

## Long-range correlations in diffusive systems away from equilibrium

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We study the dynamics of density fluctuations in purely diffusive systems away from equilibrium. Under some conditions the static density correlation function becomes long ranged. We then analyze this behavior in the framework of nonequilibrium fluctuating hydrodynamics.

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### I. INTRODUCTION

Recently, there has been a great interest in the study of long-range spatial correlations in nonequilibrium systems. A wide variety of models governed by Langevin-like equations has been proposed. A first class of models has been introduced to describe systems, in the presence of external noise sources, in which the fields are conserved only in average. As an example we could mention models of sandpiles [1] or diffusion in disordered media [2,3], which display long-range order. A second group of models would account for situations in which internal noise sources are present and the total values of the fields are conserved. Among them one finds turbulence models in fluids [4], continuum models for interface-growing or directed polymers [5], and some models for driven-diffusive systems [1,6]. In them, although fluctuations have an internal origin, they are not necessarily related to dissipation. Whereas in the former long-range behavior is always observed, in the latter long-range correlations appear when some intrinsic anisotropy is present.

Hydrodynamic systems constitute a large class of systems which also presents internal fluctuations coming from the presence of microscopic degrees of freedom. The existence of a fluctuation-dissipation theorem introduces new aspects in the behavior of the correlations. In particular, it is shown that the form of the fluctuation-dissipation theorem is determinant in order to produce such correlations. In this sense, a preliminary result was found for thermal conducting systems [7] where the temperature correlation function decays as  $s^{-1}$ ,  $s$  being the distance between the points in consideration [see Eq. (23)]. Such long-range correlations have been observed experimentally in liquids under thermal gradients [8].

Our purpose in this paper is to analyze the origin of long-range correlations in purely diffusive systems, described only by one variable: the number density of particles. In these systems diffusion is the only mechanism responsible for the maintenance of nonequilibrium steady states. Therefore, we are not considering the possibility of applying external forces (as, for example, electric fields) which could be the origin of long-range correlations even if the stationary state is homogeneous [9]. The dynamics of fluctuations around nonequilibrium steady states is dictated by nonequilibrium fluctuating hydro-

namics. We can imagine a general situation in which the diffusion coefficient may depend on the position. This is what happens, for example, when diffusion takes place in a suspension of particles in the presence of hydrodynamic interactions or in an inhomogeneous medium. In the first case the diffusion coefficient is not constant due to the local concentration value, taking into account the fact that the distance among the particles can hinder their movement, while in the second the diffusion coefficient depends on the position through the inherent properties of the medium where the particles are diffusing. While in the first system we obtain long-range correlations, in the second density fluctuations are  $\delta$  correlated.

The paper is organized as follows. In Sec. II we introduce the models. We specify the steady states and the fluctuation evolution equations. These results enable us, in Sec. III, to determine the static correlation functions. Finally, in Sec. IV, we discuss the origin of the long-range terms in the framework of nonequilibrium fluctuating hydrodynamics.

### II. DIFFUSIVE SYSTEMS

Our purpose in this section is to study the fluctuation dynamics of purely diffusive systems, which will be characterized by the number density field,  $n(\vec{r}, t)$ . According to nonequilibrium thermodynamics [10], this quantity is governed by the continuity equation

$$\frac{\partial n}{\partial t} + \vec{\nabla} \cdot \vec{J} = 0, \quad (1)$$

where  $\vec{J}$  is the number density flux, given by Fick's law

$$\vec{J} = \underline{D} \cdot \vec{\nabla} n, \quad (2)$$

with  $\underline{D}$  being the diffusion tensor, which becomes a scalar for isotropic systems. To analyze the fluctuation dynamics, we will next study two different models with particular expressions for the diffusion coefficient.

#### A. Model A

As a first model, we will consider diffusion in a system in which the diffusion coefficient depends on the lo-

cal number density, as happens, for example, when hydrodynamic interactions among suspended particles are considered [11].

The differential equation governing the evolution of the number density of particles results from the continuity equation and the linear law for the mass flux, introduced previously. Since the diffusion coefficient is not constant, this equation reads

$$\frac{\partial n}{\partial t} = \vec{\nabla} \cdot [D(n)\vec{\nabla}n], \quad (3)$$

where we have taken into account the fact that the diffusion coefficient depends on position through the local value of the density  $n$ . The expressions of both the stationary number density profile and diffusion coefficient are obtained from the solution of Eq. (3) in the stationary regime

$$\vec{\nabla}[D_s(n_s) \cdot \vec{\nabla}n_s] = 0. \quad (4)$$

We will analyze the stationary solutions of (4) in the situation in which our system fills the region between two parallel plates and a concentration gradient  $\nabla_0 n = (n_{L/2} - n_{-L/2})/L \equiv \Delta n/L$  is kept. In this last expression,  $n_{\pm L/2}$  are the values of the number density at the plates. If the dependence of  $D_s$  on  $n$  is smooth enough, we can expand both the stationary values of the diffusion coefficient and the number density in powers of the parameter  $\frac{n_0}{D_0} \left(\frac{\partial D}{\partial n}\right)_s \equiv \epsilon$ , with  $n_0$  being a characteristic value of the concentration. Up to linear order in  $\epsilon$  one obtains

$$n_s(\vec{r}) = n' + \vec{r} \cdot \vec{\nabla}_0 n + \frac{1}{2} \frac{\epsilon}{n_0} (\vec{r} \cdot \vec{\nabla}_0 n)^2, \quad (5)$$

where  $n' = \bar{n} - \frac{1}{8} \frac{\epsilon}{n_0} (\Delta n)^2$ , with  $\bar{n} = (n_{L/2} + n_{-L/2})/2$  being an averaged density value, and the expression for the diffusion coefficient in this approximation reads

$$D_s(\vec{r}) = D_0 \left[ 1 - \epsilon \frac{\bar{n}}{n_0} - \frac{\epsilon}{n_0} \vec{\nabla}_0 n \cdot \vec{r} \right]. \quad (6)$$

Note that for small  $\epsilon$ ,  $D_s$  remains always positive. Once we have obtained the stationary solutions, we proceed to study the dynamics of the fluctuations of the number density,  $\delta n \equiv n - n_s$ . According to fluctuating hydrodynamics, fluctuations follow an equation similar to (3) with stochastic sources  $\vec{J}^R(\vec{r}, t)$ . Note that care should be taken because the fluctuation dynamics will be affected due to the fact that the diffusion coefficient is a fluctuating quantity. After linearization in the expansion parameter (an analysis of the case in which mode-coupling terms are present has been given in [12] for a fluid in

stationary flow), and introducing the expression (6), one obtains

$$\begin{aligned} \frac{\partial \delta n}{\partial t} = & D_0 \left( 1 - \epsilon \frac{\bar{n}}{n_0} \right) \nabla^2 \delta n \\ & - D_0 \nabla^2 \left( \frac{\epsilon}{n_0} \vec{r} \cdot \vec{\nabla}_0 n \delta n \right) - \vec{\nabla} \cdot \vec{J}^R. \end{aligned} \quad (7)$$

The random part of the stochastic current satisfies the fluctuation-dissipation theorem

$$\begin{aligned} \langle \vec{J}^R(\vec{r}, t) \cdot \vec{J}^R(\vec{r}', t') \rangle \\ = 2k_B T \frac{\partial n}{\partial \mu} \Big|_s D_s(\vec{r}) \delta(\vec{r} - \vec{r}') \delta(t - t') \underline{1}, \end{aligned} \quad (8)$$

where  $\underline{1}$  is the unit matrix and the average is taken over a stationary ensemble. Therefore,  $\delta n$  describes a Gaussian process [13]. In the above expression, the diffusion coefficient is precisely (6) and the derivative  $\frac{\partial n}{\partial \mu} \Big|_s$  must be evaluated in the steady state. For ideal systems ([8]) reduces to

$$\begin{aligned} \langle \vec{J}^R(\vec{r}, t) \cdot \vec{J}^R(\vec{r}', t') \rangle \\ = 2k_B T n_s(\vec{r}) D_s(\vec{r}) \delta(\vec{r} - \vec{r}') \delta(t - t') \underline{1}, \end{aligned} \quad (9)$$

which constitutes a good approximation in the case of dilute systems.

The formal solution for the number density fluctuations, Eq. (7), in Fourier space is given by

$$\delta n(\vec{k}, \omega) = -iG(\vec{k}, \omega) \vec{k} \cdot \vec{J}^R(\vec{k}, \omega), \quad (10)$$

where we have defined the Green propagator

$$G(\vec{k}, \omega) = \frac{1}{-i\omega + D_0 \left( 1 - \epsilon \frac{\bar{n}}{n_0} \right) k^2 - i \frac{\epsilon}{n_0} k^2 \vec{\nabla}_0 n \cdot \vec{\nabla}_{\vec{k}}}, \quad (11)$$

with  $\vec{\nabla}_{\vec{k}}$  being the gradient with respect to the components of the vector  $\vec{k}$ .

Note that in the absence of inhomogeneities ( $\epsilon = 0$ ), this propagator reduces to

$$G(\vec{k}, \omega) = G_0(\vec{k}, \omega) = \frac{1}{-i\omega + D_0 k^2}, \quad (12)$$

which describes the fluctuation dynamics of diffusion processes when the diffusion coefficient is a constant. The propagator  $G(\vec{k}, \omega)$  can also be expanded in powers of  $\epsilon$ . To linear order one gets

$$G(\vec{k}, \omega) = G_0(\vec{k}, \omega) \left[ 1 + \epsilon \frac{D_0 \bar{n}}{n_0} k^2 G_0(\vec{k}, \omega) + i \frac{\epsilon}{n_0} G_0(\vec{k}, \omega) k^2 \vec{\nabla}_0 n \cdot \vec{\nabla}_{\vec{k}} - 2i \epsilon \frac{D_0}{n_0} G_0(\vec{k}, \omega)^2 \vec{\nabla}_0 n \cdot \vec{k} \right], \quad (13)$$

where we have considered that  $G(\vec{k}, \omega)$  is an operator. This expression will be used in the next section to compute correlation functions.

### B. Model B

We now consider a different model, in which the diffusion process takes place in an inhomogeneous medium in which the diffusion coefficient may depend on the position according to the expression

$$D(\vec{r}) = D_0 + \vec{\nabla}_0 D \cdot \vec{r}. \quad (14)$$

Here  $D(\vec{r})$  is the diffusion coefficient,  $D_0$  is a constant, and  $\vec{\nabla}_0 D$  is a constant vector that will be determined by boundary conditions. Variations of the diffusion coefficient are limited to ensure the positive character of such a quantity. The former expression could be interpreted as an expansion in the parameter  $L \frac{|\vec{\nabla}_0 D|}{D_0}$ , with  $L$  being a characteristic length of the system, in which higher-order powers of the parameter have been neglected.

The one-dimensional microscopic version of this model could be a particle performing a random walk in the presence of traps whose distribution depends on position [14]. It is possible to show that the probability distribution satisfies a Fokker-Planck equation in which one may identify the position-dependent diffusion coefficient as a function of the probability distribution of the traps. If this distribution varies slowly with the position, one obtains an expression similar to (14).

As in model A, the number density evolves according to

$$\frac{\partial n}{\partial t} = \vec{\nabla} \cdot D(\vec{r}) \vec{\nabla} n. \quad (15)$$

We will analyze the stationary solutions of (15) subjected to the same boundary conditions as in the previous model. To linear order in  $\Delta D/D_0$ , we get

$$n_s(\vec{r}) = \tilde{n} + \vec{r} \cdot \vec{\nabla}_0 n - \frac{1}{2D_0} \vec{r} \cdot \vec{\nabla}_0 n (\vec{r} \cdot \vec{\nabla}_0 D), \quad (16)$$

where we have defined  $\tilde{n} \equiv \bar{n} - \frac{1}{8} \frac{\Delta n \Delta D}{D_0}$ .

We now proceed to study the dynamics of the number density fluctuations. Note that fluctuation dynamics in this model will differ from (7) because now the diffusion coefficient does not fluctuate. To linear order in the expansion parameter one obtains

$$\begin{aligned} \frac{\partial \delta n}{\partial t} = & D_0 \nabla^2 \delta n + \vec{r} \cdot \vec{\nabla}_0 D \nabla^2 \delta n \\ & + \vec{\nabla}_0 D \cdot \vec{\nabla} \delta n - \vec{\nabla} \cdot \vec{J}^R, \end{aligned} \quad (17)$$

where use has been made of Eq. (14). Here,  $\vec{J}^R$  is again the stochastic flux which satisfies the fluctuation-dissipation theorem formulated in Eq. (9) since the system is ideal. In it, the diffusion coefficient is now given by Eq. (14).

The formal solution for the number density fluctuations in Fourier space also follows from Eq. (10), with the appropriate Green function, which now reads

$$G(\vec{k}, \omega) = \frac{1}{-i\omega + D_0 k^2 + ik^2 \vec{\nabla}_0 D \cdot \vec{\nabla}_{\vec{k}} + i\vec{\nabla}_0 D \cdot \vec{k}}. \quad (18)$$

This expression differs from (11) because now the diffusion coefficient does not fluctuate. In the same way as we have done in model A, the propagator can be expanded in powers of  $\frac{\Delta D}{D_0}$ .

### III. CORRELATION FUNCTIONS

To compute the number density correlation function, for model A, we will use (10) and the expression for the fluctuation-dissipation theorem (9), in  $(\vec{k}, \omega)$  representation. We then obtain

$$\begin{aligned} \langle \delta n(\vec{k}, \omega) \delta n(\vec{k}', \omega') \rangle = & -G(\vec{k}, \omega) G(\vec{k}', \omega') \vec{k} \cdot \vec{k}' [D_0 n' + in' \epsilon \nabla_0 n \cdot \vec{\nabla}_{\vec{k}} \\ & + iD_0 \epsilon \nabla_0 n \cdot \vec{\nabla}_{\vec{k}} - \frac{1}{2} \vec{\nabla}_0 n \vec{\nabla}_0 n : \vec{\nabla}_{\vec{k}} \vec{\nabla}_{\vec{k}}] \delta(\vec{k} + \vec{k}'), \end{aligned} \quad (19)$$

where, due to the convolution in real space,  $G(\vec{k}, \omega)$  should be understood as an operator. The equal-time correlation function in real space follows after Fourier transforming (19) according to

$$\langle \delta n(\vec{r}, t), \delta n(\vec{r}', t) \rangle = \frac{1}{(2\pi)^8} \int_{-\infty}^{\infty} d\vec{k} e^{i\vec{k} \cdot \vec{r}} \int_{-\infty}^{\infty} d\vec{k}' e^{i\vec{k}' \cdot \vec{r}'} \left\{ \int_{-\infty}^{\infty} d\omega e^{-i\omega t} \int_{-\infty}^{\infty} d\omega' e^{-i\omega' t} \langle \delta n(\vec{k}, \omega) \delta n(\vec{k}', \omega') \rangle \right\}. \quad (20)$$

As we have a finite system, one should have used Fourier series in the spatial direction in which the external gradient is applied. This fact means that our expressions will be restricted for points  $\vec{r}$  and  $\vec{r}'$  such that  $|\vec{r} - \vec{r}'| \ll 2L/\pi$ , with  $L$  being the size of the system. Otherwise, the discrete character of the reciprocal space has to be taken into account [15]. Performing the integrals, we finally arrive at

$$\begin{aligned} \langle \delta n(\vec{r}, t) \delta n(\vec{r}', t) \rangle = & n_s(\vec{r}) \delta(\vec{r} - \vec{r}') \\ & + \frac{\Gamma(d-2)}{2^d \pi^{\frac{d-2}{2}} \Gamma(\frac{d}{2})} \frac{\epsilon}{n_0} \frac{|\vec{\nabla}_0 n|^2}{|\vec{r} - \vec{r}'|^{d-2}} \end{aligned} \quad (21)$$

to linear order in  $\epsilon$ . In the former expression  $\Gamma(x)$  is the Euler function, and it has been given for dimensions  $d > 2$ . When  $d=2$  logarithmic divergencies appear. As

expected, Eq. (21) reduces to its equilibrium counterpart when the gradient is turned off.

For the sake of simplicity, the long-range contribution in Eq. (21) has been obtained keeping only the first term in the virial expansion of the derivative  $\partial n/\partial\mu$ , which corresponds to the ideal case. Potential interactions between particles would introduce additional terms in  $\partial n/\partial\mu$  [11], but long-range behavior is still present.

Following the same line of reasoning, we can also compute the correlation function for model B introduced in the previous section. Up to linear order in  $|\nabla_0 D|$ , we arrive at the stationary function

$$\langle \delta n(\vec{r}, t) \delta n(\vec{r}', t) \rangle = n_s(\vec{r}) \delta(\vec{r} - \vec{r}') . \quad (22)$$

which does not exhibit long-range correlations. This expression differs from the equilibrium result since now the local number density enters into the correlation. Equation (21) reduces to this expression as well, if a constant diffusion coefficient is considered.

At this point it is worth comparing the above results with the exact ones obtained for a thermal diffusive system [7]. Although both processes are diffusive and consequently, at the deterministic level, they are described by the same equations, one should emphasize the different behavior between concentration and temperature correlation functions in the presence of external gradients. Indeed, while the former does not always exhibit long-range behavior, in the latter long-range correlations are always present. The equal-time temperature correlation function has been found to be

$$\begin{aligned} \langle \delta T(\vec{r}, t) \delta T(\vec{r}', t) \rangle \\ = \frac{k_B}{\rho c_v} \left\{ T_s^2(\vec{r}) \delta(\vec{r} - \vec{r}') + \frac{|\vec{\nabla}_0 T|^2}{4\pi|\vec{r} - \vec{r}'|} \right\} . \quad (23) \end{aligned}$$

where  $T_s(\vec{r})$  is the stationary linear temperature profile,  $\vec{\nabla}_0 T$  the external temperature gradient,  $k_B$  Boltzmann's constant,  $\rho$  the density, and  $c_v$  the specific heat at constant volume. Note that Eqs. (21) and (23) have the same form, which indicates that the modifications in the propagator induced by the inhomogeneities in the diffusion coefficient do not modify the long-range behavior of the correlations.

From expression (21) it is straightforward to compute the static structure factor, which for  $d=3$  is

$$S(\vec{k}) = n_s(\vec{k}) + \frac{\epsilon}{n_0} \frac{|\nabla_0 n|^2}{k^2} . \quad (24)$$

An estimation of the nonlocal contribution,  $S_{nl}$ , of Eq. (24), given through the second term of its right-hand side, relative to its equilibrium value,  $S_{eq}$ , follows from

$$\frac{S_{nl}(k)}{S_{eq}(k)} = \frac{\epsilon |\nabla_0 n|^2}{n_0^2 k^2} . \quad (25)$$

In fluid systems, the concentration gradient will induce a coupling between concentration and velocity fluctuations [16]. This fact leads to a nonlocal term, propor-

tional to  $k^{-4}$ , which is more important than the above mentioned correction. However, if the wave vector is parallel to the external concentration gradient, this coupling may be avoided [17]. For this geometry,  $S_{nl}$  given through Eq. (25), is the only nonlocal correction to the static structure factor. For a colloidal suspension of hard spheres the quantity (25) is proportional to the volume fraction [11] and to the ratios  $\Delta n/n_0$  and  $(kL)^{-1}$ .

#### IV. DISCUSSION

We have shown that purely diffusive systems away from equilibrium may exhibit long-range correlations. In order to clarify the origin of such correlations we will first analyze in detail the nature of the Langevin sources.

Our starting point is the expression for the entropy production corresponding to an irreversible vectorial process, which is given as a product of a flux-force pair [18]. Denoting the flux by  $\vec{J}$  and the force by  $\vec{X}$ , one has

$$\sigma = \vec{J} \cdot \vec{X} \quad (26)$$

from which one formulates the linear law

$$\vec{J} = \underline{L} \cdot \vec{X} , \quad (27)$$

where  $\underline{L}$  is the matrix of phenomenological coefficients.

In the framework of fluctuating hydrodynamics, fluctuations are incorporated simply by adding stochastic sources to the currents. One has

$$\vec{J} = \underline{L} \cdot \vec{X} + \vec{J}^R , \quad (28)$$

where the stochastic current, denoted in general by  $\vec{J}^R$ , introduces a Gaussian white noise stochastic process with zero mean and whose correlations are given by

$$\langle \vec{J}^R(\vec{r}, t) \vec{J}^R(\vec{r}', t') \rangle = \underline{\Gamma} \delta(\vec{r} - \vec{r}') \delta(t - t') . \quad (29)$$

The matrix  $\underline{\Gamma}$  is related to the matrix of phenomenological coefficients by means of the relation  $\underline{\Gamma} = 2k_B \underline{L}$ . This last expression, together with Eq. (29), constitutes the formulation of the fluctuation-dissipation theorem.

Note that, according to fluctuating hydrodynamics, the former scheme holds when fluctuations occur around equilibrium or nonequilibrium steady states. In this last case, the matrix  $\underline{\Gamma}$  may depend on the position since the matrix of phenomenological coefficients may depend on local equilibrium quantities to ensure local Gaussianity. As a first example, let us consider the diffusion of a contaminant. In this case the entropy production reads

$$\sigma = \vec{J}_D \cdot \vec{\nabla} \frac{\mu}{T} \quad (30)$$

from which one derives the linear law

$$\vec{J}_D = \frac{1}{T} \underline{L}_D \cdot \vec{\nabla} \mu . \quad (31)$$

In these two last expressions,  $\vec{J}_D$  is the diffusion current,  $\mu$  the chemical potential,  $T$  the temperature (assumed to be constant) and  $\underline{L}_D$  the corresponding ma-

trix of phenomenological coefficients. The linear law (31) can be identified with Fick's law,  $\vec{J}_D = -\underline{D} \cdot \vec{\nabla} n$ , where  $\underline{D} = \frac{1}{T} \underline{L}_D \frac{\partial \mu}{\partial n}$ . This identification enables us to formulate the fluctuation-dissipation theorem [13]

$$\underline{\Gamma} = 2k_B T \underline{D} \frac{\partial n}{\partial \mu}, \quad (32)$$

which for noninteracting particles becomes

$$\underline{\Gamma} = 2\underline{D}n. \quad (33)$$

One can also apply our former analysis to the case of heat conduction. One arrives at

$$\underline{\Gamma} = 2k_B T^2 \underline{\lambda}, \quad (34)$$

where  $\underline{\lambda}$  is the thermal conductivity related to the corresponding matrix of phenomenological coefficients  $\underline{L}_q$  through  $\underline{\lambda} = \frac{1}{T^2} \underline{L}_q$ .

In the case of heat conduction, for a constant thermal conductivity, the quadratic dependence of the matrix  $\underline{\Gamma}$  on temperature is responsible for the long-range behavior of the static correlation function shown in (23). Concerning mass diffusion, long-range correlations are also present only when the corresponding matrix  $\underline{\Gamma}$  is quadratic in the thermodynamic field. In fact, in model A one has a quadratic dependence of  $\underline{\Gamma}$  on  $n$  owing to the dependence of the diffusion coefficient on the local density field. In model B the diffusion coefficient depends on position, but not on the density, and therefore the matrix  $\underline{\Gamma}$  is linear in  $n$ .

In order to give a simple justification of the long-range behavior, we may consider the following intuitive argument proposed by Ronis [19]. If we have a single Langevin source at a certain point generating randomly hydrodynamic excitations, it will strongly correlate, at equal times, two equidistant points which are closer to this source than the typical hydrodynamic decay length. In equilibrium, as we have uncorrelated Langevin sources everywhere, all of them being equivalent in intensity and magnitude, the correlations between these two points will drop out. Out of equilibrium, however, as the Langevin forces are detuned, long-range correlations could in principle appear.

In the models we have analyzed, while the Langevin forces are detuned long-range correlations are not always present. Consequently, the previous argument should be somehow made precise. In model A, and also in thermal diffusive systems, the ratio between the intensity of the noise  $\underline{\Gamma}$  and the local value of the corresponding field is proportional to the stationary field, whereas in model B it does not depend on it. It is our contention that long-range behavior originates from the nature of the Langevin sources, which should increase their intensity relative to the local thermodynamic field, and not only from their detuned character. Moreover, the form of Eq. (21) indicates that the modifications in the propagator due to

the inhomogeneities in the diffusion coefficient and its fluctuations do not contribute to the long-range behavior of the correlations, which appear only through the above-named mechanism. Finally, note that the correlation functions decay as  $k^{-2}$  as we have purely diffusive systems and there is no coupling with the fluctuations of other hydrodynamic fields.

Long-range correlations have also been predicted in other nonequilibrium systems, as for example for fluids under temperature gradients [20], for diffusive systems in the presence of chemical reactions [13], or in semiconductors in the presence of an electric field [9]. In fluid systems, correlations between density or temperature and velocity appear out of equilibrium owing to the breaking of time reversal symmetry and have been observed experimentally in Brillouin scattering [21]. As regards Rayleigh scattering, both in simple fluids [8] and in binary mixtures [16], long-range correlations appear due to the resonant coupling between temperature or concentration fluctuations and transverse velocity fluctuations [22], which leads to the characteristic  $k^{-4}$  behavior of the static structure factor. In the remaining examples, long-range behavior is related to the existence of characteristic frequencies or lengths. Note that in our case, as in thermal diffusion, detailed balance holds even in the presence of external constraints [23].

Contrasting with our analysis, other diffusive systems have been considered in the literature, which do not satisfy a fluctuation-dissipation theorem, either because they come from an external noise source [1] or because the internal noise does not have a thermal origin [4–6]. In all these examples, the appearance of long-range correlations typically originate from some kind of essential anisotropy. However, when a fluctuation-dissipation theorem is satisfied, anisotropy does not guarantee long-range order. In fact, it is easy to see that model A with a constant but anisotropic diffusion matrix does not produce long-range correlations.

Model A is a real hydrodynamic model in which long-range correlations originate from hydrodynamic interactions between particles. A similar model has been proposed as the hydrodynamic limit of a lattice gas of interacting particles [24]. Though a constant diffusion coefficient is considered, the interaction introduces nonlinearities in the number density in the fluctuation-dissipation theorem due to the nonideal character of the system. The long-range term that is obtained can be deduced from our model A if  $\epsilon$  is set equal to zero and the next order in the virial expansion of  $\partial n / \partial \mu$  is taken into account.

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