}(n(\mathbf{r},\tau)) - \epsilon\mathbf{L}(n(\mathbf{r},\tau))\cdot(\mathbf{F}(\mathbf{r},\tau) + \boldsymbol{\nabla}\lambda(\mathbf{r},\tau))\right], \tag{4.60}$$

with an additional external field of the form

$$\mathbf{F}(\mathbf{r},\tau) = -\boldsymbol{\nabla}U(\mathbf{r},\tau) \tag{4.61}$$

instantaneously for some potential U. Note that this is exactly the same coupling of the external field which would be required for the validity of the FDR of second type. The first observation is that, given a history $\{n(\mathbf{r},\tau)\}$, it is possible to find U as a functional of n and $\dot{n}$ at each instant

$$\begin{aligned} U(\mathbf{r};n,\dot{n}) \quad &= \quad \int_\Lambda d^d\mathbf{r}' G(\mathbf{r},\mathbf{r}';n) \\ &\qquad \times \left[\dot{n}(\mathbf{r}') + \boldsymbol{\nabla}'\cdot\hat{\mathbf{j}}(n(\mathbf{r}')) - \epsilon\boldsymbol{\nabla}'\cdot\left(\mathbf{L}(n(\mathbf{r}'))\cdot\boldsymbol{\nabla}'\lambda(\mathbf{r}')\right)\right] \end{aligned} \tag{4.62}$$

so that $U(\mathbf{r},\tau) = U(\mathbf{r};n(\tau),\dot{n}(\tau))$ defines the *unique* potential field $\{U(\mathbf{r},\tau)\}$, periodic in the domain Λ, required to produce in Eq.(4.60) the desired history $\{n(\mathbf{r},\tau)\}$. It is a simple calculation to show, further, that

$$L(n,\dot{n}) = \frac{1}{4}\int_\Lambda d^d\mathbf{r}\ L_{ml}(n(\mathbf{r}))\partial_m U(\mathbf{r})\partial_l U(\mathbf{r}). \tag{4.63}$$

Thus, the probability formula for history $\{n(\mathbf{r},\tau)\}$ becomes

$$P\left(\{n(\mathbf{r},\tau)\}\right) \sim \exp\left[-\epsilon^{-d}\cdot\frac{\epsilon}{4}\int_{-\infty}^{+\infty} d\tau \int_\Lambda d^d\mathbf{r} L_{ml}(n(\mathbf{r},\tau))F_m(\mathbf{r},\tau)F_l(\mathbf{r},\tau)\right]. \tag{4.64}$$

Observe that two factors of ϵ appeared in the exponent due to each of the field factors $\mathbf{F}$. The final result is quite analogous to the conjecture in [37]. The argument we have outlined, in conjunction with the Zubarev-Morozov derivation of the hydrodynamic Fokker-Planck equation for local equilibrium systems [7], gives, in fact, a "physicist's proof" of our original hypothesis.

Notice that the functional in the exponent of the RHS of (4.64) is one-half of what Onsager called the "dissipation function" $\Psi(\mathbf{F},\mathbf{F})$ [10, 36]. In the present case its physical interpretation is very simple (cf. [20], Section II.3.7). Since we saw in Eq.(2.60) of Section 2 that the Onsager matrix $\mathbf{L}$ is just the "conductivity tensor" associated to the "electrochemical field," the dissipation function is here precisely one-fourth of the Ohmic dissipation by the inhomogeneous external field required to produce the given history. This is in addition to the finite dissipation $\mathbf{E}\cdot\mathbf{J}$ already occurring in the steady state (per unit time), so that we may interpret the quantity as the *excess dissipation* (up to the additional factor of $\frac{1}{4}$.) Although we shall not show it here, the "entropy" $S(n)$ may be recovered from this quantity by minimizing subject to a single-time constraint. This provides also a simple thermodynamic interpretation of that quantity as the least total excess dissipation, integrated over time, required to produce a given density configuration at time zero. Incidentally, notice that the term in the exponent is of the order of the volume of the system $\sim \epsilon^{-d}$ only when integrated over times of order $\tau \sim \epsilon^{-1}$. The reason is that τ is a "drift time-scale" $\sim \epsilon^{-1}$ in microscopic units, whereas dissipation occurs on the longer "diffusive time-scale" $\sim \epsilon^{-2}$ in microscopic units. Only for such a long range of time will the cumulative dissipation be $O(1)$ in a unit macroscopic volume.

¿From these results we see finally that the most probable history subject to a given set of imposed constraints will be determined by a variational principle of *least excess dissipation,* which generalizes the famous "Onsager principle of least dissipation" to fluctuations in homogeneous nonequilibrium steady-states. One should be wary in interpreting such variational principles. The meaning of "constraint" here is "passive" rather than "active" [63]. That is, the principle does not apply to experimental situations where the constraints are enforced by some external means, but rather to the—exceedingly rare!—subensemble of fluctuation histories appearing spontaneously in the steady-state which satisfy the constraints. The most plausible application of the principle is instead to characterize the (absolute) most probable state without any constraints whatsoever, especially when there are multiple solutions of the deterministic hydrodynamic equations and internal noise is the physical selection mechanism.

5 Conclusions

(5.1) Outstanding Problems for DDS

Without a doubt the most outstanding issue for general DDS left unresolved in this work is the validity of the drift-diffusion equation (2.61). The main concern is consistency with the long-range correlations discussed in Section 3.3. We have seen that these correlations are likely to affect the thermodynamics of homogeneous steady-states of the DDS, making it shape-dependent. Therefore, several of the properties implicitly assumed in the derivation of hydrodynamics appear to fail. The doubt centers mostly about the *nonlinear* drift-diffusion equation. As discussed in Appendix VI, the correlation-function derivation of the *linearized* hydrodynamics appears to generalize to the "slow decay" case, and yields consistent results. Furthermore, as discussed in [30] (also Section 3.3) the linear drift-diffusion equation with a white-noise force actually *predicts* the expected power-law decay if the "naive" Price FDR is abandoned.

Even if the deterministic equation is valid, there is still a question whether the stochastic version Eq.(4.53) proposed in Section 4.2 gives the correct description of fluctuations in general. This nonlinear Langevin equation always leads to the naive FDR upon linearization. The general fluctuation dissipation theorem which we proved in Section 4.1 established that this nonlinear Langevin equation is *unique* subject to the conditions of Hypothesis 2 in Section 4.2. If the long-range correlations occur, then one of these conditions must be false in general. We strongly suspect that it is the assumed form of the stationary distribution, Eq.(4.28), which is wrong. In fact, we have seen in Section 3.3 that there is considerable delicacy in the description of the steady-state fluctuations even for the homogeneous state. The function $s(n)$, if it exists, may depend upon the way in which volumes go to infinity. This indeterminacy should be reflected in the stationary distribution of the nonlinear Langevin equation, although we do not have a definite proposal alternative to Eq.(4.28).

Perhaps it is fair to emphasize here the successes of our analysis: we have derived the drift-diffusion equation (2.61) by a fully microscopic procedure, yielding in principle explicit expressions for all the quantities involved (entropy function, drift velocities, and Onsager coefficients). Furthermore, we have established for this equation an H-theorem governing the approach of solutions to homogeneous steady-states. A nonlinear Langevin theory of fluctuations was proposed, in which a multiplicative white-noise force is added to the Eq.(2.61), unique subject to the condition of having the desired stationary distribution Eq.(4.28). Although we lack a rigorous proof, we are quite confident that these results are correct as stated in the "fast decay" case. Examples of the latter type are known—such as the ASEP model of Appendix III —so that these results have some nontrivial domain of validity.

(5.2) Remarks on the Nonequilibrium Distribution Function Method

Our analysis of the DLG models was meant in part to be a case study of the nonequilibrium distribution method of [2, 3, 5]. One of the main questions we set out to answer in this work was: *Where does the "nonequilibrium distribution function" method apply?* Our primary focus was on the class of systems not in local thermodynamic equilibrium, but we believe that the discussion has clarified the conditions for its applicability also in the standard situations. The key requirement is the condition of "fast decay" of correlations in space-time, and when that holds the method seems to be well-founded even if local states are not reversible. Therefore, there seems to be a basis to apply the method to a wide range of externally-driven nonequilibrium systems, discussed further in subsection 5.3 below.

The primary caveat concerns the long-range correlations, which seem to be ubiquitous in such non-equilibrium systems. The slow correlation decay makes the derivation by the nonequilibrium distribution suspect for two reasons: (i) the integrals over space-time of correlations no longer obviously converge, and (ii) there is no longer necessarily a unique relationship between density and chemical potential. It is not clear that these difficulties represent a *failure* of the method, since, as discussed above, it is possible that the drift-diffusion equation (2.61) itself is no longer valid. This is one of two alternative scenarios. The other is that the nonlinear equation is valid but requires a more refined derivation. A possibility worth exploring is the "level-2" approach of Zubarev-Morozov [7], since that method does not assume a chemical potential as a function of density but rather a *chemical potential functional* $G[f]$ required to produce a given *statistical distribution* $f[\rho]$ in the space of density fields $\rho(\mathbf{r})$. The functional relationship is elementary, $G[f] = -\log f$, so that no ambiguities from correlation effects appear at that level.

(5.3) Other Applications

Other possible applications of the methods discussed in this paper are reaction-diffusion systems (RDS) with local sources of reagents, plasmas in external electric and magnetic fields, and granular flow under the action of gravity, etc. Any system with hydrodynamic behavior can in principle be considered. Here we will just say a few words about the former two applications. The last subject is discussed in [64].

A hydrodynamic description of chemical systems is possible if the reaction rates are sufficiently slow, leading to reaction-diffusion equations. For example, see [65] in the context of stochastic lattice gas models. The homogeneous case was already treated by Zubarev with the distribution function method in Section 23.5 of his book [2]. It should be possible to treat also the inhomogeneous case with diffusion by his methods. A crucial difference with the DDS discussed in the present work is the non-conservative character of RDS. The reaction-diffusion equation is not of the form of a pure continuity equation, but contains also sources and sinks. Therefore, a number of aspects of our previous discussion would need to be changed. It should be noted that rigorous results are available for hydrodynamic large-deviations in the lattice-gas models of RDS [66, 67]. These can provide checks of formal theory.

Another interesting area of application is plasma systems, where the local equilibrium description also generally breaks down. Magnetohydrodynamics (MHD) is the generally proposed hydrodynamic description. A comprehensive discussion in the context of transport equations is contained in [68]. Here we will just make a few remarks pertinent to the issue of Onsager reciprocity in plasmas, which is reviewed by Krommes and Hu in [40]. It should be clear that our perspective largely agrees with theirs, since both works agree that OR is intrinsically connected with time-reversibility. It may be true for other reasons that a transport matrix is symmetric, such as self-adjointness of a collision operator [69], but this is not OR. In fact, as we have emphasized the *dissipative* Onsager matrix is always the symmetric part, but this is *not* a consequence of OR and has nothing to do with time-reversibility. We depart slightly from [40] in regarding GOR as a rather formal generalization of the original OR, and not nearly as useful, since it relates a physical situation to an unphysical one, the time-reversed state. The latter cannot be prepared in the laboratory since all reservoir dynamics would also need to be reversed. It is only theoretically possible to consider such reversal in the nonequilibrium steady-state for some artifical dissipative dynamics of the Nosé-Hoover type or for randomly modelled heat baths with the stochastic notion of time-reversal discussed in this work.

6 Appendices

Appendix I: Are Stationary Measures of the DLG Non-Gibbsian?

The reversible ($E = 0$) lattice gas is constructed in such a way that the canonical Gibbs measures $P_{\Lambda,N}(\eta) = Z_{\Lambda,N}^{-1} e^{-\beta H_\Lambda(\eta)} \delta_{N_\Lambda,N}$ are stationary for each $N = 0, 1, ..., |\Lambda|$, where the short-range Hamiltonian H_Λ is prescribed in the detailed balance condition for the exchange rates. Under finite drive, $E \neq 0$, the canonical Gibbs measures will in general no longer be stationary. This leads to the question whether there exists some other Hamiltonian, $\widetilde{H}_\Lambda$, such that the stationary measures are of the form $\widetilde{Z}_{\Lambda,N}^{-1} e^{-\beta \widetilde{H}_\Lambda(\eta)} \delta_{N_\Lambda,N}$. In finite volume, the answer is clearly positive, since the theory of finite-state Markov jump processes guarantees that for fixed N the stationary measure does not vanish on any configuration. Thus the question only becomes meaningful at infinite volume. Put differently, one has to understand how $\widetilde{H}_\Lambda$ depends on the volume Λ, for a given E and n.

Let P be a stationary, translation-invariant state of the DLG in infinite volume. We consider the distribution of the configuration η_B in the bounded domain B conditioned on the outside configuration η_{B^c}. The formal requirement for P to be a *Gibbs measure* is that there exist a set of interaction potentials $\{J_A : A \subset \mathbf{Z}^d\}$ on finite subsets of the lattice, which is translation-invariant, $J_A + \mathbf{x} = J_A$, and summable, $\sum_{A \ni \mathbf{0}} |A||J_A| < \infty$, such that the conditional expectations are of the form

$$P(\eta_B|\eta_{B^c}) = Z_B^{-1}(\eta_{B^c}) \exp\left[- \sum_{A \cap B \neq \emptyset} J_A \eta^A \right]. \tag{6.1}$$

Here $\eta^A = \prod_{\mathbf{x} \in A} \eta_{\mathbf{x}}$. Essentially, (6.1) states that conditional expectations do not vanish and depend continuously on η_{B^c}. See [26] for an extensive account of Gibbs measures and their properties. We only mention here that under suitable conditions on the potential— amounting to conditions of high temperature and low density—the Gibbs measure will be unique and exhibit exponential clustering. That is, for any local functions f, g on Ω, the truncated correlation will obey

$$|\langle f \cdot \tau_{\mathbf{x}} g \rangle - \langle f \rangle \langle g \rangle| \leq C_{f,g} e^{-\gamma |\mathbf{x}|}. \tag{6.2}$$

The constant $C_{f,g}$ depends upon the functions f, g but the decay constant γ does not. Since there is evidence, discussed in Section 3.3, that this clustering is not obeyed by the stationary meaures of the DLG, it is suggested that the invariant measures may not be Gibbs. However, even if the long-range decay could be established rigorously, this would not imply the measures are non-Gibbs. In other words, it would not be ruled out that the invariant measures are Gibbs states with long-range interactions (as happens for dipole systems) or are critical, where such power-law decays are commonly observed. Nevertheless, there are situations in physics where non-Gibbsian measures, violating Eq.(6.1), do appear. A notable example is the "effective distributions" produced by some real-space RG operations, treated very exhaustively in [70]. Section 4.5 of that work also discusses nonequilibrium steady-states.

However, for the DLG we simply do not yet know the answer. This is related to the fact that we have no convergent cluster expansion for the stationary measures (from which presumably Eq.(6.1) would follow.) There is only one system for which non-trivial steady-states have been constructed. It is a mixture of "first class" and "second class" particles in 1D. First and second class particles both jump only to the right, respecting exclusion at each site. However, a second class particle must always give way to a first class one. In this case, the stationary measures are non-Gibbs [54]. For $B = \{\mathbf{0}\}$ it is checked by explicit computation that the continuous dependence Eq.(6.1) fails.

Appendix II: Chemical Potential and Stationary Measures

We argued in Section 2.1 that for the DLG stationary measures, if not more generally, a variation in the density amounts to a variation in the chemical potential *only*. Here we state a theorem which supports this claim under a reasonable assumption. Let us assume that we have found a measure P on $\Omega = \{0,1\}^{\mathbf{Z}^d}$ which is time-invariant for the DLG ($P \in \mathcal{I}$) and invariant under spatial shifts ($P \in \mathcal{S}$). This means that

$$\int dP\, Lf = 0 \tag{6.3}$$

and

$$\int dP\, \tau_{\mathbf{x}} f = \int dP\, f, \qquad \mathbf{x} \in \mathbf{Z}^d, \tag{6.4}$$

for all local functions f. In addition, let us suppose there exists a potential $\{J_A : A \subset \mathbf{Z}^d\}$ which obeys the summability condition

$$\sum_{A \ni \mathbf{0}} |A| \cdot |J_A| < \infty, \tag{6.5}$$

such that for each finite $B \subset \mathbf{Z}^d$,

$$P(\eta_B | \eta_{B^c}) = Z_B(\eta_{B^c})^{-1} \exp\left[- \sum_{A \cap B \neq \emptyset} J_A \eta^A \right]. \tag{6.6}$$

Comparing with Eq.(6.1), we see that this amounts to $P \in \mathcal{G}(J)$. A related notion is that of *canonical Gibbs measures* for the potential J, denoted $\mathcal{G}_c(J)$, with conditional expectations

$$P(\eta_B | n_B, \eta_{B^c}) = Z_B(n_B, \eta_{B^c})^{-1} \exp\left[- \sum_{A \cap B \neq \emptyset} J_A \eta^A \right]. \tag{6.7}$$

The difference here is that the conditioning is also with respect to $n_B(\eta)$, the number of particles in B. The standard reference on this subject is [76]. The result we need for the DLG is the following

Theorem 1 *Suppose for some summable potential J, that $\mathcal{I}\cap\mathcal{S}\cap\mathcal{G}(J)\neq\emptyset$. Then, $\mathcal{I}\cap\mathcal{S}\subseteq\mathcal{G}_c(J)$.*

This theorem, due to A. Asselah, extends a previous result of Künsch [33] for non-reversible spin-flip processes to DLG. Our main interest here is in its consequence

Corollary 1 *Under the assumption of the previous theorem, if $Q\in\mathcal{I}$ is also ergodic to space-translations, then there exists a chemical potential $\lambda=J_{\{\mathbf{0}\}}$ such that $Q\in\mathcal{G}(J_{\{\mathbf{0}\}},\{J_A:|A|\geq 2\})$.*

This follows from the previous result based upon two well-known facts: for a translation-invariant potential J, (i) the boundary set ex$\mathcal{G}_c(J)$ of extremal elements of the Choquet simplex $\mathcal{G}_c(J)$ is characterized as the subset of elements ergodic under the space-shifts [26], and (ii) ex$\mathcal{G}_c(J)=\bigcup_\lambda$ ex$\mathcal{G}(\lambda,\{J\})$ (Theorems 5.14-5.15, [76].) Hence, we obtain our desired characterization of the invariant measures Q as Gibbs for the potential $\{J_A:|A|\geq 2\}$ and for some chemical potential λ.

We here only sketch the proof of the Theorem, which is technical because one wants to get away with minimal assumptions on the potential J. The main idea is to consider the time-derivative of the *relative entropy* per unit volume, $h(Q|P)$. Since we are in the translation-invariant set up, this quantity is well-defined. Moreover, because Q is assumed stationary,

$$\frac{d}{dt}h(Qe^{Lt}|P)\Big|_{t=0}=0. \tag{6.8}$$

Let us define for any pair of sites $\mathbf{x},\mathbf{y}$ the measure $Q\Delta_{\mathbf{xy}}$ by

$$\int d(Q\Delta_{\mathbf{xy}})\,f=\int dQ\,(\Delta_{\mathbf{xy}}f), \tag{6.9}$$

for all local functions f. Writing out the time derivative in Eq.(6.8) explicitly, one deduces after a careful estimate of boundary terms the following result: For each of the bounded sets of lattice sites $B_n = [-2^n, 2^n]^d$ and each nearest-neighbor pair $\langle \mathbf{x}, \mathbf{y} \rangle \subset \tilde{B}_n = [-2^n + n, 2^n - n]^d$,

$$Q_{B_n}\left(\eta^{\mathbf{xy}}_{B_n}\right) = \int \frac{d(P\Delta^{\mathbf{xy}})}{dP(\eta)} Q(\eta_{B_n}, d\eta_{B_n^c}). \tag{6.10}$$

Therefore, $\int \Delta^{\mathbf{xy}} f\, dQ = \int \frac{d(P\Delta^{\mathbf{xy}})}{dP} f\, dQ$ for every local function f and every nearest-neighbor pair $\langle \mathbf{x}, \mathbf{y} \rangle$. The conclusion is that

$$\frac{d(Q\Delta_{\mathbf{xy}})}{dQ(\eta)} = \frac{d(P\Delta_{\mathbf{xy}})}{dP(\eta)} \tag{6.11}$$

for all $\eta \in \Omega$. On the other hand, if B is any bounded set of lattice sites and $\mathcal{B}(n_B, \eta_{B^c})$ is the σ-field generated by the random variables $n_B, \{\eta_{\mathbf{x}} | \mathbf{x} \in B^c\}$, then a Borel function f is $\mathcal{B}(n_B, \eta_{B^c})$-measurable if and only if $\Delta_{\mathbf{x},\mathbf{y}} f = f$ for all nearest-neighbor pairs $\langle \mathbf{x}, \mathbf{y} \rangle$ with $\mathbf{x}, \mathbf{y} \in B$. In that case it is easy to see that

$$\frac{d(Q(\cdot | n_B, \eta_{B^c})\Delta_{\mathbf{xy}})}{dQ(\eta_B | n_B, \eta_{B^c})} = \frac{d(Q\Delta_{\mathbf{xy}})}{dQ(\eta)}. \tag{6.12}$$

Because any two configurations η_B, η'_B in B with n_B, η_{B^c} fixed can be obtained by a sequence of nearest-neighbor exchanges, $\eta'_B = \Delta^{\mathbf{x}_M \mathbf{y}_M} \cdots \Delta^{\mathbf{x}_1 \mathbf{y}_1} \eta_B$, it follows from the combination of Eqs.(6.12) and (6.11) that

$$Q(\eta_B | n_B, \eta_{B^c}) = P(\eta_B | n_B, \eta_{B^c}) \tag{6.13}$$

for all bounded sets B. Therefore, $Q \in \mathcal{G}_c(J)$.

There is a slightly different way to state the Theorem, which may lend plausibility to its hypothesis. According to Eq.(2.67), the rates of the time-reversed process with respect to $P \in \mathcal{I}$ are given by

$$c^r(\mathbf{x}, \mathbf{y}, \eta) = c(\mathbf{x}, \mathbf{y}, \eta^{\mathbf{xy}}) \frac{d(P\Delta_{\mathbf{xy}})}{dP(\eta)}, \tag{6.14}$$

which is defined for all η, if P is Gibbs. However, it is then true by the Theorem (cf. Eq.(6.11)) that the righthand side is the *same* for all $Q \in \mathcal{I}$. Therefore, the time-reversed process does not depend upon the stationary measure which is used to define it when one of the measures (and thus each of them) is Gibbs.

It would be of interest—in particular in view of our discussion in Appendix I—to avoid the assumption of P being Gibbs altogether. However, if also non-translation invariant states are permitted, then there must be some condition which excludes the "blocked states" P_b, defined by the conditional expectations

$$P_b(\eta_B|\eta_{B^c}) = Z_B^{-1}(\eta_{B^c}) \exp\left[-\sum_{A\cap B\neq\emptyset} \overline{J}_A\eta^A + \sum_{\mathbf{x}\in B} q\beta(\mathbf{E}\cdot\mathbf{x})\eta_{\mathbf{x}}\right], \tag{6.15}$$

where $\overline{J}$ is the short-range potential defining the Hamiltonian in Eq.(1.3). The measures P_b are stationary and reversible.

Appendix III: The Asymmetric Simple Exclusion Process

The asymmetric exclusion process (ASEP) is defined by the rates

$$c(\mathbf{x},\mathbf{y},\eta) = p(\mathbf{y}-\mathbf{x})\eta_{\mathbf{x}}(1-\eta_{\mathbf{y}}) + p(\mathbf{x}-\mathbf{y})(1-\eta_{\mathbf{x}})\eta_{\mathbf{y}}, \tag{6.16}$$

where $p(\mathbf{x}) \geq 0$ with compact support, $p(\mathbf{0}) = 0$, and $\sum_{\mathbf{x}} p(\mathbf{x}) = 1$ as a normalization. Thus after an exponentially distributed waiting time a particle at site $\mathbf{x}$ jumps with probability $p(\mathbf{y}-\mathbf{x})$ to site $\mathbf{y}$ provided this site is empty. If

$$p(\mathbf{x}) = p(-\mathbf{x})e^{\beta q\mathbf{E}\cdot\mathbf{x}}, \tag{6.17}$$

then Eq.(3) is satisfied with $H = 0$. For nearest neighbor jumps only, i.e. $p(\mathbf{x}) = 0$ whenever $|\mathbf{x}| > 1$, Eq.(6.17) can always be fullfilled. In general, we do not impose such a restriction. The very welcome simplification of ASEP originates in the assumption that the exchange rates depend *only* on the occupation variables at sites $\mathbf{x}$ and $\mathbf{y}$. For the ASEP a number of rigorous results are available which will be briefly reviewed here.

The Bernoulli measures, i.e. the $\eta_{\mathbf{x}}$'s are independent and $\langle\eta_{\mathbf{x}}\rangle_n = n$, are stationary for the rates (A.1). In fact, these are the only stationary and translation invariant measures in infinite volume [71]. If Eq.(6.17) holds, then also the inhomogeneous blocked states

$$Z^{-1}\exp[\beta q\sum_{\mathbf{x}}(\mathbf{E}\cdot\mathbf{x})\eta_{\mathbf{x}}] \tag{6.18}$$

and their translates are stationary. They play no role in hydrodynamics. The two point function $S(\mathbf{x}) = \langle \eta_{\mathbf{x}} \eta_{\mathbf{0}} \rangle_n - n^2 = \delta_{\mathbf{x}\mathbf{0}} n(1-n)$ and consequently the structure function $\hat{S}(\mathbf{k}) = n(1-n)$, which is independent of $\mathbf{k}$. Of course, large deviations Eq.(2.11) are satisfied with the entropy function $s(n) = -[n \log n + (1-n)\log(1-n) + \log 2]$ relative to density $n^* = 1/2$. The average current

$$\hat{\mathbf{j}}(n) = n(1-n) \sum_{\mathbf{x}} \mathbf{x} p(\mathbf{x}). \tag{6.19}$$

The rates of the time reversed process are given by

$$c^r(\mathbf{x}, \mathbf{y}, \eta) = p(\mathbf{x} - \mathbf{y})\eta_{\mathbf{x}}(1 - \eta_{\mathbf{y}}) + p(\mathbf{y} - \mathbf{x})(1 - \eta_{\mathbf{x}})\eta_{\mathbf{y}}, \tag{6.20}$$

which reflects the simplicity of the invariant measure. In particular, if Eq.(6.16) holds, the reversed rates also satisfy Eq.(6.16) with $\mathbf{E}$ replaced by $-\mathbf{E}$.

The exclusion process is reversible iff $p(\mathbf{x}) = p(-\mathbf{x})$ in which case it is called symmetric. This corresponds to the equilibrium situation. The average current vanishes, $\sum_{\mathbf{x}} \mathbf{x} p(\mathbf{x}) = \mathbf{0}$. Conductivity, Onsager matrix, and diffusion matrix are given by

$$(q\beta)^{-1}\sigma_{ml} = n(1-n)\frac{1}{2}\sum_{\mathbf{x}} x_m x_l p(\mathbf{x}) = L_{ml} = n(1-n)D_{ml}. \tag{6.21}$$

The latter is independent of n and the hydrodynamic equation becomes linear. Note that we could impose $\hat{\mathbf{j}}(n) = n(1-n)\sum_{\mathbf{x}} \mathbf{x} p(\mathbf{x}) = \mathbf{0}$ and still $p(\mathbf{x}) \neq p(-\mathbf{x})$. This is an example of a zero current, but not time-reversal invariant lattice gas.

For the ASEP the Euler equations read

$$\partial_\tau n(\mathbf{r}, \tau) + \partial_m \hat{\jmath}_m(n(\mathbf{r}, \tau)) = 0 \tag{6.22}$$

which are established in full generality by Rezakhanlou [72], with prior results detailed in [20], Notes to Section 4.2. He proves that on the time scale $\sim \epsilon^{-1}$ and the spatial scale $\sim \epsilon^{-1}$ in microscopic units a typical density profile follows the unique entropic solution to Eq.(6.22). In particular, his results cover times beyond the first shock and entropy may no longer be conserved.

Next we turn to the Onsager matrix. The static part reads

$$L_{ml}^{stat} = n(1-n)\frac{1}{2}\sum_{\mathbf{x}} x_m x_l p(\mathbf{x}) \tag{6.23}$$

which is clearly symmetric. Note that the second jump moments are *not* truncated. For the dynamic part we use

$$\begin{aligned}
\mathbf{j}(\mathbf{x}) &= \frac{1}{2}\sum_{\mathbf{y}} \mathbf{y}[p(\mathbf{y})\eta_{\mathbf{x}}(1-\eta_{\mathbf{x}+\mathbf{y}}) - p(-\mathbf{y})(1-\eta_{\mathbf{x}})\eta_{\mathbf{x}+\mathbf{y}}],\\
\mathbf{j}^r(\mathbf{x}) &= \frac{1}{2}\sum_{\mathbf{y}} \mathbf{y}[p(-\mathbf{y})\eta_{\mathbf{x}}(1-\eta_{\mathbf{x}+\mathbf{y}}) - p(\mathbf{y})(1-\eta_{\mathbf{x}})\eta_{\mathbf{x}+\mathbf{y}}],\\
\mathbf{j}^s(\mathbf{x}) &= \frac{1}{2}[\mathbf{j}(\mathbf{x}) + \mathbf{j}^r(\mathbf{x})] = \frac{1}{2}\sum_{\mathbf{y}} \mathbf{y}(p(\mathbf{y}) + p(-\mathbf{y}))(\eta_{\mathbf{x}} - \eta_{\mathbf{x}+\mathbf{y}}),\\
\mathbf{j}^a(\mathbf{x}) &= \frac{1}{2}[\mathbf{j}(\mathbf{x}) - \mathbf{j}^r(\mathbf{x})] = \frac{1}{2}\sum_{\mathbf{y}} \mathbf{y}((p(\mathbf{y}) - p(-\mathbf{y}))(\eta_{\mathbf{x}} - \eta_{\mathbf{x}+\mathbf{y}})^2,
\end{aligned} \tag{6.24}$$

and we note that

$$\sum_{\mathbf{x}} \mathbf{j}^s(\mathbf{x}) = 0. \tag{6.25}$$

Therefore

$$\begin{aligned}
L_{ml}^{dyn} &= -\int_0^\infty dt \left[\sum_{\mathbf{x}} \langle j_m^r(\mathbf{x}) e^{tL} j_l(\mathbf{0})\rangle_n^\top - \chi(n)\hat{\jmath}'_m(n)\hat{\jmath}'_l(n)\right]\\
&= \int_0^\infty dt \left[\sum_{\mathbf{x}} \langle j_m^a(\mathbf{x}) e^{tL} j^a(\mathbf{0})\rangle_n^\top - \chi(n)\hat{\jmath}'_m(n)\hat{\jmath}'_l(n)\right].
\end{aligned} \tag{6.26}$$

Since the RHS of Eq.(6.26) is an autocorrelation, $\mathbf{L}^{dyn} \geq \mathbf{0}$ as a matrix. Together with Eq.(6.23) this implies $\mathbf{D}(\mathbf{E}) \geq \mathbf{D}(E=0)$. Thus the driving tends to increase diffusion. In fact, it could increase it so much that $L = \infty$. According to the standard physical picture [73, 74] this will be the case whenever $\hat{\mathbf{j}}(n) \neq \mathbf{0}$ and $d = 1, 2$. If $\hat{\mathbf{j}}(n) = \mathbf{0}$, then the integral Eq.(6.26) should exist in any dimension.

In his thesis Lin Xu [75] considers the non-reversible ASEP with zero current $\hat{\mathbf{j}}(n) = \mathbf{0}$ in one spatial dimension. Although not carried out explicitly, the results are asserted for any dimension. He proves that on the (microscopic) time scale ϵ^{-2}, i.e. on the diffusive time scale, the density is approximated with probability one by the solution of the nonlinear diffusion equation

$$\partial_\tau n(\mathbf{r},\tau) = \partial_m D_{ml}(n(\mathbf{r},\tau))\partial_l n(\mathbf{r},\tau) \tag{6.27}$$

with $D_{ml} = (n(1-n))^{-1}(\mathbf{L}^{stat} + \mathbf{L}^{dyn})_{ml}$ from Eqs.(6.23), (6.26). Lin Xu also provides a lower bound on $\mathbf{D}$ which in our notation reads $\mathbf{L}^{dyn} > \mathbf{0}$. Note that while the ASEP is nonreversible the Gaussian fluctuations in the steady-state are given by a reversible Ornstein-Uhlenbeck process. Thus, on the large scale reversibility is regained.

Esposito, Marra, and Yau (EMY) [16] study the ASEP with $\hat{\mathbf{j}}(n) \neq \mathbf{0}$ in three and more dimensions. One immediate difficulty stems from the difference in Euler and Navier-Stokes time scale, equivalently, from their different scaling under spatial dilations. EMY decided to adopt a procedure familiar from incompressible Navier-Stokes equations for fluids. They fix a reference density n^* and take a small density deviation of order ϵ, i.e. the chemical potential in the local equilibrium state Eq.(2.26) is of order ϵ. Expanding Eq.(6.19) there is a constant drift velocity $\mathbf{c}(n^*) = (1-2n^*)\sum_{\mathbf{x}} \mathbf{x}p(\mathbf{x})$. Thus one is led to consider density deviations of order ϵ in a moving frame of reference. EMY prove that the average density, on the Navier-Stokes time scale,

$$\lim_{\epsilon\to 0} \epsilon^{-1}[\bar{n}_\epsilon(\mathbf{r} - \epsilon^{-1}c(n^*)\tau, \epsilon^{-1}\tau) - n^*] = n(\mathbf{r},\tau) \tag{6.28}$$

has a limit as $\epsilon \to 0$. The limit density satifies

$$\partial_\tau n(\mathbf{r},\tau) = \partial_m[-(\sum_{\mathbf{x}} x_m p(\mathbf{x}))n(\mathbf{r},\tau)^2 + D_{ml}(n^*)\partial_l n(\mathbf{r},\tau)] \tag{6.29}$$

with $D_{ml} = (n(1-n))^{-1}(\mathbf{L}^{stat} + \mathbf{L}^{dyn})_{ml}$ from Eqs.(6.23), (6.26). Again it is established that $\mathbf{L}^{dyn} > \mathbf{0}$. EMY also prove that, on the scales considered, typical density profiles follow the solution to Eq.(6.29).

Finally, Landim, Olla and Yau (LOY) [17] have studied more recently in the same model as EMY the problem equivalent to ours in the text. Namely, they prove that the average density $\bar{n}_\epsilon(\mathbf{r},\tau)$ on the Euler time-scale $\sim \epsilon^{-1}$ is given correctly to order ϵ by the solution $n_\epsilon(\mathbf{r},\tau)$ of the drift-diffusion equation

$$\partial_\tau n(\mathbf{r},\tau) + \partial_m \hat{\jmath}_m(n(\mathbf{r},\tau)) = \epsilon\partial_m D_{ml}(n(\mathbf{r},\tau))\partial_l n(\mathbf{r},\tau) \tag{6.30}$$

with again $D_{ml} = (n(1-n))^{-1}(\mathbf{L}^{stat} + \mathbf{L}^{dyn})_{ml}$ from Eqs.(6.23), (6.26). This is proved in the precise sense that

$$\int_\Lambda d^d\mathbf{r}\phi(\mathbf{r})\,[\bar{n}_\epsilon(\mathbf{r},\tau) - n_\epsilon(\mathbf{r},\tau)] = o(\epsilon), \tag{6.31}$$

for any nice test function ϕ (i.e. $\phi \in H^1$) and for each fixed $\tau < T_0$, the time of appearance of the first shock in the solution of Eq.(6.22) for the same initial data. This is a result of the same type, just slightly weaker, as that conjectured in Eq.(2.62) in the text. LOY prove no results for typical density profiles in this setting.

Appendix IV: Steady-State Stability and the FDR of Kraichnan

In a very interesting work [77], which is unfortunately little referenced outside the turbulence literature, R. H. Kraichnan in 1959 derived a microscopic version of the FDR of 2nd type for a general classical dynamics

$$dx_i(t)/dt = V_i(x(t)) \tag{6.32}$$

with a conserved quantity, or"energy" $H(x)$, and a Liouville theorem for conservation of phase volume. These guarantee that the "Gibbs measures" $\propto e^{-\beta H}$ are stationary for the dynamics. Under these same two conditions, Kraichnan established that the mean response function in the stationary "Gibbs state"

$$G_{ij}(t,s) = \langle \frac{\delta x_i(t)}{\delta f_j(s)} \rangle \tag{6.33}$$

associated to a change in the external force f_j coupled as:

$$\frac{dx_i}{dt}(t) = V_i(x(t)) + f_i(t), \tag{6.34}$$

is determined by the FDR

$$G_{ij}(t,s) = \beta\langle x_i(t)\cdot \frac{\partial H}{\partial x_j}(x(s))\rangle. \tag{6.35}$$

His two page derivation is interesting because it shows that the FDR arises as the consequence of "stability" of the Gibbs state to coupling of two independent, identical systems in a general way preserving conservation of total energy $H(x)+H(y)$ and a Liouville theorem for the coupled dynamics. This is very reminiscent of the standard argument for positive equilibrium response, or susceptibility, which we gave in Eqs.(2.23),(2.24). In fact, if the distribution on "histories" is regarded as a formal Gibbs distribution in one higher dimension, then the arguments are quite analogous. Kraichnan's method yields also the corresponding quantum KMS conditions and 2nd type FDR's in the context of stochastic Langevin equations.

Actually, it is possible to derive Kraichnan's FDR in an even simpler way as a direct corollary of the *classical KMS condition*

$$\langle \{f, g\} \rangle = \beta \langle g\{f, H\} \rangle, \tag{6.36}$$

in which $\{f, g\}$ is the canonical Poisson bracket; see [78, 79]. This condition is easily proved at finite-volume by a simple integration by parts, and also at infinite-volume for a suitable class of functions f, g. If one takes $f = p_j$ one gets

$$\langle \frac{\partial g}{\partial x_j} \rangle = \beta \langle g \frac{\partial H}{\partial x_j} \rangle, \tag{6.37}$$

which is also true (by the same arguments) even if the dynamical vector field V is not Hamiltonian. It is easy to see that Eq.(6.37) with $g = x_i(t)$ gives Kraichnan's FDR. In fact,

$$\frac{\delta x_i(t)}{\delta f_j(0)} = \frac{\partial x_i(t)}{\partial x_j}, \tag{6.38}$$

because making the small perturbation to the force $f_j(s) \to f_j(s) + \epsilon\delta(s)$ is the same as making the small perturbation $x_j \to x_j + \epsilon$ to the initial data. It is rather natural to see that Kraichnan's FDR is a consequence of the KMS condition, because the latter is a more general expression of "stability" of the state. It is known, for example, that the classical KMS condition Eq.(6.36) arises as a consequence of stability of the Gibbs measure to small perturbations of the Hamiltonian dynamics [80].

In the case of a linear Langevin dynamics such as our Eq.(3.13), it is easy to check that Kraichnan's method gives a result

$$\langle \frac{\delta \alpha_i(t)}{\delta \eta_j(0)} \rangle = \langle \alpha_i(t) X_j(0) \rangle, \tag{6.39}$$

equivalent to Eq.(3.36). However, note that while Kraichnan's argument applies also to the nonlinear Langevin equations, like our Eq.(4.2), it does not yield a relation obviously equivalent to Graham's 2nd type FDR in that context. At least it does not in Kraichnan's original form of the argument. In fact, the two FDR's of Kraichnan and Graham for this case deal with the response functions to differently imposed forces.

Appendix V: Microscopic Current Correlations in the DLG Models

For $t > s$ we calculate that

$$\begin{aligned}
\mathsf{E}_n(J_m(\mathbf{x},t)J_l(\mathbf{y},s)) &= \lim_{\delta\to 0}\frac{1}{\delta}\int_0^\delta du\ \mathsf{E}_n(J_m(\mathbf{x},t-s)J_l(\mathbf{y},u)) \\
&= \lim_{\delta\to 0}\int P_n(d\eta)\ \mathsf{E}^\eta\left[\mathsf{E}^\eta\left(J_m(\mathbf{x},t-s)|\mathcal{F}_\delta\right)\cdot\frac{1}{\delta}\int_0^\delta du\ J_l(\mathbf{y},u)\right] \\
&= \lim_{\delta\to 0}\int P_n(d\eta)\ \mathsf{E}^\eta\left[\mathsf{E}^{\eta^{\mathbf{y},\mathbf{y}+\hat{\mathbf{e}}_l}}\left(J_m(\mathbf{x},t-s-\delta)\right)\cdot\frac{1}{\delta}\int_0^\delta du\ J_l(\mathbf{y},u)\right] \\
&= \lim_{\delta\to 0}\int P_n(d\eta)\ \left(\Delta_{\mathbf{y},\mathbf{y}+\hat{\mathbf{e}}_l}e^{(t-s-\delta)L}j_m(\mathbf{x})\right)(\eta)\cdot\frac{1}{\delta}\int_0^\delta du\ \left(e^{uL}j_l(\mathbf{y})\right)(\eta) \\
&= \int P_n(d\eta)\ \left(\Delta_{\mathbf{y},\mathbf{y}+\hat{\mathbf{e}}_l}e^{(t-s)L}j_m(\mathbf{x})\right)(\eta)\cdot j_l(\mathbf{y},\eta). \qquad (6.40)
\end{aligned}$$

We can say a few words to explain and justify each line. The first uses just stationarity. The second line introduces the conditional expectation $\mathsf{E}^\eta(\cdot)$ given that the configuration at time zero is η and the sigma algebra $\mathcal{F}_t = \sigma\{N(\cdot,s) : s < t\}$ of events prior to time t. In the next line, the Markov property

$$\mathsf{E}^\eta\left(J_m(\mathbf{x},t-s)|\mathcal{F}_\delta\right) = \mathsf{E}^{N(\cdot,\delta)}\left(J_m(\mathbf{x},t-s-\delta)\right) \qquad (6.41)$$

was used, along with the fact that $N(\cdot,\delta) = \eta^{\mathbf{y},\mathbf{y}+\hat{\mathbf{e}}_l}$ with a probability going to one as $\delta\to 0$ for the histories contributing in the expectation. (Only the histories for which $\int_0^\delta du\ J_l(\mathbf{y},u) \neq 0$ contribute and the probability of an additional exchange in the time-interval of length δ goes to zero.) Finally the definitions of the semigroup e^{tL} and the systematic current $j_m(\mathbf{x},\eta)$ are used and the limit $\delta\to 0$ is taken. An entirely identical calculation gives for $s > t$ that

$$\mathsf{E}_n(J_m(\mathbf{x},t)J_l(\mathbf{y},s)) = \int P_n(d\eta)\ j_m(\mathbf{x},\eta)\cdot\left(\Delta_{\mathbf{x},\mathbf{x}+\hat{\mathbf{e}}_m}e^{(s-t)L}j_l(\mathbf{y})\right)(\eta). \qquad (6.42)$$

Another contribution appears if $s = t$ which is calculated as

$$\lim_{\delta\to 0}\frac{1}{\delta}\int_0^\delta du\int_0^\delta du'\ \mathsf{E}_n(J_m(\mathbf{x},u)J_l(\mathbf{y},u')) = \delta_{\mathbf{x},\mathbf{y}}\delta_{mn}\lim_{\delta\to 0}\frac{1}{\delta}\mathsf{E}_n\left[\mathcal{N}_{\mathbf{x},\mathbf{x}+\hat{\mathbf{e}}_m}([0,\delta])\right]$$

$$= \quad \delta_{\mathbf{x},\mathbf{y}}\delta_{mn}\langle c(\mathbf{x},\mathbf{x}+\hat{\mathbf{e}}_m)\rangle_n. \tag{6.43}$$

We introduced the random variable $\mathcal{N}_{\mathbf{x},\mathbf{x}+\hat{\mathbf{e}}_m}([s,t])$ which counts the absolute number of exchanges between sites $\mathbf{x}$ and $\mathbf{x}+\hat{\mathbf{e}}_m$ in the time-interval $[s,t]$. Altogether we see that

$$\begin{aligned} \mathsf{E}_n(J_m(\mathbf{x},t)J_l(\mathbf{y},s)) &= \langle c(\mathbf{0},\hat{\mathbf{e}}_m)\rangle_n\delta_{ml}\delta_{\mathbf{xy}}\delta(t-s) \\ &\quad +\theta(t-s)\langle\Delta_{\mathbf{y},\mathbf{y}+\hat{\mathbf{e}}_l}e^{L(t-s)}j_m(\mathbf{x})\cdot j_l(\mathbf{y})\rangle_n \\ &\quad +\theta(s-t)\langle\Delta_{\mathbf{x},\mathbf{x}+\hat{\mathbf{e}}_m}e^{L(s-t)}j_l(\mathbf{y})\cdot j_m(\mathbf{x})\rangle_n. \end{aligned} \tag{6.44}$$

On the other hand, using the identity Eq.(2.68), with $f = e^{tL}j_m(\mathbf{0})\cdot(\eta_{\mathbf{x}}-\eta_{\mathbf{x}+\hat{\mathbf{e}}_l})$, we can rewrite this as

$$\begin{aligned} \mathsf{E}_n(J_m(\mathbf{x},t)J_l(\mathbf{y},s)) &= \langle c(\mathbf{0},\hat{\mathbf{e}}_m)\rangle_n\delta_{ml}\delta_{\mathbf{xy}}\delta(t-s) \\ &\quad -\theta(t-s)\langle e^{L(t-s)}j_m(\mathbf{x})\cdot j_l^r(\mathbf{y})\rangle_n \\ &\quad -\theta(s-t)\langle e^{L(s-t)}j_l(\mathbf{y})\cdot j_m^r(\mathbf{x})\rangle_n. \end{aligned} \tag{6.45}$$

This is exactly the result claimed in Eq.(3.56) of Section 3.3.

Appendix VI: Kadanoff-Martin Approach to Hydrodynamics in DLG

We shall repeat the argument in Appendix 3 of the KLS paper, but making only the weaker regularity assumption that

$$\chi(n,\hat{\mathbf{k}}) \equiv \lim_{k\to 0} S_n(\mathbf{k}) \tag{6.46}$$

exists. By integration we obtain distinct relations $n(\lambda,\hat{\mathbf{k}})$ between density and chemical potential, depending upon the wavevector direction of the density modulation in space. Since it is still supposed to be true that

$$\frac{S(\mathbf{k},t)}{S(\mathbf{k})} \approx \exp\left[i(\mathbf{c}\cdot\mathbf{k})t - (\mathbf{k}\cdot\mathbf{D}\cdot\mathbf{k})t\right] \tag{6.47}$$

at long times and low wavenumbers, we may still define

$$\hat{\mathbf{k}}\cdot\mathbf{c}(\hat{\mathbf{k}}) \equiv \lim_{k\to 0}\lim_{t\to\infty}\frac{1}{t}\frac{\partial}{\partial k}\left[\frac{S(\mathbf{k},t)}{S(\mathbf{k})}\right], \tag{6.48}$$

and

$$\hat{\mathbf{k}}\cdot\mathbf{D}\cdot\hat{\mathbf{k}} \equiv \lim_{k\to 0}\lim_{t\to\infty}\frac{-1}{2t}\left\{\frac{\partial^2}{\partial k^2}\left[\frac{S(\mathbf{k},t)}{S(\mathbf{k})}\right] - (\hat{\mathbf{k}}\cdot\mathbf{c}(\hat{\mathbf{k}}))^2 t^2\right\}. \tag{6.49}$$

Let us examine each of these in turn.

For the first, we calculate that

$$\hat{\mathbf{k}}\cdot\mathbf{c}(\hat{\mathbf{k}}) = \lim_{k\to 0}\lim_{t\to\infty}\frac{1}{t}\cdot\frac{1}{S(\mathbf{k})}\sum_{\mathbf{x}} e^{i\mathbf{k}\cdot\mathbf{x}}(\hat{\mathbf{k}}\cdot\mathbf{x})\mathsf{E}_n(N(\mathbf{x},t)N(\mathbf{0},0))^{\top}, \tag{6.50}$$

if we assume that limits such as $\lim_{k\to 0}\sum_{\mathbf{x}} e^{i\mathbf{k}\cdot\mathbf{x}} x_m \mathsf{E}_n(N(\mathbf{x},t)N(\mathbf{0},0))^{\top}$ exist and are finite. In that case, all the contributions from the derivative $\frac{\partial}{\partial k}$ of $S^{-1}(\mathbf{k})$ vanish. Then, using the conservation law, we obtain

$$\begin{aligned}
\hat{\mathbf{k}}\cdot\mathbf{c}(\hat{\mathbf{k}}) &= \frac{\hat{\mathbf{k}}}{\chi(\hat{\mathbf{k}})}\cdot\left[\lim_{k\to 0}\lim_{t\to\infty}\frac{1}{t}\int_0^t ds\ \sum_{\mathbf{x}} e^{i\mathbf{k}\cdot\mathbf{x}}\mathsf{E}_n(\mathbf{J}(\mathbf{x},s)N(\mathbf{0},0))^{\top}\right] \\
&= \frac{\hat{\mathbf{k}}}{\chi(\hat{\mathbf{k}})}\cdot\left[\lim_{k\to 0}\sum_{\mathbf{x}} e^{i\mathbf{k}\cdot\mathbf{x}}\langle\mathbf{j}(\mathbf{x})\eta_{\mathbf{0}}\rangle_n^{\top}\right].
\end{aligned} \tag{6.51}$$

In fact, we shall show that $\mathbf{c}$ is actually $\hat{\mathbf{k}}$-*independent* under the assumption that *there is a unique stationary state P_n for each density n*. The result follows from the chain of equalities:

$$\begin{aligned}
\mathbf{c}(n,\hat{\mathbf{k}}) &= \frac{1}{\chi(n,\hat{\mathbf{k}})}\lim_{k\to 0}\frac{\partial}{\partial\lambda}\left[\frac{1}{Z(\lambda)}\langle\mathbf{j}(\mathbf{0})\exp\left(\lambda\sum_{\mathbf{x}} e^{i\mathbf{k}\cdot\mathbf{x}}\eta_{\mathbf{x}}\right)\rangle_n\right]_{\lambda=0} \\
&= \frac{1}{\chi(n,\hat{\mathbf{k}})}\frac{\partial}{\partial\lambda}\hat{\mathbf{j}}(n(\lambda,\hat{\mathbf{k}}))|_{\lambda=\lambda(n,\hat{\mathbf{k}})} \\
&= \frac{\partial\hat{\mathbf{j}}}{\partial n}(n).
\end{aligned} \tag{6.52}$$

Therefore, the same formula holds for $\mathbf{c}$ as before.

Actually, the previous argument applies generally if we define "$\hat{\mathbf{k}}$-dependent Zwanzig-Mori spaces," via the inner-product

$$\langle A|B\rangle_{n,\hat{\mathbf{k}}} = \lim_{k\to 0}\sum_{\mathbf{x}} e^{i\mathbf{k}\cdot\mathbf{x}}\mathsf{E}_n(A(\mathbf{x})B(\mathbf{0}))_n^{\top}, \tag{6.53}$$

and establishes that

$$\langle A|N\rangle_{n,\hat{\mathbf{k}}} = \chi(n,\hat{\mathbf{k}})\frac{\partial\hat{A}}{\partial n}(n). \tag{6.54}$$

Therefore, all of the $\hat{\mathbf{k}}$-dependence is through the susceptibility in this expression. Furthermore, if we assume an ergodic property of the dynamics, then it is generally true as well that

$$\lim_{t\to\infty}\langle A(t)|B\rangle_{n,\hat{\mathbf{k}}} = \chi(n,\hat{\mathbf{k}})\frac{\partial \hat{A}}{\partial n}(n)\frac{\partial \hat{B}}{\partial n}(n). \tag{6.55}$$

The proof is just that

$$\begin{aligned}
\lim_{t\to\infty}\langle A(t)|B\rangle_{n,\hat{\mathbf{k}}} &= \lim_{k\to 0}\lim_{t\to\infty}\sum_{\mathbf{x}} e^{i\mathbf{k}\cdot\mathbf{x}}\mathsf{E}_n(A(\mathbf{0},t)B(\mathbf{x},0))_n^\top \\
&= \lim_{k\to 0}\lim_{t\to\infty}\frac{\partial}{\partial\theta}\left[\frac{1}{Z(\theta)}\mathsf{E}_n\left(A(t)\exp\left(\theta\sum_{\mathbf{x}} e^{i\mathbf{k}\cdot\mathbf{x}}B(\mathbf{x})\right)\right)\right]_{\theta=0} \\
&= \frac{\partial}{\partial\theta}\hat{A}(n(\theta,\hat{\mathbf{k}}))\Big|_{\theta=0} \\
&= \frac{\partial \hat{A}}{\partial n}(n)\cdot\frac{\partial n}{\partial\theta}(\theta,\hat{\mathbf{k}})\Big|_{\theta=0} \\
&= \frac{\partial \hat{A}}{\partial n}(n)\cdot\lim_{k\to 0}\sum_{\mathbf{x}} e^{i\mathbf{k}\cdot\mathbf{x}}\mathsf{E}_n(B(\mathbf{x})N(\mathbf{0}))_n^\top \\
&= \chi(n,\hat{\mathbf{k}})\frac{\partial \hat{A}}{\partial n}(n)\frac{\partial \hat{B}}{\partial n}(n).
\end{aligned} \tag{6.56}$$

(The ergodic assumption was used in the third line.)

¿From these considerations it seems reasonable to make the following *hypothesis*: namely, that $\langle A|B\rangle_{n,\hat{\mathbf{k}}}$ *is independent of* $\hat{\mathbf{k}}$ *whenever* $A,B\in\{N\}^\perp$. The motivation of this hypothesis is that only the conserved variable (here, the particle number) is expected to show long-range correlations and the orthogonal variables (the "fast" subspace) should exhibit rapid decay.This hypothesis can be restated as saying, for arbitrary variables A,B, that $\langle A|B\rangle_{n,\hat{\mathbf{k}}} - \chi(n,\hat{\mathbf{k}})\frac{\partial\hat{A}}{\partial n}(n)\frac{\partial\hat{B}}{\partial n}(n)$ will be $\hat{\mathbf{k}}$-independent. In particular, under this hypothesis the covariance of the "fast component" of the microscopic electric currrent

$$R_{ml}\equiv\frac{1}{2}\int_{-\infty}^{+\infty}dt\left[\langle J_m(t)|J_l\rangle_{n,\hat{\mathbf{k}}} - \chi(n,\hat{\mathbf{k}})\frac{\partial \hat{\mathrm{j}}_m}{\partial n}(n)\frac{\partial \hat{\mathrm{j}}_l}{\partial n}(n)\right], \tag{6.57}$$

is *constant* (i.e. $\hat{\mathbf{k}}$-independent) for $k\to 0$. We can now calculate the diffusion tensor defined in Eq.(6.49) above, as

$$\hat{\mathbf{k}}\cdot\mathbf{D}\cdot\hat{\mathbf{k}} = \lim_{k\to 0}\lim_{t\to+\infty}\frac{1}{2t}\left[\frac{1}{\chi(n,\hat{\mathbf{k}})}\sum_{\mathbf{x}} e^{i\mathbf{k}\cdot\mathbf{x}}(\hat{\mathbf{k}}\cdot\mathbf{x})^2\mathsf{E}_n(N(\mathbf{x},t)N(\mathbf{0},0))^\top - (\hat{\mathbf{k}}\cdot\mathbf{c}(n))^2\right]$$

$$
\begin{aligned}
&= \lim_{k\to 0}\lim_{t\to+\infty} \hat{k}_m\hat{k}_l\frac{1}{2t}\int_0^t ds\int_0^t ds' \times \left[\frac{1}{\chi(n,\hat{\mathbf{k}})}\sum_{\mathbf{x}} e^{i\mathbf{k}\cdot\mathbf{x}}\mathsf{E}_n(J_m(\mathbf{x},s)J_l(\mathbf{0},s'))^\top - c_m(n)c_l(n)\right] \\
&= \hat{k}_m\hat{k}_l\cdot\lim_{k\to 0}\frac{1}{2}\int_{-\infty}^{+\infty} dt\left[\frac{1}{\chi(n,\hat{\mathbf{k}})}\sum_{\mathbf{x}} e^{i\mathbf{k}\cdot\mathbf{x}}\mathsf{E}_n(J_m(\mathbf{x},t)J_l(\mathbf{0},0))^\top - c_m(n)c_l(n)\right], \qquad (6.58)
\end{aligned}
$$

or,

$$
\hat{\mathbf{k}}\cdot\mathbf{D}(n)\cdot\hat{\mathbf{k}} = \frac{\hat{\mathbf{k}}\cdot\mathbf{R}(n)\cdot\hat{\mathbf{k}}}{\chi(n,\hat{\mathbf{k}})}. \qquad (6.59)
$$

Observe that Eq.(6.58) is precisely a microscopic version of the FDR Eq.(3.67) obtained at the macroscopic Langevin level in Section 3.3. It is a weaker version of the Price noise-diffusion relation consistent with the long-range density correlations in the steady-state.

Acknowledgements. We thank A. Asselah, R. Esposito, M. K.-H. Kiessling, Y. Oono, E. Speer, H.-T. Yau, and R. K. P. Zia for conversations on the subject of this work. The research was supported in part by AFOSR Grant AF-0115, 4–28296, by NSF–DMR–134244–20946, and by a Max Planck Research Award.

References

[1] H. Mori, Phys. Rev. **111** 694 (1958); **112** 1829 (1958).

[2] D. N. Zubarev, *Nonequilibrium Statistical Thermodynamics.* (Consultants Bureau, New York, 1974).

[3] J. A. McLennan, *Introduction to Nonequilibrium Statistical Mechanics.* (Prentice Hall, Englewood Cliffs, NJ, 1989).

[4] Y. G. Sinai, Selecta Mathematica Sovietica **7** 279 (1988)

[5] G. Eyink, "Nonequilibrium Statistical Distributions," unpublished.

[7] D. N. Zubarev and V. G. Morozov, Physica A **120** 411 (1983).

[8] R. Zwanzig, J. Chem. Phys. **33** 1338 (1960).

[9] A. Einstein, Ann. Phys. (Leipzig) **22** 180 (1907); **33** 1275 (1910).

[10] L. Onsager, Phys. Rev. **37** 405 (1931); **38** 2265 (1931).

[11] G. H. Wannier, Phys. Rev. **83** 281 (1951); **87** 795 (1952).

[12] G. H. Wannier, Bell. Syst. Techn. Journ. **32** 170 (1953).

[13] P. A. Markowich, C. A. Ringhofer, and C. Schmeisser, *Semiconductor Equations.* (Springer, Wien, 1990).

[14] S. Katz, J. L. Lebowitz, and H. Spohn, J. Stat. Phys. **34** 497 (1984)

[15] B. Schmittmann and R. K. P. Zia, "Statistical Mechanics of Driven Diffusive Systems," to appear in: *Phase Transitions and Critical Phenomena.* eds. C. Domb and J. L. Lebowitz (Academic, London, 1995).

[16] R. Esposito, R. Marra and H. T. Yau, "Diffusive Limit of Asymmetric Simple Exclusion." In: *The State of Matter,* eds. M. Aizenmann, H. Araki. (World Scientific, Singapore, 1994).

[17] C. Landim, S. Olla, and H.-T. Yau, "First-order correction for the hydrodynamic limit of asymmetric simple exclusion processes in dimension $d \geq 3$," preprint, Ecole Polytechnique, R. I. No. 307, Nov. 1994.

[18] V. P. Kalashnikov, Phys. Lett. A **26** 433 (1968).

[19] N. I. Chernov, G. L. Eyink, J. L. Lebowitz, Y. G. Sinai, Commun. Math. Phys. **154** 569 (1993).

[20] H. Spohn, *Large Scale Dynamics of Interacting Particles.* (Springer-Verlag, New York, 1991).

[21] P. J. Price, in: *Fluctuation Phenomena in Solids*, ed. R. E. Burgess. (Academic Press, New York, 1965), p.355.

[22] L. P. Kadanoff and P. C. Martin, Ann. Phys. **24** 419 (1963).

[23] D. Forster, *Hydrodynamic Fluctuations, Broken Symmetry, and Correlation Functions.* (Benjamin, Reading, MA, 1975).

[24] R. Graham, Z. Physik B **26** 397 (1977).

[25] R. Graham, Z. Physik B **26** 281 (1977).

[26] H.-O. Georgii, *Gibbs Measures and Phase Transitions.* (Walter de Gruyter, Berlin, 1988).

[27] L. D. Landau and E. M. Lifshitz, *Electrodynamics of Continuous Media* 1st ed. (Pergamon, London, 1960).

[28] S. R. de Groot and P. Mazur, *Nonequilibrium Thermodynamics.* (North-Holland, Amsterdam, 1962).

[29] H. B. Callen, *Thermodynamics.* (John-Wiley and Sons, New York, 1960).

[30] P. L. Garrido, J. L. Lebowitz, C. Maes, and H. Spohn, Phys. Rev. A **42** 1954 (1990).

[31] J. L. Lebowitz and R. H. Schonmann, Prob. Th. Rel. Fields **77** 49 (1988).

[32] R. S. Ellis, *Entropy, Large Deviations, and Statistical Mechanics.* (Springer, New York, 1985).

[33] H. Künsch, Z. Wahr. v. Geb. **66** 407 (1984).

[34] G. L. Eyink, "Entropy, Statistical Mechanics, and PDE's," unpublished.

[35] H. van Beijeren, J. Stat. Phys. **35** 399 (1984).

[36] L. Onsager and S. Machlup, Phys. Rev. **91** 1505 (1953).

[37] G. L. Eyink, J. Stat. Phys. **61** 533 (1990).

[38] H. Haken, *Synergetics.* (Springer-Verlag, Berlin, 1978).

[39] J. W. Dufty and J. M. Rubí, Phys. Rev. A **36** 222 (1987).

[40] J. A. Krommes and G. Hu, Phys. Fluids B **5** 3908 (1993).

[41] L. D. Landau and E. M. Lifshitz, *Fluid Mechanics.* (Pergamon, London, 1959).

[42] R. Stratonovich, *Nonlinear Nonequilibrium Thermodynamics I.* (Springer, Berlin, 1992).

[43] A. Einstein, Ann. Phys. (Leipzig) **17** 549 (1905).

[44] B. Callen and T. A. Welton, Phys. Rev. **83** 34 (1951).

[45] H. Nyquist, Phys. Rev. **32** 110 (1928).

[46] R. F. Fox and G. E. Uhlenbeck, Phys. Fluids **13** 1893 (1970).

[47] K. Tomita and H. Tomita, Prog. Theor. Phys. **51** 1731 (1974)

[48] S. V. Gantsevich, V. L. Gurevich, and R. Katilius, Riv. Nuov. Cim. **2** 1 (1979).

[49] A.-M. S. Tremblay, in: *Recent Developments in Nonequilibrium Thermodynamics.* Lecture Notes in Physics, 199. (Springer, Berlin, 1984).

[50] M. Q. Zhang, J. S. Wang, J. L. Lebowitz and J. L. Valles, J. Stat. Phys. **52** 1461 (1988).

[51] R. K. P. Zia and B. Schmittmann, "On Singularities in the Disordered Phase of a Driven Diffusive System," preprint.

[52] A. Aharony, Phys. Rev. B **8** 3363 (1973).

[53] J. Skalyo, B. C. Frazer, and G. Shirane, Phys. Rev. B **1** 278 (1970).

[54] E. R. Speer, "The Two Species, Totally Asymmetric Simple Exclusion Process." In: *On Three Levels,* eds. M. Fannes, C. Maes, and A. Verbeure. NATO ASI Series, Vol. 324, p.91. (Plenum Press, New York, 1994).

[55] B. Derrida, S. Janowsky, J. L. Lebowitz and E. Speer, Europhys. Lett. **22** 651 (1993); J. Stat. Phys. **73** 813 (1993).

[56] B. M. Law, R. W. Gammon, and J. V. Sengers, Phys. Rev. Lett. **60** 1554 (1988).

[57] V. G. Morozov, Physica A, **126** 461 (1984).

[58] R. Graham and H. Haken, Z. Phys. **243** 289 (1971); **245** 141 (1971).

[59] C. Kipnis, S. Olla, and S. R. S. Varadhan, Commun. Pure Appl. Math. **XLII** 243 (1989).

[60] R. Graham, in: *Order and Fluctuations in Equilibrium and Nonequilibrium Statistical Mechanics*, G. Nicolis, G. Dewel, and J. W. Turner, eds. (Wiley, New York, 1981).

[61] R. Graham, in: *Stochastic Processes in Nonequilibrium Systems.* Sitges 1978, eds. L. Garrido, P. Seglar, and P. J. Shepherd. (Springer, Berlin, 1978).

[62] M. I. Freidlin and A. D. Wentzell, *Random Perturbations of Dynamical Systems.* (Springer, New York, 1984).

[63] Y. Oono, Prog. Theor. Phys. Suppl. **99** 165 (1989).

[64] E. B. Pitman and D. G. Schaeffer, Commun. Pure Appl. Math. **40** 421 (1987).

[65] A. De Masi, E. Presutti, and J. L. Lebowitz, J. Stat. Phys. **55** 523 (1986).

[66] G. Jona-Lasinio, C. Landim, and M. E. Vares, Prob. Theor. Rel. Fields **97** 339 (1993).

[67] G. Jona-Lasinio, Ann. Inst. H. Poincaré **55** no. 2, 751 (1991).

[68] R. Balescu, *Transport Processes in Plasmas*, 2 vols. (North-Holland, Amsterdam, 1988).

[69] R. Balescu, Phys. Fluids B **3** 564 (1991).

[70] A. van Enter, R. Fernández, and A. Sokal, J. Stat. Phys. **72** 879 (1993).

[71] T. J. Liggett, *Interacting Particle Systems.* (Springer, Berlin, 1985).

[72] F. Rezakhanlou, Commun. Math. Phys. **140** 417 (1991).

[73] H. van Beijeren, R. Kutner, and H. Spohn, Phys. Rev. Lett. **54** 2026 (1985).

[74] J. Krug and H. Spohn, "Kinetic Roughening of Growing Surfaces." In: *Solids Far From Equilibrium: Growth, Morphology and Defects,* ed. C. Godréche. (Cambridge University Press, Cambridge, 1991).

[75] Lin Xu, "Diffusion Limit for Lattice Gas With Short-Range Interactions," Thesis, NYU, 1993.

[76] H. O. Georgii, *Canonical Gibbs Measures.* Lecture Notes in Mathematics, vol. 760. (Springer-Verlag, Berlin, 1979).

[77] R. H. Kraichnan, Phys. Rev. **113** 1181 (1959).

[78] G. Gallavotti and E. Verboven, Nuovo Cim. **28** 274 (1975).

[79] M. Aizenman et al., Commun. Math. Phys. **53** 209 (1977).

[80] M. Aizenman et al., Commun. Math. Phys. **48** 1 (1976).