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Higher-order hydrodynamics: Extended Fick's Law, evolution equation, and Bobylev's instability

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A higher-order hydrodynamics for material motion in fluids, under arbitrary nonequilibrium conditions, is constructed. We obtain what is a generalized—to that conditions—Fick-type Law. It includes a representation of Burnett-type contributions of all order, in the form of a continuous-fraction expansion. Also, the equation includes generalized thermodynamic forces, which are characterized and discussed. All kinetic coefficients are given as correlations of microscopic mechanical quantities averaged over the nonequilibrium ensemble, and then are time- and space-dependent as a consequence of accounting for the dissipative processes that are unfolding in the medium. An extended evolution equation for the density of particles is derived, and the conditions when it goes over restricted forms of the type of the telegraphist equation and Fick's diffusion equation are presented. © *2002 American Institute of Physics.*
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I. INTRODUCTION

Microscopic descriptions of hydrodynamics, that is, derivation of kinetic equations from classical or quantum mechanics and containing kinetic (or transport) coefficients written in terms of correlation functions, is a traditional problem of long standing. An important aspect is the derivation of constitutive laws which express thermodynamic fluxes (or currents as those of matter and of energy) in terms of appropriate thermodynamic forces (typically gradients of densities as those of matter and energy). In their most general form these laws are nonlocal in space and noninstantaneous in time. The nonlocality is usually dealt with in terms of spatial Fourier transforms, and then the laws—now expressed in reciprocal space—become dependent on wave-vector $\mathbf{Q}$. The well known expressions of classical (or Onsagerian) thermohydrodynamics is obtained performing the limit of Q going to zero (long wavelengths). They are then valid in such limit, and to go beyond it is necessary to introduce a proper dependence on Q valid, in principle, for intermediate and short wavelengths (intermediate to large wave numbers). In phenomenological theories this corresponds to go from classical (Onsagerian) irreversible thermodynamics to extended irreversible thermodynamics.[1,2] This is what has been called *generalized hydrodynamics*, that is, to go beyond traditional hydrodynamics, the latter restricted to fluctuations occurring at long wavelengths and low frequencies. This question has been extensively debated for decades by the Statistical Mechanics community. Several approaches have been used, and a description can be consulted in Chap. 6 of the classical book on the subject by Boon and Yip.[3] We do present here an approach based on a grand-canonical ensemble generalized to cover the case of systems arbitrarily away from equilibrium.

Introduction of nonlocality effects for describing motions with influence of ever decreasing wavelengths (going towards the very short limit), has been done in terms of expansions in increasing powers of the wave number which consists in what is nowadays sometimes referred to as *higher-order hydrodynamics* (HOH). Attempts to perform such expansions are the so-called Burnett and super-Burnett approaches[4] in the case of mass motion, and Guyer–Krumhansl approach[5] in the case of propagation of energy. In the present paper we concentrate the attention in the first one, namely, density motion (in a future paper we shall deal with the motion of energy).

A usual approach has been based on the moment solution procedure of the Boltzmann equation, as in the work of Hess,[6] using a higher-order Chapman–Enskog solution method. The Chapman–Enskog method provides a solution to Boltzmann equation consisting of a series in powers of the Knudsen number, say K, given by the ratio between the mean-free path of the particles and the scale of variation (relevant wavelengths in the motion) of hydrodynamic fields. Retaining the term linear in K there follows Navier–Stokes equation, the term containing K^2 introduces the so-called Burnett corrections, and the higher order ones (K^3 and up) the super-Burnett corrections.

A satisfactory development of HOH is highly desirable for covering a large class of hydrodynamic situations, and in the last instance for obtaining insights into technological and industrial processes having an associated economic interest.

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Indeed, the nonlocal terms become specially important in miniaturized devices at submicronic lengths, as for instance in hydrodynamic description of microelectronic devices[7] or in design of stratosferic planes, which fly in rarefied gases in a density regime intermediate between the independent-particle description and the purely continuous description. This is the case when the mean free path of the particles is of order of, say, 10% of the characteristic length of the device, instead of being of the order of, say, 0.1% (continuous approximation) or of 100% (independent-particle approximation). Another particular problem to it related is the one of obtaining the structure of shock waves in fluids for wide ranges of Mach numbers.[8] Moreover, Burnett approximation of hydrodynamics has been shown to provide substantial improvement on many features of the flow occurring in several problems in hydrodynamics, e.g., the case of Poiseuille flow,[9] and others.[10]

The microscopic derivation of a HOH, together with the analysis of validity of the existing theories, is still a point in question. Recently, it has been shown[11] that for the case of Maxwellian molecules, whereas Navier–Stokes approximation yields equations which are stable against small perturbations, this is not the case when are introduced Burnett corrections to the equations. It follows that small perturbations to the solution, which are periodic in the space variable with a wavelength smaller than a critical length, are exponentially unstable. This fact has been called *Bobylev's instability*. More recently, García-Colín and co-workers[12] have extended Bobylev's analysis for the case of any interaction potential, and argue that one can interpret the fact as to give a bound for the Knudsen number above which the Burnett equations are not valid. Moreover, Karlin[13] reconsidered the question looking for exact solutions to simplified models. When the linearized ten-moment Grad method is used, and the Chapman–Enskog method is applied to the model, in fact there follow instabilities in the higher-order approximations. On the other hand, resorting to the Chapman–Enskog solution for the linearized Grad ten-moment equations resummed exactly, solutions are obtained for which the stability of higher-order hydrodynamics, in various approximations, can be discussed.

In this paper we reconsider the question going beyond the earlier approaches in that: First, we use a statistical-mechanical formalism[14,15] (thus containing the quantum—or classical—microscopic dynamics and a macroscopic nonequilibrium thermodynamics,[16] with the equations of hydrodynamics following from the mechanical equations of motion averaged over the nonequilibrium statistical ensemble). Second, we obtain HOH-equations including a compact expression for the Burnett contributions of all order as an algorithm in the form of a continuous-fraction expansion, leading to a far-reaching generalization of Hess' results.

The organization of the paper is as follows: In a previous paper[17] was presented a description of a generalized nonequilibrium grand-canonical ensemble, built in the framework of a nonequilibrium ensemble formalism of a large scope. It is based on the Nonequilibrium Statistical Operator Method, with its derivation founded in a variational principle—Jaynes' Maximization of the Informational Entropy—referred to as Max-Ent-NESOM in what follows.[14,15] The formalism allows for the derivation of an associated kinetic theory, which provides for the description of the evolution of dissipative processes in highly-excited matter, and can be considered as a far-reaching generalization of Mori–Heisenberg–Langevin equations.[18] Also an associated Informational Statistical Thermo-Hydrodynamics follows,[16] which is a statistical-mechanically founded version of the so-called Mesoscopic Irreversible Thermodynamics.[19] In the present paper we further analyze such kinetic theory. Summarizing the content of this communication, (i) after some considerations on the character and properties of the transport coefficients, (ii) an expression for the flux of particles in the form of a Mori-type continuous-fraction expansion is derived, which encompasses Burnett-type contributions of all orders (in that way it is provided a higher-order hydrodynamics founded on a nonequilibrium ensemble formalism of statistical mechanics), whose summability is discussed; moreover, (iii) it is shown how the theory provides an extension for intermediate to short wavelengths, and intermediate to high frequencies, of Fick's Law for the flux of particles, and (iv) generalized thermodynamic forces, of a nonlocal character and with memory, are introduced. Furthermore, in Sec. V, is derived a generalized equation of motion for the density of particles, which, as nonlocality becomes less and less relevant, goes over the equations of evolution of extended (telegraphistlike equation) and classical (traditional Fick equation) hydrodynamics.

II. THE EQUATIONS OF EVOLUTION IN HOH

We consider the MaxEnt-NESOM kinetic theory presented in Ref. 18, hereafter referred to as I, and when equations from it will be cited the corresponding number will be preceded by the label I. However, in order to avoid the quite cumbersome expressions that will follow when using the general theory, which would somehow obscure the picture, we shall simplify the presentation attempting to provide a better clarification of the main physical characteristics. For that purpose we consider a decoupling of material and thermal motions, concentrating the analysis on the first one implying in that we are neglecting cross effects. Moreover, the Markovian limit of the theory is used.

We begin re-emphasizing a quite important point: For the description of the macroscopic state of the nonequilibrium system, one faces the question of selecting the basic set of macrovariables which describe such dissipative, in general nonhomogeneous and time-evolving, state.[14,15] This is also the case in thermodynamic theories, like Extended Irreversible Thermodynamics[1,2] Rational Thermodynamics,[20] etc., but the problem is overcome in statistical thermodynamic theories like Informational Statistical Thermodynamics.[16] Consider an open system of interacting particles under the action of external sources driving it out of a state of equilibrium with a thermal reservoir at temperature T_0. Furthermore, we admit that the system is describable in terms of individual quasiparticles, for example, the extremely important cases of radiation and solid state matter. In the latter case the lattice vibrations are described in terms of a "gas" of phonons, and the electrons are described as a "gas" of Lan-

dau quasiparticles in a mean-field approximation for the Coulomb interaction (the so-called random-phase approximation or time-dependent Hartree–Fock–Bogoliubov approximation), which, as well known, constitutes an exceptionally good description which properly accounts for all the physical properties of solid state matter.

Therefore each and all the mechanical quantities associated to this kind of systems can be expressed in terms of the reduced single-particle dynamical operator, say, $\rho_{(1)}(\mathbf{r}\sigma, \mathbf{r}'\sigma')$. Use of the second quantization is quite appropriate when dealing with interacting many-body systems, accompanied with an appropriate representation in Hilbert space of single-particle states. Hence the macroscopic state (i.e., nonequilibrium thermodynamic state) of the system is completely defined by a basic set of macrovariables which are the average over the nonequilibrium ensemble of the nonequilibrium dynamical single-particle operators. They become the components of the nonequilibrium Dirac–Landau-Wigner single-particle density matrix, namely,

$$\nu_{\mathbf{k}\sigma}(t) = \mathrm{Tr}\{a_{\mathbf{k}\sigma}^{\dagger} a_{\mathbf{k}\sigma} \rho_\epsilon(t)\}, \tag{1}$$

for the diagonal elements, and the nondiagonal ones ($\mathbf{Q} \neq \mathbf{0}$) are

$$n_{\mathbf{kQ}\sigma\sigma'}(t) = \mathrm{Tr}\{\hat{n}_{\mathbf{kQ}\sigma\sigma'} \rho_e(t)\}, \tag{2}$$

where

$$\hat{n}_{\mathbf{kQ}\sigma\sigma'} = a_{\mathbf{k}+\frac{1}{2}\mathbf{Q}\sigma}^{\dagger} a_{\mathbf{k}-\frac{1}{2}\mathbf{Q}\sigma'}, \tag{3}$$

and $\rho_\epsilon(t)$ is the MaxEnt-NESOM nonequilibrium statistical operator, $\mathbf{k}$ is the crystalline momentum, and σ the spin of the single-particle, which we take as bosons (e.g., phonons or photons) to fix a case and, as usual, $s(a^{\dagger})$ represent annihilation (creation) operators in state $\mathbf{k}\sigma$. The nonequilibrium statistical operator is given by[14–16]

$$\rho_\epsilon(t) = \exp\left\{-\hat{S}(t,0) + \int_{-\infty}^{t} dt'\, e^{\epsilon(t'-t)} \frac{d}{dt'} \hat{S}(t', t'-t)\right\}, \tag{4}$$

where $\hat{S}$ is the informational-entropy operator;[21] in this case and after disregarding the spin index $\hat{S}(t,0)$ takes the form

$$\hat{S}(t,0) = \phi(t) + \sum_{\mathbf{k}} F_{\mathbf{k}}(t) a_{\mathbf{k}}^{\dagger} a_{\mathbf{k}} + \sum_{\mathbf{k},\mathbf{Q}\neq\mathbf{0}} F_{\mathbf{kQ}}(t) \hat{n}_{\mathbf{kQ}}, \tag{5}$$

and

$$\hat{S}(t', t'-t) = \exp\left\{-\frac{1}{i\hbar}(t'-t)\hat{H}\right\} \hat{S}(t',0)$$
$$\times \exp\left\{\frac{1}{i\hbar}(t'-t)\hat{H}\right\}, \tag{6}$$

where $\hat{H}$ is the system Hamiltonian, and $\phi(t)$, $F_{\mathbf{k}}(t)$ and $F_{\mathbf{kQ}}(t)$ are the Lagrange multipliers in MaxEnt-NESOM. The function $\phi(t)$ ensures the normalization of the statistical operator, and plays the role of the logarithm of a nonequilibrium partition function. We stress that the Lagrange multipliers, which are functionals of the basic macrovariables, constitute an alternative set of nonequilibrium thermodynamical intensive variables, also completely characterizing the mac-

roscopic nonequilibrium state of the system.[15,16] It is also worth recalling that the statistical operator of Eq. (4) can be written as the composition of two terms, namely,

$$\rho_\epsilon(t) = \bar{\rho}(t,0) + \rho_e'(t), \tag{7}$$

where

$$\bar{\rho}(t,0) = \exp\{-\hat{S}(t,0)\} \tag{8}$$

is an auxiliary statistical operator (of large relevance in the derivation of the associated nonlinear quantum kinetic theory,[22] and sometimes dubbed as the "instantaneously frozen," or quasiequilibrium, statistical operator) which provides the instantaneous, at time t, nonequilibrium macroscopic state of the system, and $\rho_\epsilon'(t)$ contains the processes producing the irreversible evolution of the system.

It can be noticed that the quantities of Eq. (1) are the time-evolving populations in states $|\mathbf{k}\sigma\rangle$, and the quantities of Eq. (2) are related to spatial inhomogeneities in the system. Also, alternative descriptions of the macroscopic nonequilibrium state of the system can be introduced. One is the grand-canonical one of Ref. 17, obtained when the elements of the Dirac–Landau–Wigner single-particle density matrix in Eq. (5) are replaced by linear combinations of them, consisting of the Fourier-amplitudes of the particle and energy densities and their fluxes of all orders. As we have noted before, we will consider here only mass motion, and then we take as the basic set of macrovariables the energy (its fluxes and inhomogeneities being disregarded),

$$h(t) = \sum_{\mathbf{k}} \varepsilon_{\mathbf{k}} \nu_{\mathbf{k}}(t), \tag{9}$$

where $\varepsilon_{\mathbf{k}}$ is the single-particle energy-dispersion relation; the single-particle (Fourier-transformed) density

$$n(\mathbf{Q},t) = \sum_{\mathbf{k}} n_{\mathbf{kQ}}(t), \tag{10}$$

and its fluxes

$$I_n^{[r]}(\mathbf{Q},t) = \sum_{\mathbf{k}} u^{[r]}(\mathbf{k},\mathbf{Q}) n_{\mathbf{kQ}}(t). \tag{11}$$

The index r denotes the order (and tensorial rank) of the flux ($r=1$ is the usual vectorial one or current) and

$$u^{[r]}(\mathbf{k},\mathbf{Q}) = [\mathbf{u}(\mathbf{k},\mathbf{Q}) \cdots (r-\text{times}) \cdots \mathbf{u}(\mathbf{k},\mathbf{Q})], \tag{12}$$

with the square brackets indicating tensorial product of vectors, and where $\mathbf{u}(\mathbf{k},\mathbf{Q})$ is a generating velocity vector defined in Eq. (I.19), namely,

$$\mathbf{u}(\mathbf{k},\mathbf{Q}) = \frac{1}{\hbar}\nabla_{\mathbf{k}}\varepsilon_{\mathbf{k}} + \sum_{l=1}^{\infty} \frac{1}{(2l+1)!}\left(\frac{1}{2}\mathbf{Q}\cdot\nabla_{\mathbf{k}}\right)^{2l} \frac{1}{\hbar}\nabla_{\mathbf{k}}\varepsilon_{\mathbf{k}}. \tag{13}$$

The nonequilibrium grand-canonical statistical operator associated with this problem follows from a modified expression for the informational-entropy operator of Eq. (5), obtained by redefining the Lagrange multipliers in the form,

$$F_{\mathbf{k}}(t) = \beta(t)\varepsilon_{\mathbf{k}} + \sum_{r=0}^{\infty} F_{[r]}(t) \otimes u^{[r]}(\mathbf{k}), \tag{14}$$

$$F_{\mathbf{kQ}}(t) = \sum_{r=0}^{\infty} F_{[r]}(\mathbf{Q},t) \otimes u^{[r]}(\mathbf{k},\mathbf{Q}), \tag{15}$$

with, we recall, $\mathbf{Q} \neq \mathbf{0}$, and where $\beta(t)$ plays the role of a reciprocal of a kind of nonequilibrium temperature (usually called quasitemperature), the Lagrange multipliers with $r = 0$, $r = 1$, and $r \geqslant 2$ are those associated with the density, the first (vectorial) flux, and the higher-order fluxes, respectively, and $\otimes$ stands for fully contracted tensorial product. Hence, we do have an alternative, and also complete, description to the one provided by the ensemble characterized by the statistical operator of Eqs. (4)–(6). Moreover, the resulting auxiliary statistical operator is rewritten, as done in I, in the form given by Eq. (I.28), i.e.,

$$\bar{\rho}(t,0) = \exp\{\hat{A}(t) + \hat{B}(t)\} / \mathrm{Tr}\{\hat{A}(t) + \hat{B}(t)\}, \tag{16}$$

namely, composed of a part, $\hat{A}$, corresponding to the contribution in the homogeneous variables [i.e., the part containing the Lagrange multipliers $F_{\mathbf{k}}(t)$ of Eq. (14)], plus the part $\hat{B}$ which contains the contribution in the inhomogeneous variables [i.e., the part containing the Lagrange multipliers $F_{\mathbf{kQ}}(t)$ with $\mathbf{Q} \neq \mathbf{0}$ of Eq. (15)].

Moreover, following I, we restrict the analysis introducing a *linear approximation* in the calculations, namely, we keep only first-order contributions in the inhomogeneities, that is, in the terms with $\mathbf{Q} \neq \mathbf{0}$ which are contained in the statistical operator via the operator $\hat{B}$ of Eq. (I.29b). This implies in taking small amplitude variations of the density and its fluxes, that is, considering small inhomogeneities and weak fluxes. Using the Heims–Jaynes expansion[23] for averages in terms of the operator of Eqs. (I.28) and (I.29), up to first order in $\hat{B}$ we find that

$$I_n^{[r]}(\mathbf{Q},t) = I_{\mathrm{hs}}^{[r]}(\mathbf{Q},t)[1 - B_{\mathrm{hs}}(t)] + (\hat{S}_1(t), \hat{I}^{[r]}(\mathbf{Q})|t), \tag{17}$$

where

$$I_{\mathrm{hs}}^{[r]}(\mathbf{Q},t) = \mathrm{Tr}\{\hat{I}_n^{[r]}(\mathbf{Q}) \bar{\rho}_{\mathrm{hs}}(t,0)\} = 0, \tag{18}$$

$$B_{\mathrm{hs}}(t) = \mathrm{Tr}\{\hat{B}(t) \bar{\rho}_{\mathrm{hs}}(t,0)\} = 0, \tag{19}$$

that is, these averages over the homogeneous state characterized by

$$\bar{\rho}_{\mathrm{hs}}(t,0) = \frac{e^{\hat{A}(t)}}{\mathrm{Tr}\{e^{\hat{A}(t)}\}}, \tag{20}$$

are null, and we have introduced the correlation function

$$(\hat{S}_1(t), \hat{I}_n^{[r]}(\mathbf{Q})|t) = \mathrm{Tr}\{\hat{S}_1(t) \hat{I}_n^{[r]}(\mathbf{Q}) \bar{\rho}_{\mathrm{hs}}(t,0)\}, \tag{21}$$

where

$$\hat{S}_1(t) = \int_0^1 dx (\bar{\rho}_{\mathrm{hs}}(t,0))^x \hat{B}(t) (\bar{\rho}_{\mathrm{hs}}(t,0))^{-x}. \tag{22}$$

Therefore, in Eq. (17) the first term on the right-hand side is null and the second, once we use the explicit form of $\hat{B}$, is given by

$$I_n^{[r]}(\mathbf{Q},t) = \mathrm{Tr}\Bigg\{ \int_0^1 dx (\bar{\rho}_{\mathrm{hs}}(t,0))^x \sum_{\mathbf{k}',\mathbf{Q}' \neq 0} \sum_{r'=0}^{\infty} F_{[r']}(\mathbf{Q}',t)$$
$$\otimes u^{[r']}(\mathbf{k}',\mathbf{Q}') \hat{n}_{\mathbf{k}'\mathbf{Q}'} (\bar{\rho}_{\mathrm{hs}}(t,0))^{-x}$$
$$\times \sum_{\mathbf{k}} u^{[r]}(\mathbf{k},\mathbf{Q}) \hat{n}_{\mathbf{kQ}} \bar{\rho}_{\mathrm{hs}}(t,0) \Bigg\}, \tag{23}$$

establishing the linear relationship between the density ($r = 0$) and each flux ($r = 1, 2, ...$) with the Lagrange multipliers F. The contributions to Eq. (23) with $\mathbf{Q}' \neq \mathbf{Q}$ are null. Hence, we can rewrite Eq. (23) in the compact form,

$$I_n^{[r]}(\mathbf{Q},t) = \sum_{r'=0}^{\infty} F_{[r']}(\mathbf{Q},t) \otimes U^{[r+r']}(\mathbf{Q},t), \tag{24}$$

with the tensor U obtained by comparison with Eq. (23) [also cf. Eqs. (I.A15) and (I.A16)]. Equation (24) can in principle be inverted to have that

$$F_{[r]}(\mathbf{Q},t) = \sum_{r'=0}^{\infty} U_{[r+r']}^{(-1)}(\mathbf{Q},t) \otimes I_n^{[r']}(\mathbf{Q},t), \tag{25}$$

which give us the Lagrange multipliers in terms of the basic variables, implying to provide *nonequilibrium equations of state* (here in this linear approximation), once we recall that the Lagrange multipliers are the differential coefficients of the informational-statistical entropy, namely,[16]

$$F_{[r]}(\mathbf{Q},t) = \frac{\delta \bar{S}(t)}{\delta I^{[r]}(\mathbf{Q},t)} = \frac{\delta}{\delta I^{[r]}(\mathbf{Q},t)} \mathrm{Tr}\{\hat{S}(t,0) \rho_\epsilon(t)\}, \tag{26}$$

where δ stands for variational derivative.[24]

Equations (23)–(25) refer to the inhomogeneous contributions to the macrostate of the system; let us see those associated to the homogeneous part. The homogeneous basic variables are given by

$$I_n^{[r]}(t) = \sum_{\mathbf{k}} u^{[r]}(\mathbf{k}) \nu_{\mathbf{k}}(t), \tag{27}$$

where $\nu_{\mathbf{k}}(t)$ is defined in Eq. (1), and using again Heims–Jaynes expansion[23] in first (linear) order in the inhomogeneities we find that

$$\nu_{\mathbf{k}}(t) = \mathrm{Tr}\{a_{\mathbf{k}}^\dagger a_{\mathbf{k}} \bar{\rho}_{\mathrm{hs}}(t,0)\} = e^{F_{\mathbf{k}}(t)} - 1, \tag{28}$$

with $F_{\mathbf{k}}(t)$ given in Eq. (14). The population in Eq. (28) acquires, in this linear approximation, an expression which is a kind of generalized Bose–Einstein distribution depending on time through the Lagrange multiplier $F_{\mathbf{k}}(t)$, which carries the information on the evolution of the nonequilibrium state of the system.

Having then dealt with the nonequilibrium equation of state, relating basic variables and Lagrange multipliers, as given by Eq. (25), we now proceed to derive the equations of evolution for the basic variables [cf. Eqs. (I.11), (I.13), (I.14), (I.24), and the Markovian form in Eqs. (I.35)–(I.39)]. We rewrite the equations in the Markovian limit in the alternative form,[25]

$$\frac{\partial}{\partial t} I_n^{[r]}(\mathbf{Q},t) = \mathrm{Tr}\left\{ \frac{1}{i\hbar}[\hat{I}_n^{[r]}(\mathbf{Q}),\hat{H}]\rho_c(t) \right\}$$

$$\simeq i\mathbf{Q}\otimes I_n^{[r+1]}(\mathbf{Q},t)$$

$$+\left(\frac{1}{i\hbar}\right)^2 \sum_{\mathbf{Q}'}\sum_{r'=0}^{\infty}\int_{-\infty}^{0} dt'\, e^{\epsilon t'} M^{[r+r']}$$

$$\times(\mathbf{Q},\mathbf{Q}',t'-t,t)\otimes F_{[r']}(\mathbf{Q}',t), \qquad (29)$$

where

$$M^{[r+r']}(\mathbf{Q},\mathbf{Q}',t'-t,t) = \mathrm{Tr}\{[\hat{H}',\hat{I}_n^{[r]}(\mathbf{Q})]$$

$$\times[\hat{H}'(t'-t)_0,\hat{I}_n^{[r']}(\mathbf{Q}')]\bar{\rho}_{\mathrm{hs}}(t,0)\}. \qquad (30)$$

Here, $\hat{H}$ is the Hamiltonian of the system, which, according to the tenets of the formalism, is written as the addition $\hat{H}_0 + \hat{H}'$, where in $\hat{H}'$ are contained all the interactions responsible for the evolution of the dissipative processes that are developing in the system, and $\hat{H}_0$ (the nondissipative-related part or conservative part) is the so-called secular contribution.[14,15] The term after the equal sign in Eq. (29) indicates that the equations of the kinetic theory are in MaxEnt-NESOM the averages over the nonequilibrium ensemble of Heisenberg equations of motion of Quantum Mechanics. The approximate sign indicates that the calculation has been done in the Markovian approximation.[25] Moreover, M is a kinetic coefficient in the form of a correlation function over the homogeneous state involving the change in time of the fluxes due to the interactions contained in $\hat{H}'$, and where the subindex zero means evolution in the interaction representation, that is, under $\hat{H}_0$ alone; in the average over the homogeneous state only survive the terms with $\mathbf{Q}'=\mathbf{Q}$.

We stress that while the left-hand side of this equation is expressed in terms of basic variables, the right-hand side depends on the Lagrange multipliers: The set of equations can be closed by relating both types of nonequilibrium thermodynamic representations using the nonequilibrium equations of state [cf. Eqs. (24) and (25)]. The equations for the basic variables (the density and its fluxes) are obtained after using Eq. (25) to get, after going back to direct space, that

$$\frac{\partial}{\partial t} I_n^{[l]}(\mathbf{r},t) + \nabla\cdot I_n^{[l+1]}(\mathbf{r},t)$$

$$= \sum_{l'=0}^{\infty}\int d^3\mathbf{r}' L_{[l']}^{[l]}(\mathbf{r}-\mathbf{r}'|t)\otimes I_n^{[l']}(\mathbf{r},t), \qquad (31)$$

with $l=0,1,2,...$, and where L is a kernel (or kinetic coefficient) which is the Fourier antitransform of the expression given in reciprocal space by

$$L_{[l']}^{[l]}(\mathbf{Q},t) = \sum_{s=0}^{\infty}\int_{-\infty}^{t} dt'\, e^{\epsilon(t'-t)} M^{[r+r']}(\mathbf{Q},\mathbf{Q},t'-t,t)$$

$$\otimes U_{[s+l']}^{(-1)}(\mathbf{Q},t), \qquad (32)$$

after recalling that in M of Eq. (30) only survive the terms with $\mathbf{Q}'=\mathbf{Q}$.

Equation (31) has been worked out in detail in I to obtain the expression given by Eq. (I.39), which is the counterpart in the informational mechanical-statistical approach of the one derived in phenomenological mesoscopic thermodynamics.[19]

For the sake of completeness we write down the equations of evolution, the set of them in fact, for the density $l=0$, and its fluxes of all order $l=1,2,...$, namely [cf. Eq. (I.39)],

$$\frac{\partial}{\partial t} I_n^{[l]}(\mathbf{Q},t) = i\mathbf{Q}\otimes I_n^{[l+1]}(\mathbf{Q},t) + i\mathbf{Q}\otimes \mathcal{D}_{[l+1]}^{[l+1]}(\mathbf{Q},t)$$

$$+ \mathcal{A}_{[l]}^{[l]}(\mathbf{Q},t)\otimes I_n^{[l]}(\mathbf{Q},t) + i[\mathbf{Q}\mathcal{G}_{[l-1]}^{[l-1]}(\mathbf{Q},t)]$$

$$\otimes I_h^{[l-1]}(\mathbf{Q},t) + \mathcal{B}^{[l]}(\mathbf{Q},t), \qquad (33)$$

where on the right-hand side we have put into evidence the dependence on the corresponding variable $I_n^{[l]}$, the previous one in tensorial rank, $I_n^{[l-1]}$, and the following one, $I_n^{[l+1]}$, while the last term $\mathcal{B}$ contains all other contributions; the tensorial kinetic coefficients $\mathcal{D}$, $\mathcal{A}$, $\mathcal{G}$ and the contribution $\mathcal{B}$ have the cumbersome expressions given in Appendix B in I. We call the attention to a misleading sentence in I, namely, that $\mathcal{D}(\mathbf{Q},t)$ and $\mathcal{G}(\mathbf{Q},t)$ when given in a series expansion in $\mathbf{Q}$, have a first constant term equal to zero (and being composed of even powers of $|\mathbf{Q}|$). This is because we included that first constant term in $\mathcal{B}$ (what we failed to indicate), but which we now keep in $\mathcal{D}$ and $\mathcal{G}$. We recall that $\otimes$ stands for a contracted product of tensors and square brackets for direct product of tensors. In direct space Eq. (33) becomes

$$\frac{\partial}{\partial t} I_n^{[l]}(\mathbf{r},t) + \mathrm{Div}\int d^3\mathbf{r}'[\delta(\mathbf{r}-\mathbf{r}') + \mathcal{D}_{[l+1]}^{[l+1]}(\mathbf{r}-\mathbf{r}',t)]$$

$$\otimes I_n^{[l+1]}(\mathbf{r}',t)$$

$$= \int d^3\mathbf{r}' \mathcal{A}_{[l]}^{[l]}(\mathbf{r}-\mathbf{r}',t)\otimes I_n^{[l]}(\mathbf{r}',t)$$

$$- \mathrm{Grad}\int d^3\mathbf{r}' \mathcal{G}_{[l-1]}^{[l-1]}(\mathbf{r}-\mathbf{r}',t)\otimes I_n^{[l-1]}(\mathbf{r}',t)$$

$$+ \mathcal{B}^{[l]}(\mathbf{r},t), \qquad (34)$$

where, we again recall, $l=0$ is for the density [cf. Eq. (I.40a)] when the gradient of the previous tensorial quantity is of course absent, $l=1$ for the vectorial flux, and $l\geqslant 2$ for the higher-order fluxes. It can be noticed that if the tensorial coefficients $\mathcal{A}$, $\mathcal{G}$, $\mathcal{D}$ are taken as scalars, say $\mathcal{A}_{[l]}^{[l]}=\mathcal{A}_l$, etc., and it is introduced a local-in-space approximation, that is,

$$\mathcal{A}_l(\mathbf{r}-\mathbf{r}';t) = -\tau_l^{-1}\delta(\mathbf{r}-\mathbf{r}'), \qquad (35)$$

$$\mathcal{D}_{l+1}(\mathbf{r}-\mathbf{r}';t) = (\beta_{l+1}/\alpha_l)\delta(\mathbf{r}-\mathbf{r}'), \qquad (36)$$

and is taken into account that in $\mathcal{B}^{[l]}$ is present the thermodynamic force—as will be discussed later on as we proceed—and then we take

$$\mathcal{B}^{[l]}(\mathbf{r},t) = \mathrm{Grad}\int d^3r' C_l(\mathbf{r}-\mathbf{r}';t)I^{[l]}(\mathbf{r}',t), \qquad (37)$$

the relation involving the gradient operator reads as

$$C_l(\mathbf{r}-\mathbf{r}';t)+\mathcal{G}_{l-1}(\mathbf{r}-\mathbf{r}';t)=(\beta_{l-1}/\alpha_l)\delta(\mathbf{r}-\mathbf{r}'),\quad (38)$$

where we have introduced τ_l, α_l, and β_l, which are the coefficients of mesoscopic thermodynamics,[19] showing that the results of the latter are fully recovered in such approximation, with the understanding that there is a full formal equivalence of the equations for the flux of heat of Ref. 19, and those for matter that we pursue here.

It should be stressed that even though we are considering a system of bosons, the form of Eqs. (33) remains formally unaltered in the case of fermions; evidently only the explicit form of the coefficients would be altered because anticommutation relations enter in place of the commutation relations for bosons we have used. Hence, we have a complete general description of the thermohydrodynamics, in arbitrary nonequilibrium conditions, of any fluid of particles, let them be bosons or fermions. We stress once again, that Eqs. (33) and (34) are far-reaching generalizations of Mori–Heisenberg–Langevin equations, which, just because of the approximation we have introduced are linear in the basic macrovariables.

It may be noticed that in Eq. (34), on the right-hand side there are several interesting contributions: First, after the pump source is switched off, we have a term with $\mathcal{A}^{-1}$ which plays the role of Maxwell's characteristic time of extended thermodynamics,[1] second, one term involving the gradient of the flux one order smaller; and finally, the term $\mathcal{B}$ containing contributions from all the other macrovariables excluding the one of rank l being considered and the two corresponding to tensors of rank $(l-1)$ and of rank $(l+1)$. This is, as noticed, the result of the particular redistribution of terms we have introduced when writing Eqs. (33).

We proceed next to perform further analyses of the equations, showing in the process how a higher-order hydrodynamics is derived on the basis of the nonequilibrium statistical ensemble formalism provided by MaxEnt-NESOM.

III. A CONTINUOUS FRACTION EXPANSION

Consider Eqs. (33), and for the sake of simplicity, in order to avoid cumbersome expressions which would obscure the basics of the physics involved, are imposed on the system several restrictions:

(i) First, it is introduced a *truncated description* in the second-order flux, that is, we keep as basic variables the density, its first flux or current, and the second-order flux (it can be noticed that this second flux, multiplied by the mass of the particles, is related to the pressure tensor, with the diagonal components associated to the hydrodynamic pressure and the nondiagonal ones to the shear pressure).

(ii) Second, we take a *parabolic energy-dispersion relation* and then the generating velocity $\mathbf{u}(\mathbf{k},\mathbf{Q})$ of Eq. (13) becomes $\mathbf{Q}$-independent; hence the fluxes consist of the superposition of the movement of single-particle wave packets built in terms of the states $|\mathbf{k}\rangle$, and moving with group velocity $\mathbf{u}(\mathbf{k})=\hbar^{-1}\nabla_{\mathbf{k}}\varepsilon_{\mathbf{k}}$.

(iii) Third, we take the nonequilibrium homogeneous state as being in a *steady state*; this condition can be satisfied if the system is kept under the action of an external source providing a continuous excitation (i.e., pumping energy at a constant rate). Hence, all the kinetic coefficients ($\mathcal{A}$, $\mathcal{D}$, $\mathcal{G}$) become constant in time.

(iv) Fourth, *coefficients $\mathcal{B}^{[0]}$, $\mathcal{B}^{[1]}$ are neglected*, but it is kept the one associated to the second flux, namely $\mathcal{B}^{[2]}$, because it contains the most relevant contribution to the thermodynamic forces (in the long-wavelength limit provides the traditional term proportional to the gradient of the density—the proportionality factor being the diffusion coefficient given in the form of classical kinetic theory—recovering the traditional Fick's Law).

(v) Fifth, the second-rank tensorial flux is taken as *diagonal and isotropic* (meaning that we are neglecting shear effects in the pressure tensor), and in all the equations the kinetic coefficients are taken as scalars.

(vi) Sixth, the coefficients $\mathcal{A}$ are of the form $\mathcal{A}_l(\mathbf{Q},t)=\Theta_l^{-1}(\mathbf{Q},t)+i\Delta\mathcal{A}_l(\mathbf{Q},t)$, where $\Delta\mathcal{A}$ becomes *a fortiori* associated to renormalization of the kinetic coefficients and it is neglected, for the sake of simplicity, retaining only Θ which is the relevant contribution playing the role of *Maxwell's characteristic time* of extended irreversible thermodynamics.[1,2]

Hence, after transforming Fourier in the time coordinate, we do have that

$$i\omega n(\mathbf{Q},\omega)\simeq -i\mathbf{Q}\cdot\mathbf{I}_n(\mathbf{Q},\omega),\tag{39}$$

$$i\omega\mathbf{I}_n(\mathbf{Q},\omega)\simeq -i\mathbf{Q}I_{n2}(\mathbf{Q},\omega)-\Theta_1^{-1}(\mathbf{Q})\mathbf{I}_n(\mathbf{Q},\omega)$$
$$-\mathcal{G}_0(\mathbf{Q})i\mathbf{Q}n(\mathbf{Q},\omega)-\mathcal{D}_2(\mathbf{Q})i\mathbf{Q}I_{n2}(\mathbf{Q},\omega),\tag{40}$$

$$i\omega I_{n2}(\mathbf{Q},\omega)\simeq -i\mathbf{Q}\cdot\mathbf{I}_{n3}(\mathbf{Q},\omega)-\Theta_2^{-1}(\mathbf{Q})I_{n2}(\mathbf{Q},\omega)$$
$$-\mathcal{G}_1(\mathbf{Q})i\mathbf{Q}\cdot\mathbf{I}_n(\mathbf{Q},\omega)+\mathcal{B}_2(\mathbf{Q},\omega),\tag{41}$$

where, because of item (v) above, I_{n2} is a scalar (the diagonal term in $I_n^{[2]}$, and $\mathbf{I}_{n3}$ is a vector, the diagonal part of an odd-rank tensor); moreover the coefficients Θ, $\mathcal{G}$, and $\mathcal{D}$ are scalars and time-independent as noticed in item (iii) above.

We now look for an expression for the first flux $\mathbf{I}_n(\mathbf{Q},\omega)$, thus looking for a generalized Fick's Law; using Eqs. (40) and (41), after some algebra we obtain that

$$\mathbf{I}_n=\frac{\Theta_1\mathcal{G}_0}{1+i\omega\Theta_1}i\mathbf{Q}n+\frac{\Theta_1[1+\mathcal{D}_2]}{1+i\omega\Theta_1}\frac{\Theta_2}{1+i\omega\Theta_2}i\mathbf{Q}[i\mathbf{Q}\cdot\mathbf{I}_{n3}]$$
$$+\frac{\Theta_1[1+\mathcal{D}_2]\Theta_2}{(1+i\omega\Theta_2)(1+i\omega\Theta_1)}i\mathbf{Q}\mathcal{B}_2$$
$$+i\mathbf{Q}\left\{\frac{\Theta_1[1+\mathcal{D}_2]}{1+i\omega\Theta_1}\frac{\Theta_2\mathcal{G}_1}{1+i\omega\Theta_2}i\mathbf{Q}\cdot\mathbf{I}_n\right\}$$
$$\equiv\frac{\mathbf{N}(\mathbf{Q},\omega)}{1+i\omega\Theta_1}-\frac{\Theta_1[1+\mathcal{D}_2]\Theta_2\mathcal{G}_1}{(1+i\omega\Theta_1)(1+i\omega\Theta_2)}Q^2\mathbf{I}_n,\tag{42}$$

and then

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$$\mathbf{I}_n = \frac{\mathbf{N}(\mathbf{Q},\omega)}{1 + i\omega\Theta_1 + \dfrac{\xi_1 Q^2}{1 + i\omega\Theta_2}}, \tag{43}$$

where

$$\xi_1 = \Theta_1[1 + \mathcal{D}_1]\Theta_2\mathcal{G}_1, \tag{44}$$

$$\mathbf{N}(\mathbf{Q},\omega) = \Theta_1[1 + \mathcal{D}_2]\frac{\Theta_2}{1 + i\omega\Theta_2} i\mathbf{Q}[i\mathbf{Q}\cdot\mathbf{I}_{n3}]$$

$$+ \Theta_1\mathcal{G}_0 i\mathbf{Q} n + 1 + i\omega\Theta_2$$

$$\times \Theta_1[1 + \mathcal{D}_2]\Theta_2 i\mathbf{Q}\mathcal{B}_2, \tag{45}$$

and we recall that we have disregarded the term $\mathcal{B}^{[1]}$ [item (iv) above].

Let us now consider the quantity $\mathbf{N}$, working out its first and third term. On the one hand, according to Appendix A,

$$i\mathbf{Q}[i\mathbf{Q}\cdot\mathbf{I}_{n3}(\mathbf{Q},\omega)] = \mathcal{K}(\mathbf{Q})i\mathbf{Q}(i\mathbf{Q}\cdot\mathbf{I}_n(\mathbf{Q},\omega))$$

$$= -\mathcal{K}(\mathbf{Q})i\mathbf{Q}i\omega n(\mathbf{Q},\omega), \tag{46}$$

after using the equation of conservation for the density, i.e., Eq. (39). On the other hand, of the contributions present in $\mathcal{B}_2$, as given by Eq. (I.B4) in Appendix B of I, we single out the leading one consisting in the contribution which in the limit of classical hydrodynamics leads to the traditional Fick's Law. Using a simplified form of the theory, namely neglecting the terms with $\mathcal{D}$ and $\mathcal{G}$, and in terms of a particular model, we derive in Appendix B a Fick's Law in such conditions, showing how the expression for the diffusion coefficient is retrieved in the limit of classical (very long wavelengths and very low frequency) hydrodynamics. Hence the last term on the right-hand side of Eq. (45) is expressed in the form,

$$1 + i\omega\Theta_2\,\Theta_1[1 + \mathcal{D}_2]\Theta_2 i\mathbf{Q}\mathcal{B}_2 = -D_0(\mathbf{Q},\omega)i\mathbf{Q}n(\mathbf{Q},\omega), \tag{47}$$

with D_0 to be evaluated in each particular case (cf. Appendix B).

From Eqs. (43) to (47) there follows what can be considered as a *generalized Fick's Law*, or, better to say, an *extended Fick's Law*, namely,

$$\mathbf{I}_n(\mathbf{Q},\omega) = D_n(\mathbf{Q},\omega)i\mathbf{Q}n(\mathbf{Q},\omega), \tag{48}$$

where the wave-vector- and frequency-dependent diffusion coefficient is given by

$$D_n(\mathbf{Q},\omega) = \frac{\Lambda(\mathbf{Q},\omega)}{1 + \dfrac{\xi_1(\mathbf{Q})Q^2}{\Omega_1(\omega)\Omega_2(\omega)}}, \tag{49}$$

with

$$\Lambda(\mathbf{Q},\omega) = -D_0(\mathbf{Q},\omega)$$

$$-\frac{\Theta_1(\mathbf{Q})(1 + \mathcal{D}_2(\mathbf{Q}))\Theta_2(\mathbf{Q})\mathcal{K}(\mathbf{Q})}{\Omega_1(\omega)\Omega_2(\omega)}$$

$$+\frac{\Theta_1(\mathbf{Q})}{\Omega_1(\omega)}\mathcal{G}_0(\mathbf{Q}), \tag{50}$$

$$\Omega_{1(2)}(\mathbf{Q},\omega) = 1 + i\omega\Theta_{1(2)}(\mathbf{Q}), \tag{51}$$

and

$$\xi_1(\mathbf{Q}) = \Theta_1(\mathbf{Q})(1 + \mathcal{D}_1(\mathbf{Q}))\Theta_2(\mathbf{Q})\mathcal{G}_1(\mathbf{Q}). \tag{52}$$

Equation (48) tells us that the response to the perturbation, that has driven the system out of homogeneity in space, is nonlocal in space and not instantaneous in time. In fact we do have in direct space that

$$\mathbf{I}_n(\mathbf{r},t) = \int d^3r' \int_0^t dt'\, D_n(\mathbf{r}-\mathbf{r}',t-t')\nabla' n(\mathbf{r}',t'), \tag{53}$$

where we have introduced the Fourier antitransforms of the quantities involved, with the double convolution product being a result that in Eq. (48) we have an algebric product of Fourier transforms in reciprocal $(\mathbf{Q},\omega)$-space.

The nonlocality in space mirrors the dependence on wave vector $\mathbf{Q}$ in the different quantities that have appeared in the theory, and has, in the general case, a triple origin, namely,

(1) The dependence on $\mathbf{Q}$ of the kinetic coefficients $\Theta_j(\mathbf{Q})$ (Maxwell's characteristic times) and a part in $\mathcal{G}_j(\mathbf{Q})$ and $\mathcal{D}_j(\mathbf{Q})$;

(2) Parts in $\mathcal{G}_j(\mathbf{Q})$ and $\mathcal{D}_j(\mathbf{Q})$ associated with the presence of the generating velocity vector $\mathbf{u}(\mathbf{k},\mathbf{Q})$ of Eq. (13), which, as already noticed, is a kind of group velocity of the particle in state $\mathbf{k}$ depending on position as the particle moves;

(3) The explicit presence of the square modulus of Q in Eq. (43), which, we recall, is associated with the fact of having introduced a higher-order flux (in this case I_{n2}) as a basic variable (and since the coefficient ξ_1 is proportional to $\mathcal{G}_1$, it arises from the coupling with the gradient of $\mathbf{I}_n$ in the equation of motion for I_{n2}).

We recall that we have used for simplicity a parabolic energy-dispersion law, and then item 2 above does not contribute in the present case.

It can be noticed that we can write Eq. (43) as

$$\mathbf{I}_n(\mathbf{Q},\omega) = \frac{\mathbf{N}(\mathbf{Q},\omega)}{\Omega_1(\mathbf{Q},\omega)}\frac{1}{1 + \dfrac{\xi_1(\mathbf{Q})Q^2}{\Omega_1(\mathbf{Q},\omega)\Omega_2(\mathbf{Q},\omega)}}$$

$$= \frac{\mathcal{N}(\mathbf{Q},\omega)}{1 + a_1(\mathbf{Q},\omega)Q^2}, \tag{54}$$

where

$$\mathcal{N}(\mathbf{Q},\omega) = \mathbf{N}(\mathbf{Q},\omega)/\Omega_1(\mathbf{Q},\omega), \tag{55}$$

$$a_1(\mathbf{Q},\omega) = \xi_1(\mathbf{Q})/\Omega_1(\mathbf{Q},\omega)\Omega_2(\mathbf{Q},\omega). \tag{56}$$

We can see that Eq. (43) is a truncated *continuous fraction expansion* containing only a first term, as a result of the

truncation we have introduced in the set of basic variables, where we have kept the fluxes only up to the second order. As very briefly described in Appendix C a complete continuous fraction follows when introducing the complete set of fluxes; we can see that retaining the fluxes only up to order n, the continuous fraction is truncated in order $n-1$ with the last contribution being $\xi_{(n-1)}Q^2/\Omega_n\Omega_{n-1}$.

For further simplifying matters we neglect nonlocal effects arising out of the contributions of the kinetic coefficients [item (1) above], i.e., we disregard their dependence on $\mathbf{Q}$. Hence, the nonlocality is associated only with the presence of the term Q^2 in, now, $a_1(\omega)Q^2=\xi_1Q^2/\Omega_1(\omega)\Omega_2(\omega)$, which corresponds to item (3) above, namely, to have included the second order flux for the characterization of the nonequilibrium macroscopic state of the system.

The continuous fraction expansion of Appendix C derived in MaxEnt-NESOM can be put in correspondence with the one of phenomenological mesoscopic thermo-dynamics,[19,25,26] with the coefficients in the former given in the present formulation as correlation functions determined by the microscopic dynamics of the system. We can see that the equivalence of these coefficients corresponds to the relationship,

$$-\beta_l\tau_l\tau_{l+1}(\alpha_l\alpha_{l+1})^{-1}\leftrightarrow[1$$
$$+\mathcal{D}_{l+1}]\mathcal{G}_{l-1}\Theta_l\Theta_{l+1}(\Omega_l\Omega_{l+1})^{-1}, \tag{57}$$

where on the left are those given in Ref. 19 and on the right are those of Appendix C.

IV. BURNETT EQUATIONS AND BOBYLEV'S INSTABILITY

Now, whereas it is verified that

$$|a_1(\omega)|Q^2=\left|\frac{\xi_1Q^2}{\Omega_1(\omega)\Omega_2(\omega)}\right|<1, \tag{58}$$

we can write Eq. (54) in the form of the series expansion,

$$\mathbf{I}_n(\mathbf{Q},\omega)=-\mathcal{N}(\mathbf{Q},\omega)[1-a_1(\omega)Q^2+a_1^2(\omega)Q^4-\cdots]. \tag{59}$$

It can be noticed that Eq. (58) above introduces a cutoff in the value of the wave number, that is, the series expansion is valid for not too large $Q[Q^2<|a_1(\omega)|^{-1}]$, i.e., for not too short wavelengths. On the other hand, we must recall that using a less truncated description (see Appendix C), for example, to fix ideas, introducing the third-order flux $I^{[3]}$ would provide contributions quartic in Q. Then, to be consistent we must keep terms that are only quadratic in Q, implying that in Eq. (59) the series is truncated retaining only $a_1(\omega)Q^2$. On the basis of the results in Appendix C it can be noticed that, in the imposed condition of neglecting the contributions of items (1) above, an important consequence is that truncating the description in a given, say, order n of the fluxes (corresponding to a truncation in order $n-1$ of the continuous fraction), and whereas it is verified that $|a_l(\omega)|Q^2<1$ [with a_l of Eq. (C5) and disregarding its $\mathbf{Q}$-dependence], leads to a finite series, which can be consid-

ered to be correct up to a power $2(n-1)$ of the wave number Q (or inverse of wavelength λ). Hence, we do have an expression of the type;

$$\mathbf{I}_n(\mathbf{Q},\omega)=\mathcal{N}(\mathbf{Q},\omega)\left[1+\sum_{l=1}^{n-1}b_l(\omega)Q^{2l}\right], \tag{60}$$

or in direct space

$$\mathbf{I}_n(\mathbf{r},\omega)=\left[1+\sum_{l=1}^{n-1}b_l(\omega)\nabla^{2l}\right]\mathcal{N}(\mathbf{r},\omega), \tag{61}$$

with the b_l's being complicated coefficients resulting in the process of transforming the truncated continuous fraction in the series expansion.

Equation (61) resembles a Burnett (super-Burnett in fact) equation,[4,11–13] however valid, as noticed, for the range of values of Q for which $|a_l(\omega)|Q^2<1$ for any $l=1,2,...,n-1$. Such limitation has been noticed by Garcia-Colin and co-workers,[12] and ascribed equivalently in terms of the expansion in powers of the Knudsen number. To give an example, and without going into details, the condition of Eq. (59) can be shown to be verified in the case of a continuum fluid in Debye model, where $\omega(\mathbf{Q})=s|\mathbf{Q}|$, when the thermal velocity of the particles is smaller than the sound velocity s in the medium. It is worth noticing that the resulting instability of the solution, may be related to the possible occurrence of a normal emission of phonons in a kind of acoustic-phonons-Cherenkov-type effect.[27] Summarizing, we can say that in writing Burnett-type equations one faces restrictions for their correctness, that is, it is restricted the range of values of wave number (or wavelength) for which can be considered to be valid. This result, together with Bobylev's analysis,[11] suggest conjecturing that the instability may point to the emergence of complex behavior, a question worth considering.

Evidently, such conditions depend on the quantities $\mathcal{G}_j$ and Θ_j which appear in the expression for $a_j(\omega)$, besides, of course, on the range of frequencies being considered. Such quantities, as given in I, have quite general cumbersome expressions which have to be evaluated in each particular case under consideration. We discuss this point in Appendix D where a calculus for a particular model system, consisting of two ideal classical gases in mutual interaction, is given.

As already noticed in the Introduction, the question of stability of Burnett equations has been discussed by Bobylev,[11] who showed the existence of instabilities for sufficiently large Knudsen numbers (very short wavelengths). Garcia-Colin and collaborators[8] extended such analysis to more general conditions, and Karlin[13] obtained exact solutions to simplified models in terms of which he analyses the stability problem in various approximations. Particularly, Karlin conjectures that, for circumventing the noted difficulties, the expression for the stress tensor and momentum equations should present a denominator in the form of a monomial in the Laplacian; a form also suggested earlier by Rosenau.[28] This is precisely the result that we have for the first flux (which multiplied by the mass of particles is the

momentum density). Equation (54) after transformed to direct space [once it is assumed that the condition of Eq. (58) is satisfied] becomes

$$\mathbf{I}_n(\mathbf{r},\omega)=[1-a_1(\omega)\nabla^2]^{-1}\mathcal{N}(\mathbf{r},\omega),\tag{62}$$

after neglecting the dependence on space of coefficient a_1.

However, we recall that this is the result in the truncated description which contained only the second-order flux $I^{[2]}$. This can be extended by including higher-order fluxes up to, say, order R (see Appendix C), provided the relevant wavelengths satisfy the conditions that $|a_j(\omega)|Q^2<1$ for $j=1,2,...,R-1$, when we can write in direct space the expression,

$$\mathbf{I}_n(\mathbf{r},\omega)=-D(\mathbf{r},\omega)\nabla n(\mathbf{r},\omega),\tag{63}$$

with

$$D(\mathbf{r},\omega)=\Lambda(\mathbf{r},\omega)/\Delta(\mathbf{r},\omega),\tag{64}$$

$$\Delta(\mathbf{r},\omega)=1-\cfrac{a_1(\omega)\nabla^2}{1-\cfrac{a_2(\omega)\nabla^2}{1-\cfrac{a_3(\omega)\nabla^2}{\cfrac{\ddots}{1-a_{R-1}(\omega)\nabla^2}}}},\tag{65}$$

and Λ is the back-transform in space of the one in Eq. (50).

Equation (63) above has the form of a generalized Fick equation—in a higher-order hydrodynamics, and with D being a generalized space- and time-dependent diffusion coefficient—whose general expression is given in the form of the double convolution,

$$\mathbf{I}_n(\mathbf{r},t)=-\int d^3r'\int_0^t dt' D(\mathbf{r}-\mathbf{r}';t-t')\nabla' n(\mathbf{r}',t'),\tag{66}$$

with

$$D(\mathbf{r}-\mathbf{r}';\tau)=\int_{-\infty}^{\infty}d\omega e^{-i\omega\tau}\delta(\mathbf{r}-\mathbf{r}')\hat{L}(\mathbf{r},\omega)\Lambda(\mathbf{r},\omega),\tag{67}$$

and $\tau=t-t'$.

We stress at this point that a truncation in the description (i.e., keeping a certain restricted number of higher-order fluxes, what is mirrored in a truncation of the continuous fraction, as shown), requires a criterion of validity which restricts the region in wave number and frequency space where can be applied, as discussed in Ref. 28.

V. THE EQUATION OF EVOLUTION FOR THE DENSITY

Using the antitransforms to direct space and time of Eqs. (39), (40), and (41), deriving twice in time the equation for the density, once the one for the current, and using the equation for the second flux, Eq. (46), after some calculus we find that

$$\frac{\partial^3}{\partial t^3}n(\mathbf{r},t)-\frac{1}{\tau}\frac{\partial^2}{\partial t^2}n(\mathbf{r},t)+\alpha_1\frac{\partial}{\partial t}n(\mathbf{r},t)-\alpha_2\nabla^2 n(\mathbf{r},t)$$

$$-\alpha_3\nabla^2\frac{\partial}{\partial t}n(\mathbf{r},t)=0,\tag{68}$$

where

$$\frac{1}{\tau}=\frac{1}{\tau_1}+\frac{1}{\tau_2},\quad\tau_i=-\Theta_i\tag{69}$$

$$\alpha_1=\frac{1}{\Theta_1\Theta_2};\quad\alpha_2=\frac{\mathcal{G}_0}{\Theta_2}+[1+\mathcal{D}_2](v_{th}^2/3\Theta_2);$$

$$\alpha_3=\mathcal{G}_0+[1+\mathcal{D}_2](\mathcal{G}_1+\mathcal{K}),\tag{70}$$

and use was made of the result of Appendix B, namely, that $i\mathbf{Q}\mathcal{B}_2(\mathbf{Q},t)=-(v_{th}^2/3\Theta_2)i\mathbf{Q}n(\mathbf{Q},t)$.

We can see that when it is verified that $\omega\tau\ll1$, the first term (the triple derivative in time) can be neglected when compared with the next term (the second time derivative in time), and the last term (the one associated to the Burnett-type contribution) can also be neglected when compared with the previous one. Then Eq. (68) becomes a telegraphistlike (hyperbolic) equation of the form,

$$\left[\frac{1}{s^2}\frac{\partial^2}{\partial t^2}-\frac{1}{D}\frac{\partial}{\partial t}-\nabla^2\right]n(\mathbf{r},t)=0,\tag{71}$$

where

$$s^2=-\alpha_2\tau;\quad D=-\frac{\alpha_2}{\alpha_1}.\tag{72}$$

This equation corresponds to propagation of waves of the sound type, with velocity s, which has an associated dispersion relation given by

$$\lambda_\pm(\mathbf{Q})=-\frac{\gamma}{2}\pm i\sqrt{s^2Q^2-\frac{\gamma^2}{4}},\tag{73}$$

where $\gamma=s^2/D$ is the reciprocal of the lifetime of the wave.

It can be noticed that for small Q, such that $sQ\ll\gamma/2$, the solutions are $\lambda_+=-DQ^2$ and $\lambda_-=-\gamma$, corresponding, the latter to a purely decaying mode, and the former one to diffusive motion, solution of the traditional Fick's equation.

These considerations show us that with decreasing values of Q and ω, i.e., going over the classical hydrodynamic regime, the movement can be well described with an ever-decreasing number of higher-order fluxes. In the present case, the presence of the second-order flux leads to a third-degree in time equation, together with nonlocal corrections; when only the first-order flux is a basic variable, it follows the telegraphistlike equation corresponding to a particular kind of propagation of sound waves, and finally, the equation of conservation of the density together with the traditional Fick's Law produces diffusive motion.

A corollary is that the inclusion of an ever increasing number of r-order fluxes, $I^{[r]}$, describes more and more complicated motions (increasing number of hydrodynamic modes), corresponding to steeper and steeper variations in space and time.

VI. CONCLUSIONS

The powerful kinetic theory based on a generalized non-equilibrium grand-canonical ensemble, which provides the foundations for the construction of a nonclassical thermohy-

drodynamics, has been applied to the study of the motion of a fluid when in the presence of space inhomogeneities.

The generalized Mori–Heisenberg–Langevin equations that the theory provides have been used to derive a generalized Fick's Law, in which the expression for the flux of particles contains a denominator in the form of a continuous fraction, whose sucessive terms involve a coupling with all the higher-order fluxes. But, as shown, when it is performed a truncation of the set of basic variables, say retaining fluxes only up to order R, the continuous fraction also becomes truncated in the contribution of order $R-1$.

We have been able to show that the sucessive terms in the continuous fraction provide contributions to the expression for the current of particles in the form of a series of even powers of the reciprocal of the wavelength. When rewritten in direct space, these contributions become powers of the Laplacian acting on the thermodynamic force—thus giving a generalized thermodynamic force—producing what can be considered a Burnett-type equation. Thus, the theory here described allows to go beyond the classical hydrodynamic limit by covering the situations when the motion is not restricted to long wavelength and low frequencies. Taking in the general expression we have derived the classical hydrodynamic limit, that is when $\lambda \to \infty$ and $\omega \to 0$, it is recovered the traditional Fick's Law.

It emerges from the study thus presented, that a truncation of description implies into obtaining expressions that are correct only up to certain limiting values of wavelength and frequency (both are related through the dispersion relations of the corresponding normal modes of motion). On the one hand, this implies that the shorter the wavelengths (with accompanying larger frequencies) that have relevant contributions for the motion of the fluid, a proper description requires to incorporate an ever extended number of fluxes.

Equation (43) is then an extended form of Fick's Law, going beyond the classical hydrodynamic limit by incorporating—in the spirit of Extended Irreversible Thermodynamics and within this Informational-Statistical Thermodynamic approach—the fluxes (the vectorial one or current and higher-order ones) as basic variables. An extended generalization of Fick's Law, involving any number R of fluxes, is presented in Appendix C as Eqs. (C3) *et seq.*

Such generalized Fick's Law has a numerator, the vectorial one of Eq. (45), containing a term which is a multiple divergence of the tensor of rank next to the last one incorporated in the basic set [this is $\mathbf{I}_{n3}$ in Eq. (42), and $I_n^{[R+1]}$ in Appendix C]. Additional contributions are present in Eq. (45), with the second one with coefficient $\mathcal{G}_0$ (which is proportional to the square of the interactions responsible for the collisional processes) giving *a posteriori* a perturbative correction to the diffusion coefficient [cf. Eq. (72), with α_2 given in Eq. (70)], while the last one contains the main contribution to the thermodynamic force, i.e., the one that in the limit of classical hydrodynamics ($\mathbf{Q} \to \mathbf{0}, \omega \to 0$) recovers the traditional form for the flux $\mathbf{I}_n$, namely, $(v_{\text{th}}^2 \Theta_1/3) \nabla n(\mathbf{r}, t)$.

The denominator acquires the form of a continuous fraction expansion [however finite, with a cutoff term of order $R-1$ if we have truncated the description in the flux of order R in Eq. (C4), and 1 in the case of Eq. (49) when $R=2$]. This

denominator takes the value 1 in the classical hydrodynamic limit of very large wavelengths (or Q going to zero) and low frequencies (ω going to zero), and the usual Fick's Law is recovered, as already noticed.

Moreover, as we have shown, the presence of the continuous-fraction implies that when it is handled as described in Eq. (60), it produces contributions in increasing powers of the reciprocal of the wavelength. This means that the inclusion of fluxes as basic variables in the nonequilibrium thermodynamic space removes the restriction to work in the classical hydrodynamic limit, entering the domain of Extended Irreversible Thermodynamics (here done within the statistical treatment of Informational-Statistical Thermodynamics). Therefore, for the case of fluid motions where the relevant wavelengths that characterize it includes short ones, can be better described by requiring an ever-increasing dimension in the nonequilibrium thermodynamic space (additional higher order fluxes).

Once the equations are transformed back to direct space [cf. Eqs. (62)], it can be noticed that it follows an extended version of the equations of the Burnett-type. The coefficients are given as correlations, over the nonequilibrium ensemble, of mechanical quantities, being then well defined and calculable for ulterior comparison with experiment. However, care must be exercised once, as shown by Bobylev and others,[11–13] these equations may display instabilities when they are applied to the description of motions characterized by too short wavelengths. As we have shown in this paper, a truncation in the description, meaning to keep a reduced number of higher-order fluxes as basic variables, impose a restriction delimiting a range of values of wave numbers (or wavelengths) for which the equations can be applied; this requires a case by case analysis (in Ref. 28, a particular model of two classic ideal gases with mutual interaction, has been studied). Apparently an exact expansion of all Burnett contributions free of instabilities would be given by a continuous fraction expansion including all fluxes. This is then a contribution to the debated question concerning the validity of Burnett and super-Burnett expansions. Certain analysis in kinetic theory pointed to the fact that the coefficients in the expansion could be nonanalytic functions of the wave number, as $\ln Q$ or $Q^{1/2}$. But the present expansion in fluxes of all orders, in the spirit of mesoscopic thermohydrodynamics, appears as quite appropriate, with the coefficients being analytic and providing a rapid convergence. However, it must be kept in mind that, according to our results, it appears to be a general property of the expansion the existence of a cutoff wave number, determined by the characteristics of the system and its nonequilibrium macroscopic state, see for example, Eq. (58). When this property is not verified the convergence of the continuous fraction fails.

ACKNOWLEDGMENTS

APPENDIX A: DOUBLE DIVERGENCE OF THE THIRD-ORDER FLUX

Using Eq. (9b) we have that

$$i\mathbf{Q}\otimes i\mathbf{Q}\otimes I_n^{[3]}(\mathbf{Q},t)=-\sum_{\mathbf{k}}(\mathbf{Q}\cdot\mathbf{u}(\mathbf{k},\mathbf{Q}))^2\mathbf{u}(\mathbf{k},\mathbf{Q})n_{\mathbf{kQ}}(t). \tag{A1}$$

Using the Heimes–Jaynes expansion[23] in the homogeneities [cf. Eqs. (16) and (17)], in linear approximation we can express the quantity $n_{\mathbf{kQ}}(t)$ in terms of the basic macrovariables, and substitute it in Eq. (A1) to obtain

$$i\mathbf{Q}\otimes i\mathbf{Q}\otimes I_n^{[3]}(\mathbf{Q},t)=-\gamma_1(\mathbf{Q})\sum_{\mathbf{k}}\mathcal{C}(\mathbf{k},\mathbf{Q},t)$$
$$\times(\mathbf{Q}\cdot\mathbf{u}(\mathbf{k},\mathbf{Q}))^2u^{[2]}(\mathbf{k},\mathbf{Q})\cdot\mathbf{I}_n(\mathbf{Q},t), \tag{A2}$$

where the factor $\gamma_1(\mathbf{Q})$ is the proportionality coefficient between the current $\mathbf{I}_n(\mathbf{Q},t)$ and its associate Lagrange multiplier, as obtained from a proper calculation of Eq. (24)—$\mathbf{I}_n$ does not depend on the other Lagrange multipliers because the correlation is null for symmetry—and we recall that Eq. (24) is a linearized expansion. Moreover, we used that we are considering a purely longitudinal regime, i.e., the macrovariable $\mathbf{I}_n(\mathbf{Q},t)$ is parallel to the vector wave number $\mathbf{Q}$, and quantity $\mathcal{C}(\mathbf{k},\mathbf{Q},t)$ is defined in Appendix A of article I, by Eq. (I.A13).

Using the definition of $\mathbf{u}(\mathbf{k},\mathbf{Q})$ and the parity of $\mathcal{C}(\mathbf{k},\mathbf{Q},t)$ it follows that the sum in $\mathbf{k}$ in Eq. (A2) takes the form,

$$\sum_{\mathbf{k}}\mathcal{C}(\mathbf{k},\mathbf{Q},t)(\mathbf{Q}\cdot\mathbf{u}(\mathbf{k},\mathbf{Q}))^2u^{[2]}(\mathbf{k},\mathbf{Q})$$
$$=\gamma_2(\mathbf{Q})|\mathbf{Q}|^2\mathbb{1}+\gamma_3(\mathbf{Q})[\mathbf{QQ}], \tag{A3}$$

where γ_2 and γ_3 are coefficients resulting from the summation, whose details we omit. Using Eq. (A4) in Eq. (A2) and the fact that $\mathbf{I}_n(\mathbf{Q},t)$ is parallel to the vector wave number $\mathbf{Q}$ we obtain that

$$i\mathbf{Q}\otimes i\mathbf{Q}\otimes I_n^{[3]}(\mathbf{Q},t)=\mathcal{K}(\mathbf{Q})(i\mathbf{Q}\cdot\mathbf{I}_n(\mathbf{Q},t))i\mathbf{Q}, \tag{A4}$$

where

$$\mathcal{K}(\mathbf{Q})=\gamma_1(\mathbf{Q})(\gamma_2(\mathbf{Q})+\gamma_3(\mathbf{Q})). \tag{A5}$$

APPENDIX B: A SIMPLIFIED MODEL AND THE CLASSICAL FICK'S LAW

We here briefly describe the case of a fluid of fermions in interaction with a bath of bosons, specifically the model used in Ref. 29. In this simplified description, i.e., not involving the HOH extension of the main text, the equations of evolution take the form,

$$\frac{\partial}{\partial t}n(\mathbf{Q},t)=i\mathbf{Q}\cdot\mathbf{I}_n(\mathbf{Q},t), \tag{B1}$$

$$\frac{\partial}{\partial t}\mathbf{I}_n(\mathbf{Q},t)=i\mathbf{Q}I_{n2}(\mathbf{Q},t)-\Theta_1^{-1}\mathbf{I}_n(\mathbf{Q},t), \tag{B2}$$

$$\frac{\partial}{\partial t}\mathbf{I}_{n2}(\mathbf{Q},t)=i\mathbf{Q}\cdot\mathbf{I}_{n3}(\mathbf{Q},t)+\tfrac{1}{3}v_{\text{th}}^2\Theta_2^{-1}n(\mathbf{Q},t)$$
$$-\Theta_2^{-1}I_{n2}(\mathbf{Q},t), \tag{B3}$$

where it has been kept only the diagonal and isotropic part of the second and third order fluxes, β is the reciprocal of the quasitemperature, Θ_1 and Θ_2 are minus the characteristic Maxwell's times, and v_{th} is the thermal velocity, given by $mv_{\text{th}}^2/2=(3/2)\beta^{-1}$; and we recall that we have introduced $\tau_i=-\Theta_i$.

We can see in the right-hand side of these equations a first contribution associated with the conserved part (divergence of the fluxes in the direct space), followed in Eqs. (B2) and (B3) by the contributions associated to the collisional events governed by the fermion–boson interaction in the model. Transforming Fourier in time we obtain

$$i\omega n(\mathbf{Q},\omega)=i\mathbf{Q}\cdot\mathbf{I}_n(\mathbf{Q},\omega), \tag{B4}$$

$$(1+i\omega\theta_1)\mathbf{I}_n(\mathbf{Q},\omega)=\Theta_1 i\mathbf{Q}I_{n2}(\mathbf{Q},\omega), \tag{B5}$$

$$(1+i\omega\theta_2)I_{n2}(\mathbf{Q},\omega)=\Theta_2 i\mathbf{Q}\cdot\mathbf{I}_{n3}(\mathbf{Q},\omega)+\tfrac{1}{3}v_{\text{th}}^2 n(\mathbf{Q},\omega). \tag{B6}$$

The third-order flux is not a basic variable and needs be expressed in terms of them; using the linearized form of Heims–Jaynes expansion for averages, it follows that $i\mathbf{Q}\cdot\mathbf{I}_{n3}(\mathbf{Q},\omega)=(10/3\beta)i\mathbf{Q}\cdot\mathbf{I}_n(\mathbf{Q},\omega)$. Finally, rearranging these equations we arrive to the relation

$$\mathbf{I}_n(\mathbf{Q},\omega)=D(\mathbf{Q},\omega)i\mathbf{Q}I_{n2}(\mathbf{Q},\omega), \tag{B7}$$

where

$$D(\mathbf{Q},\omega)=(1+i\omega\theta_1)^{-1}(1+i\omega\theta_2)^{-1}\tfrac{1}{3}v_{\text{th}}^2\Theta_1$$
$$+(10/3\beta)\Theta_1\Theta_2(1+i\omega\theta_1)^{-1}$$
$$\times(1+i\omega\theta_2)^{-1}i\omega. \tag{B8}$$

In the limit of classical hydrodynamics ($\mathbf{Q}\to\mathbf{0},\omega\to0$) and in direct space, one recovers the usual Fick's Law,

$$\mathbf{I}_n(\mathbf{r},t)=-\tfrac{1}{3}v_{\text{th}}^2\tau_1\nabla n(\mathbf{r},t). \tag{B9}$$

APPENDIX C: THE CONTINUOUS FRACTION EXPANSION

Let us recall that from equations of evolution, it follows that

$$\mathbf{I}^{[l]}(\mathbf{Q},\omega)=-\frac{\Theta_l(\mathbf{Q})}{\Omega_l(\mathbf{Q},\omega)}\{(1+\mathcal{D}_{l+1}(\mathbf{Q}))i\mathbf{Q}$$
$$\otimes I^{[l+1]}(\mathbf{Q},\omega)+\mathcal{G}_{l-1}(\mathbf{Q})[i\mathbf{Q}I^{[l-1]}(\mathbf{Q},\omega)]$$
$$+\mathcal{B}^{[l]}(\mathbf{Q},\omega)\}, \tag{C1}$$

where

$$\Omega_l(\mathbf{Q},\omega)=1+i\omega\Theta_l(\mathbf{Q}). \tag{C2}$$

Introducing a truncation retaining the higher-order fluxes up to an order, say R, after some calculation it follows the general expression for the current ($l=1$) or generalized Fick's Law, in the form,

$$\mathbf{I}(\mathbf{Q},\omega|R) = -\frac{\mathcal{N}(\mathbf{Q},\omega|R)}{\Delta(\mathbf{Q},\omega|R)}, \tag{C3}$$

with R indicating the order of the truncation introduced, and where the denominator is the continuous fraction,

$$\Delta(\mathbf{Q},\omega|R) = 1 + \cfrac{a_1(\mathbf{Q},\omega)Q^2}{1 + \cfrac{a_2(\mathbf{Q},\omega)Q^2}{\ddots \cfrac{}{1 + \cfrac{a_{R-2}(\mathbf{Q},\omega)Q^2}{1 + a_{R-1}(\mathbf{Q},\omega)Q^2}}}}, \tag{C4}$$

where [cf. Eqs. (56) and (44)]

$$a_r(\mathbf{Q},\omega) = \frac{\xi_r(\mathbf{Q})}{\Omega_r(\mathbf{Q},\omega)\Omega_{r+1}(\mathbf{Q},\omega)}, \tag{C5}$$

$$\xi_r(\mathbf{Q}) = \Theta_r(\mathbf{Q})(1 + \mathcal{D}_{r+1}(\mathbf{Q}))\Theta_{r+1}(\mathbf{Q})\mathcal{G}_r(\mathbf{Q}). \tag{C6}$$

The numerator $\mathcal{N}(\mathbf{Q},\omega)$ stands for a generalized thermodynamic force taking the cumbersome expression,

$$\mathcal{N}(\mathbf{Q},\omega|R) = -\left(\prod_{j=0}^{R-3} C_j^{R-1}\right) D_R (i\mathbf{Q})^{R\otimes} I^{[R+1]}(\mathbf{Q},\omega) + \left(\prod_{j=0}^{R-3} C_j^{R-1}\right)\sum_{j=0}^{1} D_{R-j-1}\frac{\Theta_{R-j}}{\Omega_{R-j}}(i\mathbf{Q})^{(R-j-1)\otimes}\mathcal{B}^{[R-j]}(\mathbf{Q})$$

$$+ \sum_{k=1}^{R-3}\left(\prod_{j=k}^{R-3} C_j^{R-1}\right) D_{R-k-2}\frac{\Theta_{R-k-1}}{\Omega_{R-k-1}}(i\mathbf{Q})^{(R-k-2)\otimes}\mathcal{B}^{[R-k-1]}(\mathbf{Q}) + \frac{\Theta_1}{\Omega_1}\mathcal{B}^{[1]}(\mathbf{Q}) + \frac{\Theta_1\mathcal{G}_0}{\Omega_1}[i\mathbf{Q}n(\mathbf{Q},\omega)], \tag{C7}$$

where

$$D_r = (-1)^r \prod_{j=1}^{r} \frac{\Theta_j(1+\mathcal{D}_{j+1})}{\Omega_j}, \tag{C8}$$

$$C_0^k = \frac{1}{1+a_k Q^2}; \quad C_j^k = \frac{1}{1+C_{j-1}^k a_{k-j}Q^2}. \tag{C9}$$

We should notice that $\mathcal{D}_{R+1}(\mathbf{Q})=0$ and $D_0(\mathbf{Q})=0$, and that for $R=2$ the products should be taken equal to one and the third term on the right-hand side of Eq. (C7) is equal to zero.

According to the results above, we can write the generalized Fick's Law in the compact form,

$$\mathbf{I}(\mathbf{Q},\omega|R) = -C_{R-2}^{R-1}(\mathbf{Q},\omega)\mathcal{N}(\mathbf{Q},\omega|R). \tag{C10}$$

Equation (C7) can be written in an alternative way, which, for better visualization we write down for the case $R=3$, namely,

$$\mathcal{N}(\mathbf{Q},\omega|R) = -\frac{1}{\mathcal{Z}_1}\frac{\Theta_1[1+\mathcal{D}_2]}{\Omega_1}\frac{\Theta_2[1+\mathcal{D}_3]}{\Omega_2}\frac{\Theta_3}{\Omega_3}i\mathbf{Q}[Q^2 I_4(\mathbf{Q},\omega)] - \frac{1}{\mathcal{Z}_1}\frac{\Theta_1[1+\mathcal{D}_2]}{\Omega_1}\frac{\Theta_2[1+\mathcal{D}_3]}{\Omega_2}\frac{\Theta_3}{\Omega_3}\mathcal{G}_2 i\mathbf{Q}[Q^2 I_2(\mathbf{Q},\omega)]$$

$$+ \frac{1}{\mathcal{Z}_1}\frac{\Theta_1[1+\mathcal{D}_2]}{\Omega_1}\frac{\Theta_2[1+\mathcal{D}_3]}{\Omega_2}\frac{\Theta_3}{\Omega_3}i\mathbf{Q}[i\mathbf{Q}\cdot\mathcal{B}_3^{[1]}(\mathbf{Q},\omega)] - \frac{1}{\mathcal{Z}_1}\frac{\Theta_1[1+\mathcal{D}_2]}{\Omega_1}\frac{\Theta_2}{\Omega_2}[i\mathbf{Q}\mathcal{B}_2(\mathbf{Q},\omega)] + \frac{\Theta_1}{\Omega_1}\mathcal{B}_1^{[1]}(\mathbf{Q},\omega)$$

$$+ \frac{\Theta_1}{\Omega_1}\mathcal{G}_0 i\mathbf{Q}n(\mathbf{Q},\omega), \tag{C11}$$

where $\mathcal{Z}_1(\mathbf{Q},\omega) = 1 + a_1(\mathbf{Q},\omega)Q^2$ and, for simplicity, we have taken the tensors as diagonal and isotropic (i.e., we neglect shear contributions and anisotropy).

We can then see the particular structure for the generalized thermodynamic force $\mathbf{N}$. Besides the main contribution—the one that recovers Fick's Law in the limit of classical hydrodynamics—namely, proportional to the gradient of the concentration [$i\mathbf{Q}n(\mathbf{Q},\omega)$ in the reciprocal

space] being contained in the term with $\mathcal{B}_2$ as shown in Appendix B, there appear to be a number of additional contributions which also add nonlocal contributions to the final expression, besides those arising out of the continuous fraction expansion in the denominator. In the shown case of $R=3$ we can see the presence of a triple divergence of the flux of fourth order (which is not a basic variable and then needs be expressed in terms of the latter to close the expression),

which here reduces to the form of the first contribution on the right-hand side of this Eq. (C11). Besides, we can see the gradient of the double divergence of the second-order flux (i.e., contributions of the fluxes of even order), which appears in the second contribution. Moreover, it can be noticed the presence of the terms of type $\mathcal{B}(\mathbf{Q},\omega)$—of a very complicated structure—in this case all the possible three of them, involved with the gradient of the divergence of the vector $\mathcal{B}_3^{[1]}(\mathbf{Q},\omega)$, and the vector $\mathcal{B}^{[1]}(\mathbf{Q},\omega)$. We shall return to this question of generalized thermodynamic forces in a future paper.

APPENDIX D: ON THE ELEMENTS OF THE CONTINUOUS-FRACTION EXPANSION

We recall that the convergence of the continuous-fraction expansion is ensured by the condition,

$$|a_r(\omega)|Q^2 = |\xi_r|Q^2/|\Omega_r(\omega)\Omega_{r+1}(\omega)|$$

$$= |\Theta_r\Theta_{r+1}(1+\mathcal{D}_{r+1})$$

$$\times\mathcal{G}_r|Q^2/|\Omega_r(\omega)\Omega_{r+1}(\omega)| < 1, \qquad (D1)$$

where

$$|\Omega_r(\omega)\Omega_{r+1}(\omega)| = |(1+\omega^2\Theta_r^2)(1+\omega^2\Theta_{r+1}^2)|^{1/2}, \quad (D2)$$

and $a_r(\omega)$ is given in Eq. (C5). The limit of convergence is the case of all $|a_r|Q^2=1$, when the continuous-fraction is equal to the "golden number" $\phi=1.618...$. Let us analyze the relation in modulus of two successive $a_r(\omega)$, i.e.,

$$\left|\frac{a_{r+1}(\omega)}{a_r(\omega)}\right| = \left|\frac{\xi_{r+1}}{\xi_r}\right|\left|\frac{1+i\omega\theta_r}{1+i\omega\theta_{r+2}}\right|, \qquad (D3)$$

and after using Eq. (C6), where we neglect the contribution $\mathcal{D}$ in comparison with 1, we have that

$$\left|\frac{a_{r+1}(\omega)}{a_r(\omega)}\right| = \left|\frac{\Theta_{r+2}}{\Theta_r}\frac{\mathcal{G}_{r+1}}{\mathcal{G}_r}\right|\left[\frac{1+\omega^2\theta_r^2}{1+\omega^2\theta_{r+2}^2}\right]^{1/2}. \quad (D4)$$

In the limit of low frequencies ($\omega\Theta\ll1$) Eq. (D4) becomes

$$|a_{r+1}(\omega)/a_r(\omega)|\simeq|\Theta_{r+2}\mathcal{G}_{r+1}/\Theta_r\mathcal{G}_r|, \qquad (D5)$$

and at high frequencies we do have that

$$|a_{r+1}(\omega)/a_r(\omega)|\simeq|\mathcal{G}_{r+1}/\mathcal{G}_r|; \qquad (D6)$$

hence the relation of the coefficients along the continuous-fraction is determined by the quantities Θ and $\mathcal{G}$, which depend on each particular system under consideration.

For illustration we take the case of an ideal gas of particles of mass m, which is in interaction, via a central force potential, with an ideal gas of particles of mass M, acting as a reservoir with temperature T, and a classical approach is used. We take $m\gg M$, and then we are in the presence of a typical Brownian motion problem. It is considered a truncation with $R=2$, i.e., including the density and its first and second fluxes. It is obtained for Maxwell's characteristic times associated to the first and second fluxes the expressions,[30]

$$\Theta_1^{-1} = \sqrt{\frac{\pi}{2M}\frac{n_R}{3}}\mathcal{F}\beta^{3/2}\frac{x^{3/2}}{(1+x)^{1/2}}, \qquad (D7)$$
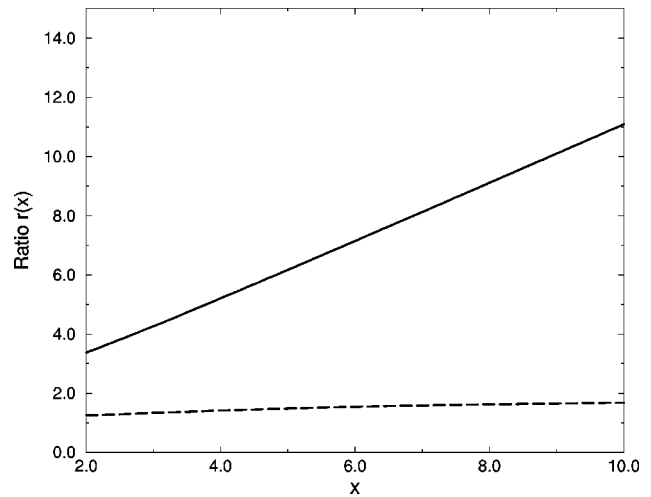

FIG. 1. The ratios of Eqs. (D16) and (D17) as a function of $x=m/M$: full line for r_{lf} and dashed line for r_{hf}.

$$\Theta_2^{-1} = \sqrt{\frac{\pi}{2M}\frac{n_R}{3}}\mathcal{F}\beta^{3/2}\frac{x^{3/2}}{(1+x)^{3/2}}, \qquad (D8)$$

where n_R is the density of the particles with mass M, $\beta^{-1}=k_BT$, with T being the common temperature of the system and reservoir, $x=m/M$ is the relation of the masses of the particles in system and reservoir, and

$$\mathcal{F}=\frac{1}{V}\sum_{\mathbf{q}}|\mathbf{q}||\mathcal{V}(\mathbf{q})|^2, \qquad (D9)$$

with $\mathcal{V}(\mathbf{q})$ being the Fourier transform of the interaction potential. Hence, using Eqs. (D3) and (D4) we obtain the relation,

$$\frac{\Theta_2}{\Theta_1}=\frac{1}{2}(1+x), \qquad (D10)$$

and we recall that $x>1$ (Brownian system) and then $\Theta_2>\Theta_1$, an inequality that also stands for the case of Maxwells' characteristic times for the quantum description of carriers and polar phonons in the direct-gap polar semiconductor GaAs.[31]

On the other hand, coefficients $\mathcal{G}$ are given by[30]

$$|\mathcal{G}_0|=\frac{n_R}{3}G\frac{\beta}{m}\frac{2+x}{(1+x)^2}, \qquad (D11)$$

$$|\mathcal{G}_1|=\frac{n_R}{3}G\frac{\beta}{m}\frac{2}{x}, \qquad (D12)$$

where

$$G=\frac{1}{V}\sum_{\mathbf{q}}\frac{1}{|\mathbf{q}|}|\mathcal{V}(\mathbf{q})|^2, \qquad (D13)$$

and then

$$\left|\frac{\mathcal{G}_1}{\mathcal{G}_0}\right|=\frac{2(1+x)^2}{x(2+x)}. \qquad (D14)$$

According to Eq. (D4), we have now that

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$$\left|\frac{a_2(\omega)}{a_1(\omega)}\right| = \frac{1}{2}(1+x)\frac{2(1+x)^2}{x(2+x)}\left[\frac{1+\omega^2\Theta_2^2}{1+\omega^2\Theta_1^2}\right]^{1/2}, \quad (D15)$$

where we can notice, on account of Eq. (D10) that the square root is a number smaller than 1. Let us analyze two limiting cases, first, the one of low frequencies ($\omega\Theta \ll 1$) when

$$r_{\mathrm{lf}}(x) \equiv \left|\frac{a_2(\omega)}{a_1(\omega)}\right|_{\mathrm{lf}} \simeq \frac{(1+x)^3}{x(2+x)}, \quad (D16)$$

and the one at high frequencies ($\omega\Theta \gg 1$) when

$$r_{\mathrm{hf}}(x) \equiv \left|\frac{a_2(\omega)}{a_1(\omega)}\right|_{\mathrm{hf}} \simeq \frac{2(1+x^2)}{x(2+x)}. \quad (D17)$$

These two ratios, depending on x, are shown in Fig. 1 for a certain range of values of x. We can notice that for this particular Brownian-type system it follows that $|a_2(\omega)| > |a_1(\omega)|$ for any value of x.

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