Statistical Mesoscopic Hydro-Thermodynamics: The Description of Kinetics and Hydrodynamics of Nonequilibrium Processes in Single Liquids

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Hydrodynamics, a term apparently introduced by Daniel Bernoulli (1700-1783) to comprise hydrostatic and hydraulics, has a long history with several theoretical approaches. Here, after a descriptive introduction, we present so-called mesoscopic hydro-thermodynamics, which is also referred to as higher-order generalized hydrodynamics, built within the framework of a mechanical-statistical formalism. It consists of a description of the material and heat motion of fluids in terms of the corresponding densities and their associated fluxes of all orders. In this way, movements are characterized in terms of intermediate to short wavelengths and intermediate to high frequencies. The fluxes have associated Maxwell-like times, which play an important role in determining the appropriate contraction of the description (of the enormous set of fluxes of all orders) necessary to address the characterization of the motion in each experimental setup. This study is an extension of a preliminary article: Physical Review E 91, 063011 (2015).

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I. INTRODUCTION

The name hydrodynamics was apparently first introduced by Daniel Bernoulli (1700-1783) to comprise the disciplines of hydrostatics and hydraulics. Leonard Euler (1707-1783) developed the equations of motion of a perfect fluid and the accompanying mathematical theory. Stokes (1819-1903) derived the equations of motion of a viscous fluid and can be considered to be a founder of the modern theory of hydrodynamics [1].

Microscopic descriptions of hydrodynamics, that is, the derivation of kinetic equations from classical or quantum mechanics with the kinetic (or transport) coefficients written in terms of correlation functions, is a traditional long-standing problem. An important aspect is the derivation of constitutive laws that express thermodynamic fluxes (or currents in the case of matter and energy) in terms of appropriate thermodynamic forces (typically gradients of densities in the case of matter and energy). In their most general form, these laws are nonlocal in space and noninstantaneous in time. The nonlocality is usually addressed by spatial Fourier transforms, and then the laws – expressed in reciprocal space - become dependent on the wavevector \( \mathbf{Q} \). The well-known expressions of classical (or Onsagerian) hydrothermodynamics are obtained by performing the limit of \( \mathbf{Q} \) going to zero (long wavelengths). The expressions are then valid in this limit and to go beyond, it is necessary to introduce a proper dependence on \( \mathbf{Q} \) that is valid, in principle, for intermediate and short wavelengths (intermediate to large wavenumbers). In phenomenological theories, this corresponds to going from classical (Onsagerian) irreversible thermodynamics to extended irreversible thermodynamics [2,3]. This is what has been called generalized hydrodynamics, that is, to go beyond traditional hydrodynamics, the latter is restricted to fluctuations occurring at long wavelengths and low frequencies. This idea has been extensively debated for decades by the statistical mechanics community. Several approaches have been used, and a description can be consulted in chapter 6 of the classic book on the subject by Boon and Yip [4].

Here, we present a statistical approach based on a grand canonical ensemble generalized to cover the case of fluids arbitrarily away from equilibrium and at intermediate to short wavelengths.

Nonlocal effects for describing motion with the influence of decreasing wavelengths (going towards the very short limit) have been introduced in terms of expansions in increasing powers of the wavenumber, which currently consists of what is sometimes referred to as higher-order hydrodynamics (HOH) or mesoscopic hydro-thermodynamics (MHT) [5].

Earlier attempts to perform such expansions are the so-called Burnett and super-Burnett approaches, for the case of mass motion, and the Guyer-Kruhsmans approach [6] in the case of propagation of energy in semiconductors. The usual approach is based on the moment solution procedure of the Boltzmann equation, as in the work of Hess [7], using a higher-order Chapman-Enskog solution method. The Chapman-Enskog method provides a solution to the Boltzmann equation consisting of a series in powers of the Knudsen number, \( K_n \), given by the ratio between the mean free path of the particles and the scale of the variation (relevant wavelengths in the motion) of the hydrodynamic fields. Retaining the linear term in \( K_n \) gives the Navier-Stokes equation, the term

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containing $K^2$ introduces the so-called Burnett correction, and the higher order terms ($K^3$ and up) give the super-Burnett corrections [8].

The satisfactory development of MHT is highly desirable for covering a large class of hydrodynamic situations and, in the last instance, for obtaining insights into technological and industrial processes with an associated economic interest: for example, oil recovery, pollution decontamination or CO$_2$ sequestration in soils that are important environmental processes. Indeed, the nonlocal terms become particularly important in miniaturized devices with constrained geometries, for example, the nanometer scales in electronic and optoelectronic devices.

We have considered the question of going beyond earlier approaches by means of a nonequilibrium statistical-mechanical formalism [9-22], which thus contains the quantum – or classical - microscopic dynamics and the macroscopic nonequilibrium thermodynamics [23-26], with the equations of hydrodynamics following from the mechanical equations of motion averaged over a nonequilibrium statistical ensemble, which is provided by the associated kinetic theory of the formalism [12-15,27-34]. MHT was derived based on the method of moments for the solution of the single-particle (or quasi-particle) kinetic equation [35,36] (generically referred to as the Boltzmann equation), initiated by Grad [37]. The case of a dilute solution of many Brownian particles was reported in Ref. [38]. MHT of quasi-particle phonons was presented under different conditions in Refs. [39-42], and the case with complex structure fluids (with “hidden constraints”) was presented in Ref. [43].

In this paper, we present an extensive description of the MHT of a classical fluid of molecules with internal interactions. Generalizations of the so-called Maxwell time arise in the treatment, the issue of the contraction of the description (of general MHT involving many fluxes of all orders) is discussed, and the theory is illustrated with an MHT description of order 2.

II. THEORETICAL BACKGROUND

Let us consider a fluid of $N$ molecules (with density $n$) of mass $m$, with $\mathbf{r}_j$ and $\mathbf{p}_j$ being the coordinate and linear momentum of the $j$-th molecule in contact with an external reservoir at temperature $T_0$. We write the Hamiltonian as

$$\hat{H} = \hat{H}_0 + \hat{H}',$$

where

$$\hat{H}_0 = \sum_{j=1}^{N} \frac{\mathbf{p}_j^2}{2m},$$

is the kinetic energy, and

$$\hat{H}' = \frac{1}{2} \sum_{j \neq k} V(\mathbf{r}_j - \mathbf{r}_k),$$

represents the pair interaction of the molecules.

A macroscopic (thermodynamic) description is made in terms of the non-equilibrium statistical ensemble formalism (NESEF) described in Refs. [9-22], the associated NESEF kinetic theory in Refs. [27-29], and the NESEF-nonequilibrium thermodynamic theory in Refs. [23-26]. A very briefly summary is given in Appendix A.

The nonequilibrium statistical operator, $g_{\varepsilon}(t)$ in Appendix A, is dependent on a set of microdynamical variables, which for the present problem are the hydrodynamical variables consisting of the density of particles

$$\hat{n} = \int d^3\mathbf{p} \hat{n}_1(\mathbf{r},\mathbf{p}),$$

and the density of energy

$$\hat{e} = \int d^3\mathbf{p} \hat{e}_1(\mathbf{r},\mathbf{p}),$$

where

$$\hat{n}_1(\mathbf{r},\mathbf{p}) = \frac{1}{N} \sum_{j=1}^{N} \delta(\mathbf{r} - \mathbf{r}_j) \delta(\mathbf{p} - \mathbf{p}_j)$$

is the single-particle dynamical operator, and the fluxes of all orders of these densities are,

$$\hat{J} = \int d^3\mathbf{p} \frac{\mathbf{p}}{m} \hat{n}_1(\mathbf{r},\mathbf{p}),$$

$$\hat{J}_t^{(0)} = \int d^3\mathbf{p} \mathbf{u}(\mathbf{p}) \hat{n}_1(\mathbf{r},\mathbf{p}),$$

$$\hat{J}_t^{(1)} = \int d^3\mathbf{p} \frac{\mathbf{p}^2}{2m} \hat{n}_1(\mathbf{r},\mathbf{p}),$$

$$\hat{J}_t^{(2)} = \int d^3\mathbf{p} \frac{\mathbf{p}^2}{2m} \mathbf{u}(\mathbf{p}) \hat{n}_1(\mathbf{r},\mathbf{p}),$$

where

$$\mathbf{u}(\mathbf{p}) = \left[ \frac{\mathbf{p}}{m} : \ldots (\ell - \text{times}) : \frac{\mathbf{p}}{m} \right]$$

is a $\ell$-order tensor (the inner product of $\ell$-times the velocity $\mathbf{p}/m$) with $\ell = 2, 3, \ldots$.

Hence, according to the formalism

$$g_{\varepsilon}(t) = \exp \left\{ -\int_0^t dt' \varepsilon(t' - t) \frac{d}{dt'} \ln \hat{g}(t', t' - t) \right\},$$

where $\hat{g}$ is an auxiliary (also dubbed instantaneous quasi-equilibrium) probability distribution

$$\hat{g}(t_1, t_2) = \exp \left\{ -\phi(t_1) - \int d^3\mathbf{r} [F_n(\mathbf{r}, t) \hat{n}(\mathbf{r}) + F_h(\mathbf{r}, t) \hat{h}(\mathbf{r})] + \mathbf{F}_n(\mathbf{r}, t) \cdot \hat{I}_n(\mathbf{r}) + \mathbf{F}_h(\mathbf{r}, t) \cdot \hat{I}_h(\mathbf{r}) + \sum_{\ell \geq 2} F_n^{(\ell)}(\mathbf{r}, t) \otimes \hat{I}_n^{(\ell)}(\mathbf{r}) + \sum_{\ell \geq 2} F_h^{(\ell)}(\mathbf{r}, t) \otimes \hat{I}_h^{(\ell)}(\mathbf{r}) \right\}.$$
where are associated (or conjugated) with the basic hydrodynamic variables, the nonequilibrium thermodynamic variables

\[
\{ F_n(r, t), F_h(r, t), F_n(r, t), F_h(r, t), \{ F_n^{[\ell]}(r, t) \}, \{ F_h^{[\ell]}(r, t) \}\},
\]

(14)

and \( \odot \) stands for a fully contracted product of tensors. We recall that \( \varepsilon \) is a real infinitesimal number that goes to +0 after the calculation of average values have been performed. The contribution involved accounts for historicity and irreversibility in the process (see Appendix A). An initial time in the remote past \( (t_0 \to -\infty) \) has been used, implying an adiabatic switch-on of relaxation processes.

We can now formulate the basic equations of NESEF-based MHT, that is, the equations of evolution for the average values of the basic microvariables of Eqs. (4) to (10); we denote the basic macrovariables as

\[
\{ \eta(r, t), h(r, t), \eta(r, t), \eta(r, t), \{ \eta_n^{[\ell]}(r, t) \}, \{ \eta_h^{[\ell]}(r, t) \}\},
\]

(15)

\[ \ell = 2, 3, \ldots \], meaning that

\[
\eta(r, t) = \text{Tr}\{\eta_1(r, p)\} \varrho(t),
\]

(16)

\[
h(r, t) = \text{Tr}\{\eta_1(r, p)\} \varrho(t),
\]

(17)

and so on for all the others. In this classical mechanics approach, \( \text{Tr} \) stands for integration in phase space.

Hence, we have that

\[
\frac{\partial}{\partial t} I_n^{[\ell]}(r, t) = \int d^3 p w_n^{[\ell]}(p) \frac{\partial}{\partial t} f_1(r, p; t),
\]

(18)

which we call the hydrodynamic family \( n \), and

\[
\frac{\partial}{\partial t} I_h^{[\ell]}(r, t) = \int d^3 p \frac{p^2}{2m} w_h^{[\ell]}(p) \frac{\partial}{\partial t} f_1(r, p; t),
\]

(19)

which we call the hydrodynamic family \( h \).

Here, \( \ell = 0 \) for the densities, \( \ell = 1 \) for the first fluxes, \( \ell = 2, 3, \ldots \) for the higher-order fluxes, and \( f_1(r, p; t) \) is the single-particle distribution function

\[
f_1(r, p; t) = \text{Tr}\{\eta_1(r, p)\} \varrho(t),
\]

(20)

which can be generically called the Boltzmann distribution function.

The distribution \( f_1(r, p; t) \) satisfies the single-particle kinetic equation [33] (also called the generalized Boltzmann equation) and therefore the set of Eqs. (18), that is, the equations of MHT are the multiple moment equations in the solution of the generalized Boltzmann equation (initiated with the 14-moment approach by Grad [37]). (We noticed that in theoretical statistics, the method of moments for the solution of evolution equations of probability distributions seems to have been originally proposed by the renowned Russian mathematician Tchebycheff in the 19th century). We call attention to the fact that there is a type of redundancy in the pair of the basic equations, in the sense that

\[
I_h^{[\ell]}(r, t) = \frac{1}{2} m \text{Tr}_{12} I_n^{[\ell+2]}(r, t),
\]

(21)

where \( \text{Tr}_{12} \) stands for the contraction of the first two indexes.

Inspection of Eqs. (18) and (19) clearly tells us that the evolution equations for the hydrodynamic variables are exclusively dependent on the evolution equation for the single-particle distribution function \( f_1(r, p; t) \), which is the mechanical equation of motion for the single-particle dynamical operator \( \eta_1 \) averaged over the nonequilibrium ensemble, namely,

\[
\frac{\partial}{\partial t} f_1(r, p; t) = \frac{\partial}{\partial t} \text{Tr}\{\{\eta_1(r, p), \hat{H}\} \varrho(t)\} = \text{Tr}\{[\hat{L} \eta_1(r, p)] \varrho(t)\},
\]

(22)

where \( \hat{L} \) is the Liouville operator of the system. However, solving these equations is a quite difficult task of almost unmanageable proportions. Hence, this situation calls for the derivation of an appropriate kinetic theory of practical use. This can be done in a way that was initiated by several authors [9,12,13,27,28] and systematized and extended by Lauck et al. [29]. In summary, Eq. (22) acquires the form of a generalization of Mori’s equation of motion with a highly nonlinear character, namely,

\[
\frac{\partial}{\partial t} f_1(r, p; t) = J_1^{(0)}(r, p; t) + J_1^{(1)}(r, p; t) + \sum_{n \geq 2} \Omega_1^{(n)}(r, p; t),
\]

(23)

where on the right-hand side, the first contribution \( J_1^{(0)} \) is in Mori’s nomenclature a precession term, the second \( J_1^{(1)} \) is a term involving the action of the external and internal forces, and the last contribution is a general collision integral consisting of a series in increasing powers \( (n \geq 2) \) of the interaction strengths. These partial collision integrals are fully described in Refs. [12,13,29], and here we simply note that they involve pair collisions \( (n = 2) \), triple collisions \( (n = 3) \), and so on, with each one including memory and vertex renormalization effects.

The first two terms on the right-hand side of Eq. (23) are given by

\[
J_1^{(0)}(r, p; t) = \text{Tr}\{\{\eta_1(r, p), \hat{H}_0\} \varrho(t, 0) \varrho_R\},
\]

(24)

\[
J_1^{(1)}(r, p; t) = \text{Tr}\{\{\eta_1(r, p), \hat{H}'\} \varrho(t, 0) \varrho_R\},
\]

(25)

where \{\ldots, \ldots\} stands for Poisson brackets, and \( \varrho_R \) is the statistical distribution of the external reservoir at temperature \( T_0 \). In the lowest-order approximation, which is generally valid, keeping the contribution to second order
in the interaction strengths, (“binary collisions” without memory) \[12,13,29,30,33\], the last contribution in Eq. (23) reduces to:

\[
J^{(2)}_1(\mathbf{r}, \mathbf{p}; t) = \int_{-\infty}^{t} dt' e^{\ell(t'-t)} \times \\
\text{Tr} \left\{ \{\hat{H}'(t'-t)_{0}, \{\hat{H}', \tilde{n}_1(\mathbf{r}, \mathbf{p})\} \times \right.
\]

\[
\tilde{\varrho}(t,0)\mathcal{Q}_R \} + \\
\int_{-\infty}^{t} dt' e^{\ell(t'-t)} \int d^3r' d^3p' \times \\
\text{Tr} \left\{ \{\hat{H}'(t'-t)_{0}, \tilde{n}_1(\mathbf{r}', \mathbf{p}')\} \tilde{\varrho}(t,0)\mathcal{Q}_R \} \times \right.
\]

\[
\frac{\delta J^{(1)}_1(\mathbf{r}, \mathbf{p}; t)}{\delta f_1(\mathbf{r}', \mathbf{p}'; t)}, \tag{26}
\]

where \(\tilde{\varrho}(t,0)\) is given in Eq. (13), and the nought subindex indicates time evolution in the interaction representation. The two contributions on the right-hand side of Eq. (26) consist of the so-called irreducible contribution and the vertex renormalization (effects of the forces present in \(J^{(1)}_1\) acting at the time of the “collision”) \[44\]; \(\delta\) stands for functional differentiation \[45\].

The NESEF single-particle kinetic equation was presented in Ref. \[34\], which included the interaction with a thermal bath (a multiple Brownian particle system) and interparticle interactions given in the so-called weak-coupling limit. We briefly describe the equation in Appendix B, including a complete treatment of the two-particle interaction, i.e., going beyond the weak coupling limit.

In the absence of a thermal bath, that is, considering the single fluid described at the beginning of this Section, we have that the kinetic equation for \(f_1(\mathbf{r}, \mathbf{p}; t)\) is given by

\[
\frac{\partial}{\partial t} f_1(\mathbf{r}, \mathbf{p}; t) = J^{(0)}_1(\mathbf{r}, \mathbf{p}; t) + J^{(1)}_1(\mathbf{r}, \mathbf{p}; t) + J^{(2)}_1(\mathbf{r}, \mathbf{p}; t), \tag{27}
\]

where the first terms on the right-hand side are

\[
J^{(0)}_1(\mathbf{r}, \mathbf{p}; t) = \text{Tr} \left\{ \{\tilde{n}_1(\mathbf{r}, \mathbf{p}), \hat{H}_0\} \tilde{\varrho}(t,0)\mathcal{Q}_R \} \right.
\]

\[
= -\frac{\mathbf{p}}{m} \cdot \nabla f_1(\mathbf{r}, \mathbf{p}; t), \tag{28}
\]

which is, as already noticed, the precession term in Moris’s terminology, and

\[
J^{(1)}_1(\mathbf{r}, \mathbf{p}; t) = \text{Tr} \left\{ \{\tilde{n}_1(\mathbf{r}, \mathbf{p}), \hat{H}'\} \tilde{\varrho}(t,0)\mathcal{Q}_R \} \right.
\]

\[
= [\nabla U_{ar}(\mathbf{r}; t) + \nabla U(\mathbf{r}; t)] \cdot \nabla \mathbf{p} f_1(\mathbf{r}, \mathbf{p}; t), \tag{29}
\]

is the contribution containing the action of the external and internal forces, where

\[
U(\mathbf{r}; t) = \int d^3r' d^3p' V(|\mathbf{r} - \mathbf{r}'|) f_1(\mathbf{r}', \mathbf{p}'; t) \tag{30}
\]

plays the role of a mean-field potential of the interaction between particles or a Vlasov-like potential, and

\[
J^{(2)}_1(\mathbf{r}, \mathbf{p}; t) = J^{(2)}_{11}(\mathbf{r}, \mathbf{p}; t) + J^{(2)}_{12}(\mathbf{r}, \mathbf{p}; t) + J^{(2)}_{13}(\mathbf{r}, \mathbf{p}; t), \tag{31}
\]

is the collision integral resulting from the interaction between particles, shown in Appendix B. The first contribution on the right-hand side is, we recall, the weak-coupling contribution.

Consequently, the hydrodynamic equations, Eq. (18) and (19), take the form

\[
\frac{\partial}{\partial t} I^{[\ell]}_h(\mathbf{r}; t) = \int d^3p u^{[\ell]}(\mathbf{p}) \left\{ J^{(0)}_1(\mathbf{r}, \mathbf{p}; t) + J^{(1)}_1(\mathbf{r}, \mathbf{p}; t) + \right.
\]

\[
+ J^{(2)}_1(\mathbf{r}, \mathbf{p}; t) \right\}, \tag{32}
\]

\[
\frac{\partial}{\partial t} I^{[\ell]}_h(\mathbf{r}; t) = \int d^3p \frac{\mathbf{p}^2}{2m} u^{[\ell]}(\mathbf{p}) \left\{ J^{(0)}_1(\mathbf{r}, \mathbf{p}; t) + \right.
\]

\[
+ J^{(1)}_1(\mathbf{r}, \mathbf{p}; t) + J^{(2)}_1(\mathbf{r}, \mathbf{p}; t) \right\}, \tag{33}
\]

recalling that \(\ell = 0\) stands for the densities, \(\ell = 1\) stands for the first fluxes, and \(\ell \geq 2\) stands for the higher-order fluxes.

In Section IV, we address in complete detail the case of contraction in an MHT description of order 2, considering the \(n\)-family.

### III. CONTRACTION OF DESCRIPTION

On the issue of the contraction of the description, we noticed that a truncation criterion can be derived, which rests on the characteristics of the hydrodynamic motion that develops under the given experimental procedure.

Since inclusion of higher- and higher-order fluxes implies describing motion with increasing Knudsen numbers for each hydrodynamic mode (that is governed by smaller and smaller wavelengths — larger and larger wavelengths — accompanied by higher and higher frequencies), in a qualitative manner, we can say, as a general “rule of thumb,” that the criterion indicates that an increasingly restricted contraction can be used when the prevalent wavelengths in the motion are larger. Therefore, in simpler words, when the motion becomes smoother in space and time, the dimension of the space of basic macrovariables used for the description of the nonequilibrium thermodynamic state of the system can be further reduced.

As shown elsewhere \[42\], a general contraction criterion can be conjectured, namely, a truncation of order \(r\) (keeping the densities and their fluxes to order \(r\)) can be introduced, once we show that in the spectrum of wavelengths, the motion predominates over the “frontier” motion, \(\lambda^2_{(r,r+1)} = \nu^2 \theta_r \theta_{r+1}\), where \(\nu\) is the order of the thermal velocity, and \(\theta_r\) and \(\theta_{r+1}\) are the corresponding Maxwell times associated with the \(r\)- and \(r+1\)-fluxes.
We recall that the Maxwell time was originally introduced by Maxwell in his fundamental article of 1867 [48] on the dynamical theory of gases in what was related to viscoelasticity. A family of Maxwell times is present in MHT associated with the dampening of particle and energy densities and their fluxes of all orders [38].

**IV. MESOSCOPIC HYDRO-THERMODYNAMICS OF ORDER 2**

We consider a contraction of the description of order 2, MHT[2], in the evolution of the family \( n \), that is, we introduce the set of hydrodynamic variables

\[
\left\{ n(r, t), I_n(r, t), I_n^2(r, t) \right\},
\]

a thirteen-moments approach (or Grad’s fourteen moments once the energy is incorporated in \( I^2 \), which is related to the pressure tensor; see Eq. (21)). We call attention to the fact that the approach here has a purely mechanical basis, in a laboratory reference frame, while the Grad’s approach is a hybrid approach including hydrodynamic variables in a barycentric frame of reference in the definitions.

Recalling that the second flux contains the density of energy, namely, (cf. Eq. (21))

\[
h(r, t) = m \text{Tr} \left\{ I_n^2(r, t) \right\},
\]

it is convenient to improve the physical discussions to redefine the set of basic hydrodynamic variables as

\[
\left\{ h(r, t), n(r, t), I_n(r, t), I_n^2(r, t) \right\},
\]

where we have introduced the traceless part of the second flux, that is, \( I_n^2(r, t) \).

The conjugated nonequilibrium thermodynamic variables, see Appendix A, are designed by

\[
\left\{ F_h(r, t), F_n(r, t), F_n^2(r, t), F_n^2(r, t) \right\},
\]

which are redefined as

\[
F_h(r, t) \equiv \beta(r, t) = \frac{1}{k_B T^*(r, t)}, \tag{38}
\]

\[
F_n(r, t) \equiv -\beta(r, t) \mu^*(r, t), \tag{39}
\]

\[
F_n^2(r, t) \equiv -\beta(r, t) \mathbf{v}(r, t), \tag{40}
\]

introducing a nonequilibrium temperature (usually called quasi-temperature), \( T^*(r, t) \), where \( \mathbf{v}(r, t) \) is the field of the barycentric velocity, and \( \mu^*(r, t) \) is a nonequilibrium chemical potential [49,50]. Moreover, we write

\[
n(r, t) = n_0 + \Delta n(r, t), \tag{41}
\]

\[
T^*(r, t) = T_0 + \Delta T^*(r, t), \tag{42}
\]

where \( n_0 \) and \( T_0 \) are values in equilibrium, and we admit that \( \Delta n(r, t) \ll n_0 \) and \( \Delta T^*(r, t) \ll T_0 \).

Using the Heims-Jaynes perturbation expansion for averages [51] to *first order* (see Appendix C), we obtain the nonequilibrium equations of state, which consist of a linear relation between the nonequilibrium thermodynamic variables in Eq. (35) and the basic variables in Eq. (34), given by

\[
h(r, t) = \frac{3}{2} n(r, t) T^*(r, t), \tag{43}
\]

\[
I_n(r, t) = n(r, t) \mathbf{v}(r, t) \simeq n_0 \mathbf{v}(r, t), \tag{44}
\]

\[
j_n^2(r, t) \simeq \frac{2 (k_B)^2}{m^2} n_0 F_n^2(r, t), \tag{45}
\]

We recall that the evolution equations are

\[
\frac{\partial}{\partial t} n(r, t) = J_n^{(0)}(r, t) + J_n^{(1)}(r, t) + J_n^{(2)}(r, t), \tag{46}
\]

\[
\frac{\partial}{\partial t} I_n(r, t) = J_n^{(0)}(r, t) + J_n^{(1)}(r, t) + J_n^{(2)}(r, t), \tag{47}
\]

\[
\frac{\partial}{\partial t} I_n^2(r, t) = J_n^{(0)}(r, t) + J_n^{(1)}(r, t) + J_n^{(2)}(r, t). \tag{48}
\]

Lengthy calculations lead to cumbersome expressions that we omit here. We introduce the following approximations:

1. In the expressions of the kinetic coefficients, we take \( n(r, t) \simeq n_0 \) and \( T^*(r, t) \simeq T_0 \) (cf. Eqs. (39) and (40)).

2. Spatial correlations are neglected; that is, a local approximation is introduced, and the set of coupled nonlinear integro-differential equations is greatly simplified.

Moreover, of the three contributions to the collision integral registering the two-particle interaction, only one contributes to the weak-coupling interactions; the others become null because of symmetry effects. We recall that weak coupling involves collisions with a low-angle transfer of momentum.

The evolution equation for the density is

\[
\frac{\partial}{\partial t} n(r, t) + \nabla \cdot I_n(r, t) = 0, \tag{49}
\]

which is the conservation equation for the number of molecules. For the first flux, we obtain that

\[
\frac{\partial}{\partial t} I_n(r, t) \simeq - \nabla \cdot I_n^2(r, t) - \frac{a_{10}}{m} \nabla V(r, t) + a_{12} \nabla \cdot I_n^2(r, t) + -\theta_1 I_n(r, t), \tag{50}
\]
where

\[ \mathbf{V}(r, t) = \int d^3r' V(r - r') n(r', t) \]  

is the mean field (Vlasov-Landau-like field) of the two-particle interactions,

\[ a_{10} = \frac{7}{5} n_0 \frac{A}{mk_BT_0}, \]  

\[ a_{12} = \frac{2}{5} n_0 \frac{A}{(kB_T0)^2}, \]  

\[ \mathcal{A} = \frac{1}{4\pi^2} \int dq d\theta |\Phi(q)|^2 q^3 \cos^2 \theta \sin \theta, \]  

where \( \Phi(q) \) is the Fourier transform of the two-particle potential, and \( \theta_1 \) is the Maxwell time associated with the first flux given by

\[ \theta_1^{-1} = \frac{A}{kB_T0} \sqrt{\frac{2\pi}{mk_BT_0}}. \]  

In Eq. (48), the first term on the right-hand side comes from \( J^{(0)}_f \), the second term \( J^{(1)}_f \) contains the force arising out of the mean field originating in the two-molecule interaction, and the other three terms have their origin in \( J^{(2)}_n \). The first two terms are contributions from a self-energy correction involving the gradient of the previous term, that is, \( \nabla n(r, t) \), and the divergence of the next term, that is, \( \nabla \cdot I^{(2)}_n(r, t) \) (quite analogously to those present in Ref. [5], here given in an expression at the microscopic-statistical level), and the last term can be referred to as a generalized Maxwellian relaxation that introduces a generalized Maxwell time. The origin of the Maxwell time goes back to a fundamental article by J. C. Maxwell in 1867 [48], interpreted as the time during which stresses in media are damped [51]; each flux in MHT has an associated Maxwellian time, and the relaxation times of the hydrodynamic motion are a combination of these multiple Maxwell times.

For the second-order flux, we recall that it contains the density of energy (cf. Eq. (35)), and we have that

\[ \frac{\partial}{\partial t} I^{(2)}_n(r, t) = -\nabla \cdot I^{(2)}_n(r, t) \]

\[-\frac{1}{m} \{ \{ I_n(r, t) : \nabla \nabla (r, t) \} + \text{transposed} \}
+ a_{20} n(r, t) 1[2] + a_{21} \nabla \cdot I_n(r, t) 1[2]
- \frac{8}{4} a_{12} \{ \nabla \cdot I_n(r, t) + \text{transposed} \}
- \theta_2^{-1} I^{(2)}_n(r, t), \]  

where

\[ a_{20} = \frac{kB_T0}{m} \theta_2^{-1}, \]  

\[ a_{21} = \frac{4}{5} \frac{B}{mk_BT_0}, \]  

and \( \theta_2 \) is Maxwell time associated with the second-order flux given by

\[ \theta_2^{-1} = \frac{3}{5} \theta_1^{-1}. \]  

Moreover, taking into account that (cf. Eq. (21)) the density of energy is

\[ h(r, t) = \frac{1}{2} m \text{Tr} \left\{ I^{(2)}_n(r, t) \right\}, \]  

\[ \text{I}_h(r, t) = \frac{1}{2} m \text{Tr}_{12} \left\{ I^{(3)}_n(r, t) \right\}, \]  

resorting to the use of Eqs. (46), (48), (55) and that

\[ \nabla \left\{ \nabla \cdot I^{(3)}_n(r, t) \right\} \]  

\[ \simeq \frac{k_BT0}{m} \left\{ \nabla^2 I_n(r, t) + 2 \nabla (\nabla \cdot I_n(r, t)) \right\} I^{(2)}_n(r, t), \]

after using the Heims-Jaynes formalism in the linear approximation, we obtain the evolution equation for the density energy given by

\[ \frac{\partial}{\partial t} h(r, t) + \nabla I_h(r, t) = -\nabla \mathbf{V}(r, t) \cdot \mathbf{I}_n(r, t) + \]

\[ + \frac{3}{2} \frac{k_BT0}{m} \theta_2^{-1} n(r, t) + \]

\[ + n_0 \frac{B}{kB_T0} \nabla \cdot \mathbf{I}_n(r, t) + \]

\[ - \theta_2^{-1} h(r, t), \]

and according to Eq. (59), the momentum relaxation time is smaller than the energy relaxation time.

However, after some mathematical handling of Eqs. (46), (48), (55) and using Eq. (61), we obtain for the density that

\[ \frac{\partial^3 n(r, t)}{\partial t^3} + \frac{1}{\theta_{ef}} \frac{\partial^2 n(r, t)}{\partial t^2} + \frac{1}{(\theta_{(2)})^2} \frac{\partial n(r, t)}{\partial t} + \]

\[ - \frac{C_{N,L}^2}{\theta_2} \nabla^2 n(r, t) = \]

\[ = 3n_0 \frac{B}{5mk_BT_0} \nabla^2 [\nabla \cdot I_n(r, t)] + \]

\[ - \left( 1 - \frac{2n_0 B}{5(k_BT_0)^2} \right) \frac{k_BT0}{m} \times \]

\[ \nabla \cdot [\nabla^2 I_n(r, t) + 2 \nabla (\nabla \cdot I_n(r, t))] \]

\[ - \frac{n_0 B}{5mk_BT_0} \nabla \cdot \left\{ \nabla \cdot [\nabla I_n(r, t) + (\nabla I_n(r, t)^{tr})] \right\}, \]  

(64)
where we discarded terms in $B^2$,

$$C_{G,NL}^2 = \frac{k_B T_0}{m} + \frac{n_0 B}{mk_B T_0} = C_{GL}^2 + \frac{n_0 B}{mk_B T_0}, \quad (65)$$

$$C_{GL}^2 = \frac{k_B T_0}{m} + \frac{n_0 B}{mk_B T_0} = C_{GL}^2 + \frac{n_0 B}{mk_B T_0}, \quad (66)$$

$$\frac{1}{\theta_{\varepsilon}} = \frac{1}{\theta_1} + \frac{1}{\theta_2} \quad \text{and} \quad \theta_{(2)} = \sqrt{\theta_1 \theta_2}, \quad (67)$$

This equation has a third-order differentiation in time: neglecting the third derivative leads to a generalized Maxwell-Cattaneo equation, and if the second derivative is neglected, we have a generalized Fick diffusion equation. This implies hydrodynamic motion that becomes smoother in time.

V. CONCLUDING REMARKS

After an introduction with a brief discussion of some aspects of hydrodynamics, we presented so-called mesoscopic hydro-thermodynamics, or higher-order generalized hydrodynamics, built in the framework of a mechanical-statistical formalism (the non-equilibrium statistical ensemble formalism).

MHT (HOGH) allows, in principle, to describe hydrodynamic motion without restrictions in the values of the wavelengths and frequencies involved. The theoretical description involves including the densities of particles and energy together with their fluxes of all orders, which are related by an enormous set of coupled nonlinear integro-differential equations describing the motion. The solution is not practically feasible, and one needs to introduce a contraction of the description, that is, one needs to retain a finite small number of fluxes, up to a certain order $n$, considered to be appropriate for describing the motion under consideration. The order of the contracted description is determined by a criterion (see Section III), which heavily depends on the values of the Maxwell-like times that are associated with the different fluxes. These times are clearly characterized in the equations of motion.

It may be noted that this is a completely analytical theory, and competing theories are computational modeling theories derived from the nonequilibrium molecular dynamics formalism; they seem to lead to comparable numerical results.

Appendix A: The Nonequilibrium Statistical Operator

The construction of nonequilibrium statistical ensembles, that is, a nonequilibrium statistical ensemble formalism, NESEF for short, which essentially consists of a derivation of a nonequilibrium statistical operator (probability distribution in the classical case), has been attempted along several lines. In a brief summarized way, we describe the construction of the NESEF within a heuristic approach. First, it should be noticed that for systems away from equilibrium, several important points need to be carefully taken into account in each case under consideration:

1. **The choice of the basic variables** (a wholly different choice from the case of equilibrium when it suffices to take a set of variables that are constants of motion), which is based on an analysis of what sort of macroscopic measurements and processes are actually possible; moreover, one should focus attention not only on what can be observed but also on the character and expectation concerning the equations of evolution for these variables [12,13,25].

2. **The question of irreversibility** (or Eddington’s arrow of time):

3. **Historicity needs be introduced**, that is, the idea that the past dynamics of the system (or historicity effects) must be incorporated along the time interval going from an initial description of the macrostate of the sample in a given experiment, say at $t_0$, to the time $t$ when a measurement is performed.

Concerning the choice of the basic variables, in contrast to the case of equilibrium, immediately after an open system of $N$ particles, in contact with external sources and reservoirs, has been driven out of equilibrium, it becomes necessary to describe its state in terms of all the observables and, eventually, introduce direct and cross-correlation. However, as time elapses, Bogoliubov’s principle of correlation weakening allows us to introduce increasing contractions of the descriptions. Let us say that we can introduce a description based on the observables $\{P_j\}, j = 1, 2, ..., n$, on which the nonequilibrium statistical operator depends.

On the question of irreversibility, in the absence of a proper way to introduce such an effect, one needs to resort to the interventionist’s approach, which is based on the ineluctable process of randomization leading to the asymmetric evolution of the macrostate.

The “intervention” consists of introducing into the Liouville equation of the statistical operator, of an otherwise isolated system, a particular source accounting for Krylov’s “jolting” effect [52], in the form (written for the logarithm of the statistical operator)

$$\frac{\partial}{\partial t} \ln \mathcal{R}_\varepsilon(t) + \frac{1}{i\hbar} \ln \mathcal{R}_\varepsilon(t), \hat{H} = -\varepsilon[\ln \mathcal{R}_\varepsilon(t) - \ln \overline{\mathcal{R}}(t, 0)], \quad (A1)$$

where $\varepsilon$ (a kind of reciprocal of a relaxation time) is taken to go to $+0$ after the calculations of average values have been performed. Such a mathematically inhomogeneous term, in the otherwise normal Liouville equation, implies
a continuous tendency of relaxation of the statistical operators towards a referential distribution, \( \mathcal{R} \), which represents an instantaneous quasi-equilibrium condition.

We can see that Eq. (A.1) consists of a regular Liouville equation but with an infinitesimal source, which provides Bogoliubov’s symmetry breaking of time reversal and is responsible for disregarding the advanced solutions. This is described by a Poisson distribution, and the result at time \( t \) is obtained by averaging over all \( t' \) in the interval \((t_0, t)\), and the solution of Eq. (A.1) is

\[
\mathcal{R}_\varepsilon(t) = \exp \left\{ -\hat{S}(t, 0) + \int_{t_0}^{t} dt' e^\varepsilon(t' - t) \frac{d}{dt'} \hat{S}(t', t' - t) \right\},
\]

where

\[
\hat{S}(t, 0) = -\ln \mathcal{R}(t, 0),
\]

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

and the initial-time condition at time \( t_0 \), when the application of the formalism begins, is

\[
\mathcal{R}_\varepsilon(t_0) = \mathcal{R}(t_0, 0).
\]

In \( \mathcal{R} \) and \( \hat{S} \), the first time variable in the argument refers to the evolution of the nonequilibrium thermodynamic variables, and the second time variable refers to the time evolution of the dynamical variables, both of which have an effect on the operator. The statistical operator can be written in the form

\[
\mathcal{R}_\varepsilon(t) = \mathcal{R}(t, 0) + \mathcal{R}'_\varepsilon(t).
\]

Involving the auxiliary probability distribution \( \tilde{\varrho}(t, 0) \), with \( \varrho'_R(t) \), which contains the historicity and irreversibility effects. Moreover, in most cases, we can consider a system that consist of the system of interest (on which we are performing an experiment) in contact with ideal reservoirs. Thus, we can write

\[
\mathcal{R}(t, 0) = \tilde{\varrho}(t, 0) \times \varrho_R.
\]

and

\[
\mathcal{R}_\varepsilon(t) = \varrho_\varepsilon(t) \times \varrho_R,
\]

where \( \varrho_\varepsilon(t) \) is the statistical operator of the nonequilibrium system, \( \tilde{\varrho} \) is the auxiliary operator, and \( \varrho_R \) is the stationary operator of the ideal reservoirs, with \( \varrho_\varepsilon(t) \) given by

\[
\varrho_\varepsilon(t) = \exp \left\{ -\hat{S}(t, 0) + \int_{-\infty}^{t} dt' e^\varepsilon(t' - t) \frac{d}{dt'} \hat{S}(t', t' - t) \right\},
\]

with the initial value \( \tilde{\varrho}(t_0, 0) \) \( (t_0 \to -\infty) \), and where

\[
\hat{S}(t, 0) = -\ln \tilde{\varrho}(t, 0).
\]

Finally, the auxiliary statistical operator \( \tilde{\varrho}(t, 0) \) must be provided. This operator defines an instantaneous distribution at time \( t \), which describes a “frozen” equilibrium defining the macroscopic state of the system at the given time, and for that reason the operator is sometimes dubbed as the quasi-equilibrium statistical operator. On this basis (or, alternatively, via the variational procedure \([12,13,14]\)) and considering the description of the nonequilibrium state of the system in terms of the basic set of dynamical variables \( \hat{P}_j \), the reference or instantaneous quasi-equilibrium statistical operator is taken as a canonical-like operator given by

\[
\tilde{\varrho}(t, 0) = \exp \left\{ -\phi(t) - \sum_j F_j(t) \hat{P}_j \right\},
\]

with \( \phi(t) \) ensuring the normalization of \( \tilde{\varrho} \) and playing the role of a logarithm of a partition function, say, \( \phi(t) = \ln \tilde{Z}(t) \). Moreover, in Eq. (A.11), \( F_j \) are the nonequilibrium thermodynamic variables associated with each kind of basic dynamical variable \( \hat{P}_j \). The nonequilibrium thermodynamic space of states consists of the basic variables \( \{Q_j(t)\} \), which consist of averages of \( \{\hat{P}_j\} \) over the nonequilibrium ensemble, namely,

\[
Q_j(t) = \text{Tr} \{\hat{P}_j \varrho_\varepsilon(t)\},
\]

which are then functionals of \( \{F_j(t)\} \), and the equations of state are

\[
Q_j(t) = -\frac{\delta \phi(t)}{\delta F_j(t)} = -\frac{\delta \ln \tilde{Z}(t)}{\delta F_j(t)},
\]

where \( \delta \) stands for a functional derivative \([45]\).

Moreover,

\[
\hat{S}(t) = \text{Tr} \{\hat{S}(t, 0) \tilde{\varrho}(t, 0)\} = -\text{Tr} \{\tilde{\varrho}(t, 0) \ln \tilde{\varrho}(t, 0)\},
\]

is the so-called informational entropy characteristic of the distribution \( \tilde{\varrho} \), a functional of the basic variables \( \{Q_j(t)\} \), and the alternative form of the equations of state is given by

\[
-\frac{\delta \hat{S}(t)}{\delta Q_j(t)} = F_j(t).
\]

In the contracted description used for the construction of HOGH of order 2, that is, in terms of the variables of Eq. (36), we have that the auxiliary statistical operator, better written in reciprocal space, is

\[
\tilde{\varrho}(t, 0) = \exp \left\{ -\phi(t) - \sum_{Q} \{F_{h\varepsilon}(Q, t) \tilde{h}(Q) + F_{\varepsilon}(Q, t) \tilde{n}(Q) + F_{\varepsilon}[2](Q, t) \otimes \tilde{I}_{n}[2](Q) \right\}.
\]

(A16)
Quantities with a triangular hat are the mechanical operators corresponding to the quantities in Eq. (3). In Eq. (A.16), the conjugated nonequilibrium thermodynamical variables, \( F \)'s listed in Eq. (37), are introduced.

**Appendix B: Single-Particle Kinetic Equation**

The NESEF-based single-particle kinetic equation was reported in Ref. [34], which contained an interaction with a surrounding thermal bath and assumed that in a dilute solution the interparticle interaction could be treated in the weak-coupling approximation. We reproduce below this result but incorporate the collision integral associated with the two-particle interaction in full. We have

\[
\frac{\partial}{\partial t} f_1(r, p; t) = J^{(0)}_1(r, p; t) + J^{(1)}_1(r, p; t) + J^{(2)}_1(r, p; t),
\]

where the first terms on the right-hand side are

\[
J^{(0)}_1(r, p; t) = \text{Tr} \left\{ \{ \tilde{a}_1(r, p), \tilde{H}_0 \} \tilde{\theta}(t, 0) \mathbf{Q} R \right\} = -\frac{p}{m} \cdot \nabla f_1(r, p; t),
\]

which are, as already noticed, the precession term in Moris’s terminology, and

\[
J^{(1)}_1(r, p; t) = \text{Tr} \left\{ \{ \tilde{a}_1(r, p), \tilde{H} \} \tilde{\theta}(t, 0) \mathbf{Q} R \right\} = [\nabla U_{\text{ex}}(r, t) + \nabla U(r, t)] \cdot \nabla p f_1(r, p; t),
\]

is the contribution containing the action of the external and internal forces, where

\[
U(r, t) = \int d^3 r' d^3 p' V(|r - r'|) f_1(r', p'; t)
\]

plays the role of a mean-field potential of the interaction between particles or a Vlasov-like potential. Observe that \( \circ \) for the scalar product of vectors, and we write \( \cdot \) for the full contraction of tensors.

Taking all the interactions in Eq. (26), denoted by \( H' \), that is, pair interactions with applied external sources, lengthy but straightforward calculations give the final expression of the collision integral \( J^{(2)}_1 \). Taking this final expression, Eqs. (B2) and (B3) into Eq. (B1) leads to the kinetic equation:

\[
\frac{\partial}{\partial t} f_1(r, p; t) + \frac{p(r, p; t)}{m} \cdot \nabla f_1(r, p; t) + F(r, p; t) \cdot \nabla p f_1(r, p; t) - B(p) f_1(r, p; t) +
\]

\[-A_1^{(2)}(p) \circ [\nabla_p \nabla] f_1(r, p; t) - B_1^{(2)}(p) \circ [\nabla_p \nabla_p] f_1(r, p; t) = J^{(2)}_S(r, p; t),
\]

where

\[
P(p; t) = p - m A_1(p),
\]

plays the role of a generalized momentum,

\[
F(r, p; t) = -\nabla U_{\text{ex}}(r, t) - B_1(p) - F_{\text{NL}}(r, t) - \nabla U(r, t),
\]

is a generalized force, in which

\[
F_{\text{NL}}(r; t) = \int d^3 r' d^3 p' G_{\text{NL}}(r' - r, p') f_1(r', p'; t),
\]

and the coefficients are as follows:

\[
A_1(p) = -\frac{n_B M \beta_0}{\mathcal{V}} \sum_Q \frac{\left| \psi(Q) \right|^2}{Q^2} \cdot \nabla_p F(p, Q),
\]

where

\[
F(p, Q) = 1 + \sum_{n=1}^{\infty} \frac{(-1)^n}{(2n-1)!!} \left( \frac{M \beta_0}{Q^2 m^2} \right)^n (Q \cdot p)^{2n},
\]

\[
B_1(p) = \frac{n_B \pi \sqrt{M \beta_0}}{m \sqrt{2 \pi}} \times \sum_Q \frac{\left| \psi(Q) \right|^2}{Q} \left( \frac{1}{M} - \frac{1}{m} \right) (Q \cdot p) e^{-\frac{M \beta_0 (Q \cdot p)^2}{2 Q^2 m^2}},
\]

\[
G_{\text{NL}}(r' - r, p') = \frac{n_B R \beta_0}{\mathcal{V}} \sum_Q \left| \psi(Q) \right|^2 \left\{ i F(Q, p') + \left( \frac{M \beta_0}{2 \pi} \right)^{1/2} \frac{Q \cdot p'}{Q} e^{-\alpha (Q \cdot p')^2} \right\} e^{i Q \cdot (r' - r)},
\]

\[
A_2^{(2)}(p) = \frac{n_B M \beta_0}{\mathcal{V}} \sum_Q \frac{\left| \psi(Q) \right|^2}{Q^2} F(p, Q) [Q Q],
\]

\[
B_2^{(2)}(p) = \frac{n_B \sqrt{M \beta_0}}{m \mathcal{V} \sqrt{2 \pi}} \sum_Q \frac{\left| \psi(Q) \right|^2}{Q^2} e^{-\frac{M \beta_0 (Q \cdot p)^2}{2 Q^2 m^2}} [Q Q],
\]

\[
B(p) = \frac{n_B \pi \sqrt{M \beta_0}^3}{m M \sqrt{2 \pi}} \times \sum_Q \frac{\left| \psi(Q) \right|^2}{Q} \left( Q^2 - \frac{M \beta_0}{m^2} (Q \cdot p)^2 \right) e^{-\frac{M \beta_0 (Q \cdot p)^2}{2 Q^2 m^2}}.
\]

In Eqs. (B.13) and (B.14) \([Q Q]\) stands for a tensor product of vectors, i.e., a rank-2 tensor. Moreover, in these equations, \( \psi(Q) \) is the Fourier transform of the potential energy \( w(|r_j - R_k|) \), \( \mathcal{V} \) is the volume and \( n_B = N_B / \mathcal{V} \).
Appendix C: Summary of Heims-Jaynes Procedure

Given a statistical operator of the form
\[ \hat{\varrho} = \frac{1}{Z} e^{\hat{\Lambda} + \hat{B}}, \tag{C1} \]
where
\[ Z = \text{Tr}\{e^{\hat{\Lambda} + \hat{B}}\}, \tag{C2} \]
ensures its normalization and introducing
\[ \hat{\varrho}_0 = \frac{e^{\hat{\Lambda}}}{\text{Tr}\{e^{\hat{\Lambda}}\}}, \tag{C3} \]
according to Heims-Jaynes, given an operator \( \hat{\Theta} \), it follows that
\[ \text{Tr}\{\hat{\Theta}\hat{\varrho}\} = \langle \hat{\Theta}\rangle_0 + \sum_{n=1}^{\infty} \langle \hat{Q}_n (\hat{\Theta} - \langle \hat{\Theta}\rangle_0) \rangle, \tag{C4} \]
where
\[ \langle \hat{\Theta}\rangle_0 = \text{Tr}\{\hat{\Theta}\hat{\varrho}_0\}, \tag{C5} \]
with
\[ \hat{Q}_n = \hat{S}_n - \sum_{k=1}^{n-1} \langle \hat{Q}_n\rangle_0 \hat{S}_{n-k}, \tag{C6} \]
for \( n \geq 2 \), and \( \hat{Q}_0 = \hat{1} \) and \( \hat{Q}_1 = \hat{S}_1 \),
\[ \hat{S}_n = \frac{B^n}{n!}, \quad \hat{S}_0 = \hat{1}. \tag{C7} \]

Equation (C4) consists of the average value of \( \hat{\Theta} \) with \( \varrho_0 \) (that is, only depending on \( A \)) plus a contribution in the form of a series expansion in powers of \( B \). In a first-order approximation, we have that
\[ \text{Tr}\{\hat{\Theta}\hat{\varrho}\} \simeq \langle \hat{\Theta}\rangle_0 + \text{Tr}\{\hat{B}(\hat{\Theta} - \langle \hat{\Theta}\rangle_0)\varrho_0\}. \tag{C8} \]

In Section III, we have used
\[ \overline{\varrho}(t, 0) = \frac{1}{Z(t)} e^{\hat{\Lambda} + \hat{B}}, \tag{C9} \]
where
\[ \hat{\Lambda}(t) = - \sum_Q F_h(Q, t) \hat{n}(Q), \tag{C10} \]
\[ \hat{B}(t) = - \sum_Q [F_n(Q, t) \hat{n}(Q) + F_n(Q, t) \cdot \hat{I}_n(Q)] + \hat{F}^{[2]}_n(Q, t) \otimes \hat{I}^{[2]}_n(Q), \tag{C11} \]
and
\[ \overline{Z}(t) = e^{\phi(t)}, \tag{C12} \]
with the calculations performed in the first-order (linear) Heims-Jaynes expansion.

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