Consistent multiphase Lattice Boltzmann model for gas-liquid systems

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A new lattice Boltzmann method for simulating multiphase flows is developed theoretically. The method is adjusted such that its continuum limit is the Navier-Stokes equation, with a driving force derived from the Cahn-Hilliard free energy. In contrast to previous work, however, the bulk and interface terms are decoupled, the former being incorporated into the model through the local equilibrium populations, and the latter through a forcing term. We focus on gas-liquid phase equilibria with the possibility to implement an arbitrary equation of state. The most novel aspect of our approach is a systematic Chapman-Enskog expansion up to the third order. Due to the third-order gradient in the interface forcing term, this is needed to obtain a model that is fully consistent with both hydrodynamics and thermodynamics. In order to satisfy all conditions, we need 59 velocities in three dimensions, and 21 velocities for simulating two-dimensional systems. Even with such a large number of velocities, there are restrictions on the equation of state that can only be lifted by increasing the set further. Moreover, we find that it is necessary to solve a self-consistent equation for the hydrodynamic flow velocity, in order to enforce the identity of momentum density and mass current on the lattice. The analysis completely identifies all spurious terms in the Navier-Stokes equation, and thus shows how to systematically eliminate each of them, by constructing a suitable collision operator. The commonly noticed inconsistency of most existing models is thus traced back to their insufficient number of degrees of freedom. Therefore, the gain of the new model is in its clear derivation, full thermo-hydrodynamic consistency, and expected complete elimination of spurious currents in the continuum limit. Numerical tests are deferred to future work.

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I. INTRODUCTION

The Lattice Boltzmann (LB) method [1–6] is based upon solving a fully discretized version of the Boltzmann equation known from the kinetic theory of gases. Space is discretized in terms of the sites of a regular lattice with spacing $a$ and time in terms of a finite time step $h$, while the velocity space is reduced to a small set of discrete velocities $c_i$ that are chosen such that one time step will always connect sites on the lattice (i.e. $c_i h$ is a lattice vector). The central objects of the algorithm are the populations $n_i(r,t)$ at site $r$ at time $t$, corresponding to the velocity $c_i$, and the algorithm proceeds by an alteration of streaming and collision steps. Typically, $n_i$ is assigned the physical interpretation of a mass density.

The simplest version of this algorithm is for an isothermal ideal gas, where the collision step is done by linearly relaxing the populations towards a set of local pseudo-equilibrium populations $n_i^{eq}$, which in turn are determined from the local mass density $\rho = \sum_i n_i$ and the local flow velocity $u = \rho^{-1} j = \rho^{-1} \sum_i n_i c_i$. This scheme can be analyzed in detail by a multiple timescale Chapman-Enskog (CE) expansion (see, e.g. Ref. [4]), from which one finds that the algorithm provides a valid solution to the isothermal Navier-Stokes equation (NSE) in the continuum limit, provided that the Mach number (flow velocity $u$ relative to the speed of sound $c_s$) is sufficiently small, i.e. terms of order $u^3$ may be safely neglected. In turn, this means that the scheme is confined to flows that are (close to) incompressible. It is this analysis that provides essentially all of the deeper insights: one starts by introducing a dimensionless scale separation parameter $\varepsilon \ll 1$ and setting $r_1 = \varepsilon r$. At fixed $r_1$, the limit $\varepsilon \to 0$ then automatically implies an analysis at large length scales. Since in standard hydrodynamics two time scales are involved, one for sound waves, where time $\propto (length)^{1/2}$, and a slower one for diffusive momentum transfer, where time $\propto (length)^2$, the analysis takes this into account by explicitly introducing two time variables, $t_1 = \varepsilon t$ (wave-like scaling) and $t_2 = \varepsilon^2 t$ (diffusive scaling), and formally treating the dynamic variables as depending on $t_1$ and $t_2$ independently.

The limiting behavior is then obtained by a leading-order Taylor expansion with respect to $\varepsilon$. It should be noted that the CE expansion may also be viewed as an expansion with respect to gradients — wave-like scaling corresponds to first-order gradients, while diffusive terms imply second-order gradients in the NSE. For the ideal-gas LB algorithm, the CE analysis then provides a host of important results [4]):

- The non-dissipative Euler equation is obtained at first order of the CE expansion, while dissipation corresponds to the second order.
- The Euler dynamics is completely encoded in the algebraic form of the equilibrium populations, i.e. in the dependence of $n_i^{eq}$ on $\rho$ and $u$ — and the analysis also shows that $n_i^{eq}$ may only depend on the conserved quantities, which are mass and momentum.
In order to avoid spurious terms in the Euler equation, one needs at least three velocity shells. In this case, the equation of state is fixed to $p = \rho c_s^2$, where $p$ is the thermodynamic pressure, while $c_s$ cannot be chosen at will, but takes the value $c_s = 3^{-1/2}$ in lattice units for the D3Q19 model [6]. If another value is desired, more shells are needed.

Viscous dissipation corresponds to the relaxation towards local equilibrium, and the analysis provides explicit expressions for the relation between the shear and bulk viscosities in the NSE on the one hand, and the LB relaxation rates on the other.

Since roughly two decades, there has been extensive work that tries to extend this algorithm to the case of multiphase flows, where the interest was mainly focused on the case of a binary mixture on the one hand, and the case of gas-liquid phase coexistence in a one-component system on the other. The literature on this topic is vast, and hence we do not attempt here to provide anything like a comprehensive review. Instead, we try to re-examine the LB method for multiphase flows. In view of this unsatisfactory situation we here try to transfer this observation to the case of a non-ideal gas, i.e. to assume that the non-ideal gas is characterized by an equation of state $p = \rho c_s^2(\rho)$ with a non-trivial density dependence of $c_s^2$, and, furthermore, to assume that the equilibrium populations have the same form as in the ideal-gas case, only with a modified $c_s^2$.

This means that consistency with thermodynamics and Galilean invariance will necessarily involve more than three velocity shells.

We also notice that with this interpretation it is possible to achieve $u^2 \ll c_s^2$ throughout the phase diagram, which means that it should be justified to neglect $O(u^3)$ terms, just as in the CE analysis of the ideal gas.

Furthermore, the ideal-gas case tells us that we should view $n_i^{eq}$ as the solution to a maximum-entropy problem [28–31]. Transferring this to the non-ideal gas, we find that the thermodynamics should probably be viewed as derived from a non-ideal (i.e. non-Boltzmann) entropy, which we expect (in future work) to ultimately pave the way towards a consistent formulation of the stochastic version of the method, which includes thermal fluctuations [31]. This notion should not come as a big surprise to physicists with a background in modern soft-matter theory, where the concepts of energy and entropy are often used interchangeably (what ultimately matters is just the free energy).

Aiming at thermodynamic consistency, it is then most natural to derive the macroscopic equations of motion from a Ginzburg-Landau type free energy functional, as in the LB approaches pioneered by Swift and Yeomans [9, 10]. Such a functional describes the bulk thermodynamics by a free energy density $f(\rho)$, which, below the critical point, exhibits a double-well structure, while phase separation is driven by an interface free energy, which is typically modeled by a gradient-square term in the functional.

Since $n_i^{eq}$ corresponds to the leading order of the CE expansion, which does not contain any gradients, $n_i^{eq}$ should correspond to only the bulk free energy, or the bulk equation of state (and we have already outlined how to do this). In contrast, it should not depend on the interfacial term, as it was introduced in the original Swift-Yeomans model [9, 10].
• Rather, we model the interface term similar to the effect of an external force. This is somewhat reminiscent of the work by Lee and Fischer [11]; however, they confined the driving not to the interface term (as we do), but rather to all terms that deviate from the ideal gas (which, in our opinion, is inconsistent as well).

• On the NSE level, the interfacial driving shows up via a term that involves a third-order gradient of the density. This means however that the CE analysis has to be done up to third order as well, which is obviously a cumbersome task.

• One is therefore naturally led to the introduction of a third time scale, \( t_3 = \varepsilon^3 t \), which is yet slower than momentum diffusion. We believe that the physical interpretation of the process corresponding to \( t_3 \) is simply domain coarsening, which is typically the slowest process in a phase-separating system.

• Apart from the obvious terms in the collision operator (relaxation towards \( n_i^{eq} \), interfacial force-like driving), we also construct a “correction” collision operator that cancels various spurious terms, such that the final equation of motion up to order \( \varepsilon^3 \) is just the NSE. This correction is analogous to the correction that is known in the case of driving an ideal gas via an external force [4, 32]. It is essentially impossible to guess the form of such a correction without doing the CE analysis, and it is (we believe) a non-trivial result that it is possible to construct such an operator at all. We believe that this approach is most likely the only way to systematically eliminate all numerical artifacts from the method. A numerical test is however deferred to future work.

It should be noted that in previous work [19] it has already been realized that multiphase LB models should be subjected to a higher-order CE analysis. However, it seems that the present paper is the first attempt in which this program is actually being carried out. The analysis will show that the higher-order CE expansion also implies more stringent requirements on the isotropy of lattice tensors. While for the ideal gas (second-order CE expansion) isotropy up to fourth-rank tensors is needed, the present model requires isotropy up to sixth-rank tensors (and a concomitant larger set of velocities). The fact that improved isotropy is helpful to construct better multiphase LB models has been noted before as well [20, 22]. For our needs, the work of Chen, Goldhirsh and Orszag [33] on isotropy of lattice tensors turned out to be particularly useful.

The remainder of this paper is organized as follows: Section II outlines the general LB setup of our model, and discusses the target NSE. In Sec. III we then discuss how to find a proper set of velocities, and how to implement the equation of state in terms of the equilibrium populations. Section IV then presents a brief excursion on central-difference approximations to various gradients that need to be evaluated on the lattice. Section V discusses how the momentum transfer derived from the interface force is split up between the various contributions of the collision operator, and from this we construct the interface force collision operator in Sec. VI. The central part of the paper is then Sec. VII, in which the CE analysis is done. The results derived there then allow us to construct the correction collision operator, which is done in Sec. VIII. Section IX outlines the implicit algorithm that needs to be applied in order to find the hydrodynamic flow velocity with third-order CE accuracy. Finally, we summarize in Sec. X. Appendix A derives the third-order interface force from the Cahn-Hilliard model, while App. B works out how to construct an equation of state that is compatible with the various restrictions derived in the main text. Some algebraic details that have been omitted in the main text are presented in App. C.

II. BASIC CONSIDERATIONS

Our starting point are the hydrodynamic equations of motion that should be simulated by the multiphase LB method. Mass conservation is expressed by the continuity equation

\[ \partial_t \rho + \partial_\alpha (\rho u_\alpha) = 0, \tag{1} \]

where \( \rho(r, t) \) is the mass density, \( u(r, t) \) the flow velocity, Greek letters denote Cartesian indexes, the Einstein summation convention is implied, and \( \partial_i \equiv \partial / \partial t \), \( \partial_\alpha \equiv \partial / \partial r_\alpha \). Momentum conservation implies an equation of motion for the momentum density \( j = \rho u \), of the form

\[ \partial_t (\rho u_\alpha) + \partial_\beta (\rho u_\alpha u_\beta) + \partial_\alpha p = \partial_\beta \sigma_{\alpha \beta} + f_\alpha, \tag{2} \]

where \( p \) is the thermodynamic pressure, \( f \) the interfacial force density and \( \sigma \) the viscous stress tensor involving the shear viscosity \( \eta \) and the bulk viscosity \( \eta_V \):

\[ \sigma_{\alpha \beta} = \eta \left[ \partial_\alpha u_\beta + \partial_\beta u_\alpha - \frac{2}{d} \partial_\gamma u_\gamma \delta_{\alpha \beta} \right] + \eta_V \partial_\gamma u_\gamma \delta_{\alpha \beta}, \tag{3} \]

where \( \delta_{\alpha \beta} \) denotes the Kronecker symbol and \( d \) the spatial dimension. Pressure and interface forces are related to the Cahn-Hilliard Hamiltonian [34]

\[ \mathcal{H} = \int d^d r \left[ \frac{1}{2} \rho u^2 + \rho e + \frac{\kappa}{2} (\nabla \rho)^2 \right], \tag{4} \]

where the first term denotes the kinetic energy density, \( e = e(\rho) \) is the internal energy per unit mass and \( \kappa \) the interfacial stiffness. In the absence of viscous dissipation, the dynamics should conserve the total Hamiltonian,

\[ \frac{d}{dt} \mathcal{H} = 0, \tag{5} \]
and since the pressure is related to $e$ via
\[ p = \rho^2 \frac{\partial e}{\partial \rho}, \tag{6} \]
this condition allows the determination of $f$ (see App. A):
\[ f = \kappa \rho \nabla \nabla^2 \rho. \tag{7} \]

LB simulations are based on the kinetic theory of gases and the Boltzmann equation. The algorithm can be summarized by the following discretized version of the Boltzmann equation:
\[ n_i(r + c_i h, t + h) = n_i^*(r, t) = n_i(r, t) + \Delta_i, \tag{8} \]
where $n_i(r, t)$ indicates the pre-collisional populations — mass density of particles at site $r$ and at time $t$ that have the velocity $c_i$. $n_i^*$ indicates the post-collisional populations. The difference between the pre- and post-collisional populations $\Delta_i$ is the collision operator. The algorithm is hence performed in two steps from the right to the left hand side; the first step is the collision step and the second step is the streaming step. In the streaming step the post-collisional populations $n_i^*$ are moved to neighboring sites $r + c_i h$, where $h$ is the time step of the simulation. \{cih\} is a discrete set of vectors which connect each lattice site with its neighboring sites. The connection between this microscopic equation and the macroscopic equations of motion (Eqs. 1 and 2) is found via the CE analysis. After Taylor expansion with respect to the scaling parameter $\varepsilon \ll 1$, one studies various velocity moments of the populations and their equations of motion at different levels of the multiple time-scale analysis. At the end the velocity moments of different orders are gathered back together, so that the zeroth velocity moment results in the continuity equation (Eq. 1) and the first velocity moment in the Navier-Stokes equation (Eq. 2). As discussed in the Introduction, the third-order gradient in the interface force requires that we introduce three time scales for our CE analysis, and hence we write
\begin{align*}
  r_1 &= \varepsilon r, \tag{9} \\
  t_1 &= \varepsilon t, \tag{10} \\
  t_2 &= \varepsilon^2 t, \tag{11} \\
  t_3 &= \varepsilon^3 t, \tag{12}
\end{align*}
such that the corresponding space and time derivatives can be written as
\begin{align*}
  \partial_s &= \varepsilon \partial_{s_1}, \tag{13} \\
  \partial_t &= \varepsilon \partial_{t_1} + \varepsilon^2 \partial_{t_2} + \varepsilon^3 \partial_{t_3}. \tag{14}
\end{align*}
The time scales $t_1$ and $t_2$ are already known from the ideal gas CE expansion [4] and correspond to sound waves and diffusion of momentum, respectively, while the physical interpretation of the newly introduced time scale $t_3$ can be found in the coarsening process of the multiphase system. With this the lattice Boltzmann equation (Eq. 8) is rewritten as
\[ n_i(r_1 + \varepsilon c_i h, t_1 + \varepsilon h, t_2 + \varepsilon^2 h, t_3 + \varepsilon^3 h) - n_i(r_1, t_1, t_2, t_3) = \Delta_i, \tag{15} \]
and its CE analysis will be done in Sec. VII.

The collision operator $\Delta_i$ is composed of the bulk, the interface and the correction term:
\[ \Delta_i = \Delta_i^{bulk} + \Delta_i^{int} + \Delta_i^{corr}. \tag{16} \]
For the sake of simplicity we just use the standard BGK (Bhatnagar-Gross-Krook) operator for $\Delta_i^{bulk}$:
\[ \Delta_i^{bulk} = (\gamma - 1)(n_i - n_i^{eq}), \tag{17} \]
where $\gamma$ with $-1 < \gamma < 1$ is the relaxation parameter. The detailed derivation is done in Secs. V and VI. The correction collision operator is constructed so that all spurious terms, which would occur in the continuum equations of motion in case there was no such correction operator, are cancelled out exactly up to the third order in the CE expansion. The final construction of the correction collision operator is done after the CE expansion based on the same principle as previously used to implement an external force in the ideal gas LB algorithm [4, 32] (see Sec. VIII).

It turns out that the construction of the correction collision operator is quite elaborate and requires an LB algorithm which is isotropic up to sixth rank tensors, in contrast to standard algorithms which usually satisfy the isotropy up to the fourth rank only. Because of this a large number of velocities $c_i$ is necessary. The relation between lattice velocity sets and the tensors of their equations of motion in case there was no such correction operator, are cancelled out exactly up to the third order in the CE expansion. The final construction of the correction collision operator is done after the CE expansion based on the same principle as previously used to implement an external force in the ideal gas LB algorithm [4, 32] (see Sec. VIII).

In three dimensions, analogous considerations yield an even larger set of 59 velocities on the simple-cubic lattice. In what follows, we will restrict attention to these two lattices.
flow velocity $\mathbf{u}$ via the prescription (see also Sec. V)

$$\mathbf{j} = \rho \mathbf{u} = \sum_i n_i \mathbf{c}_i + \frac{h}{2} \mathbf{f} + \tilde{\mathbf{j}}. \quad (20)$$

We first discuss the set of velocities and weights that we use in our model. It is clear that for symmetry reasons the weights must be identical for vectors within the same shell, i.e. vectors that have the same length. Lattice tensors composed from the velocities $\mathbf{c}_i$ via moments weighted with the $w_i$ have to satisfy sixth rank isotropy (this is needed for the proper construction of the reaction collision operator as described in Sec. VIII). Odd moments trivially vanish for reasons of reflection symmetry of the lattice. For the even orders cubic symmetry and proper normalization implies

$$\sum_i w_i = 1, \quad (21)$$

$$\sum_i w_i c_{i\alpha} c_{i\beta} = \sigma_2 \delta_{\alpha\beta}, \quad (22)$$

$$\sum_i w_i c_{i\alpha} c_{i\beta} c_{i\gamma} c_{i\delta} = \kappa_4 \delta_{\alpha\beta} \delta_{\gamma\delta} \quad (23)$$

$$+ \sigma_4 (\delta_{\alpha\beta} \delta_{\gamma\delta} + \text{perm.}),$$

$$\sum_i w_i c_{i\alpha} c_{i\beta} c_{i\gamma} c_{i\delta} c_{i\mu} c_{i\nu} = \kappa_6 \delta_{\alpha\beta} \delta_{\gamma\delta} \delta_{\mu\nu} \quad (24)$$

$$+ \eta_6 (\delta_{\alpha\beta} \delta_{\gamma\delta} \delta_{\mu\nu} + \text{perm.}) + \sigma_6 (\delta_{\alpha\beta} \delta_{\gamma\delta} \delta_{\mu\nu} + \text{perm.}) .$$

Here the various $\delta$ symbols are one if all indexes are identical and zero otherwise. Furthermore, the term “perm.” means that one has to take into account all possibilities to assign subsets of indexes to the various $\delta$ symbols, such that, for example, the bracket near $\sigma_4$ contains three terms and the bracket near $\sigma_6$ contains 15 terms.

The isotropy condition requires that $\kappa_4 = 0$, $\kappa_6 = 0$, $\eta_6 = 0$. Furthermore, we require $\sigma_4 = \sigma_2^2$ as well as $\sigma_6 = \sigma_4^2$. These latter two conditions can be motivated from the continuum analog, i.e. the Maxwell-Boltzmann velocity distribution at vanishing flow velocity, $\mathbf{u} = 0$. This is a Gaussian distribution, whose tensorial moments result from Wick’s theorem, and have the same form as given above. We also require, in analogy to the ideal-gas case, that $\sigma_2^2 = \sigma_2$. The moment relations are therefore simplified to

$$\sum_i w_i = 1, \quad (25)$$

$$\sum_i w_i c_{i\alpha} c_{i\beta} = c_2^2 \delta_{\alpha\beta}, \quad (26)$$

$$\sum_i w_i c_{i\alpha} c_{i\beta} c_{i\gamma} c_{i\delta} = c_4^2 (\delta_{\alpha\beta} \delta_{\gamma\delta} + \text{perm.}), \quad (27)$$

$$\sum_i w_i c_{i\alpha} c_{i\beta} c_{i\gamma} c_{i\delta} c_{i\mu} c_{i\nu} = c_6^2 (\delta_{\alpha\beta} \delta_{\gamma\delta} \delta_{\mu\nu} + \text{perm.}) . \quad (28)$$

For a freely picked value of $c_2^2$, we thus have six conditions for the weights $w_i$ ($\kappa_4 = 0$, $\kappa_6 = 0$, $\eta_6 = 0$, $\sigma_4 = c_4^2$,
We here use lattice units, i.e., velocities is $39$. The total number of velocities is $39$. $b$ is the number of velocities within one shell. For each shell, we only list one representative lattice vector. We here use lattice units, i.e., $h = a = 1$.

$\sigma_6 = c^6$, and normalization), resulting in seven “degrees of freedom” or velocity shells that are expected to be needed in order to satisfy all constraints. As a matter of fact, however, we use eight shells in three dimensions, while in two dimensions it is possible to satisfy all conditions with only six shells. The set of shells and the corresponding weights are given in Tabs. I and II for the 3D and 2D cases, respectively. They have been constructed by making use of the work of Chen et al. [33], and we refer interested readers to that paper. Briefly, the D3Q59 model is obtained by projecting a set of 4D lattice velocities which satisfy sixth rank isotropy down to 3D, while the D2Q21 model is obtained by further projecting the D3Q59 model down to two dimensions. In particular, the weights for the lower-dimensional sets are directly obtained from the weights of the original 4D model.

With these conditions on the moments of the $w_i$, we can evaluate moments of $n_i^{eq}$. As in the ideal-gas case, the low-order moments are the hydrodynamic variables mass density, momentum density, and Euler stress:

$$\sum_i n_i^{eq} = \rho,$$

$$\sum_i n_i^{eq} c_{\alpha} = j_{\alpha} = \rho u_{\alpha},$$

$$\sum_i n_i^{eq} c_{\alpha} c_{\beta} = \Pi_{\alpha\beta}^{eq} = p c^2_{s,eq}(\rho) \delta_{\alpha\beta} + \rho u_{\alpha} u_{\beta}$$

$$= p(\rho) \delta_{\alpha\beta} + \rho u_{\alpha} u_{\beta}. \quad (31)$$

Furthermore, we evaluate the third and fourth order moments as

$$\sum_i n_i^{eq} c_{\alpha} c_{\beta} c_{\gamma} c_{\delta} = \Phi_{\alpha\beta\gamma\delta}^{eq}$$

$$= \rho c^2_{s,eq}(\rho) \delta_{\alpha\beta} u_{\gamma} u_{\delta} + \delta_{\alpha\gamma} u_{\beta} u_{\delta} + \delta_{\alpha\delta} u_{\beta} u_{\gamma}, \quad (32)$$

$$\sum_i n_i^{eq} c_{\alpha} c_{\beta} c_{\gamma} c_{\delta} c_{\epsilon} = \Phi_{\alpha\beta\gamma\delta\epsilon}^{eq}$$

$$= \rho c^2_{s,eq}(\rho) \delta_{\alpha\beta} u_{\gamma} u_{\delta} u_{\epsilon} + \delta_{\alpha\gamma} u_{\beta} u_{\delta} u_{\epsilon} + \delta_{\alpha\delta} u_{\beta} u_{\gamma} u_{\epsilon} + \delta_{\alpha\epsilon} u_{\beta} u_{\gamma} u_{\delta} + \delta_{\beta\epsilon} u_{\alpha} u_{\gamma} u_{\delta} + \delta_{\beta\delta} u_{\alpha} u_{\gamma} u_{\epsilon} + \delta_{\gamma\delta} u_{\alpha} u_{\beta} u_{\epsilon} + \delta_{\gamma\epsilon} u_{\alpha} u_{\beta} u_{\delta} + \delta_{\delta\epsilon} u_{\alpha} u_{\gamma} u_{\beta}.$$. \quad (33)

Since the weights have to be positive, $w_i > 0$, $c^2_s$ is limited to a certain interval, i.e., $c^2_{s, min} < c^2_s < c^2_{s, max}$. Graphical analysis of the $w_i$ reveals (in lattice units) that $c^2_{s, min}$ takes the values 0.3510760 and 0.3850612 for the two- and three-dimensional models, respectively, while the corresponding values for $c^2_{s, max}$ are 1.3333333 and 1.1917145. This means that one is limited in the construction of the equation of state, too (cf. Eq. 18). More details on this are found in App. B.

### IV. DISCRETIZATION OF DERIVATIVES

#### A. Lattice sets and Taylor expansion

The interface collision operator and the correction collision operator will turn out to depend on different orders of derivatives of hydrodynamic variables like density $\rho$ or velocity $u$. For the CE expansion and the implementation of the algorithm we hence need to know how different orders of derivatives are systematically discretized. This is explained within this section.
We start from an arbitrary but symmetric set of dimensionless lattice vectors $\mathbf{d}_i$, with dimensionless coefficients $\tau_i$. Again we organize the vectors in shells and require that the $\tau_i$ are the same within each shell. In contrast to the weights $w_i$, there is no restriction on the sign of the $\tau_i$, and there is in general no normalization condition either. The odd moments of $\tau_i$ vanish for symmetry reasons, while the even moments are defined similarly as in Eqs. 21–23 up to fourth order:

\begin{align}
\sum_i \tau_i &= \bar{\sigma}_0, \quad \text{(34)} \\
\sum_i \tau_i d_{i\alpha} d_{i\beta} &= \bar{\sigma}_2 \delta_{\alpha\beta}, \quad \text{(35)} \\
\sum_i \tau_i d_{i\alpha} d_{i\beta} d_{i\gamma} d_{i\delta} &= \bar{\sigma}_4 (\delta_{\alpha\beta}\delta_{\gamma\delta} + \text{perm.}); \quad \text{(36)}
\end{align}

again we require that the fourth moment is isotropic whenever it occurs.

We assume that $g = g(\mathbf{r}_1)$ is some hydrodynamic variable like mass density $\rho$, momentum density $\mathbf{j}$, or flow velocity $\mathbf{u} = \mathbf{j}/\rho$. Within the framework of the CE expansion terms of the form

$$g_i \equiv g(\mathbf{r}_1 + \varepsilon \mathbf{a} \mathbf{d}_i)$$

will appear, and they should be expanded (at least up to third order) with respect to $\varepsilon$:

$$g_i = g + \varepsilon a d_{i\alpha} \partial_{\alpha} g + \frac{1}{2} \varepsilon^2 a^2 d_{i\alpha} d_{i\beta} \partial_{\alpha} \partial_{\beta} g \quad \text{(38)}$$

$$+ \frac{1}{6} \varepsilon^3 a^3 d_{i\alpha} d_{i\beta} d_{i\gamma} \partial_{\alpha} \partial_{\beta} \partial_{\gamma} g + O(\varepsilon^4).$$

We will now discuss discretizations case by case. For each case, we will derive a set of conditions that a specific set of vectors $\mathbf{d}_i$ and its set of coefficients $\tau_i$ has to fulfill in order to correctly calculate the respective derivative up to third order. Different derivatives are constructed by taking different tensorial moments of Eq. 38 and by using Eqs. 34–36.

### B. First derivative $\partial_\alpha g$

This is a vector, and therefore it is sufficient to study a vectorial moment:

$$\sum_i \tau_i d_{i\alpha} g_i = \varepsilon a \bar{\sigma}_2 \partial_{\alpha} g$$

$$+ \frac{1}{2} \varepsilon^2 a^2 \bar{\sigma}_4 \partial_{\alpha} \partial_{\beta} \partial_{\gamma} g + O(\varepsilon^4).$$

We now require

$$\bar{\sigma}_2 = 1,$$

$$\bar{\sigma}_4 = 0,$$

resulting in

$$\frac{1}{a} \sum_i \tau_i d_{i\alpha} g_i = \varepsilon \partial_{\alpha} g + O(\varepsilon^4).$$

### C. Second derivative $\partial_\alpha \partial_\beta g$

This is a scalar, and hence one should study a simple scalar moment:

$$\sum_i \tau_i g_i = \bar{\sigma}_0 g + \frac{1}{2} \varepsilon^2 a^2 \bar{\sigma}_2 \partial_{\alpha} \partial_{\alpha} g + O(\varepsilon^4),$$

We now require

$$\bar{\sigma}_0 = 0,$$

$$\bar{\sigma}_2 = 2,$$

resulting in

$$\frac{1}{a^2} \sum_i \tau_i g_i = \partial_\alpha \partial_\beta g + O(\varepsilon^4).$$

### D. Second derivative $\partial_\alpha \partial_\beta g$

This is a symmetric second-rank tensor, and its trace has already been discussed in the previous subsection. We therefore confine attention to its traceless part,

$$\bar{\partial}_\alpha \partial_\beta g = \left( \partial_\alpha \partial_\beta - \frac{1}{a} \delta_{\alpha\beta} \partial_{\gamma} \partial_{\gamma} \right) g,$$
where \( d \) is the spatial dimension, and, correspondingly, study the traceless moment
\[
\sum d_{ia}d_{ib}g_i = \tilde{\sigma}_4 a^2 d_{ia}d_{ib}g + O(\varepsilon^4),
\]
where the traceless part of the tensor \( d_{ia}d_{ib} \) is given by
\[
\overline{d_{ia}d_{ib}} = d_{ia}d_{ib} - \frac{1}{d} d_{ia}d_{ib}d_{ia}.
\]
Requiring \( \tilde{\sigma}_4 = 1 \) then results in
\[
\frac{1}{a^2} \sum \tau d_{ia}d_{ib}g_i = \partial_a\partial_b\partial_cg + O(\varepsilon^4),
\]
i.e., a valid approximation for the desired derivative. The corresponding coefficients are again given in Tab. III. For the non-traceless tensor \( \partial_a\partial_b\partial_cg \) one simply has to superimpose the present result with the approximation derived in the previous subsection.

E. Third derivative \( \partial_a\partial_b\partial_cg \)

This is again a vector, so a first moment is sufficient. We find
\[
\sum \tau d_{ia}g_i = a\tilde{\sigma}_2\partial_a g + \frac{1}{2} a^2 \tilde{\sigma}_4 \partial_a\partial_b\partial_cg + O(\varepsilon^4).
\]
We now require
\[
\tilde{\sigma}_2 = 0, \quad \tilde{\sigma}_4 = 2,
\]
resulting in
\[
\frac{1}{a^2} \sum \tau d_{ia}g_i = \partial_a\partial_b\partial_cg + O(\varepsilon^4),
\]
i.e., just the desired derivative. The coefficients are again given in Tab. III.

V. MOMENTUM TRANSFER

We start from the condition that the total momentum density is changed as a result of the acting force:
\[
h f_{\alpha} = \sum \Delta_i c_{ia} = \sum \Delta_i^{\text{bulk}} c_{ia} + \sum \Delta_i^{\text{int}} c_{ia} + \sum \Delta_i^{\text{corr}} c_{ia} = \sum (\gamma - 1)(n_{i\alpha} - n_{i\alpha}^0) c_{ia} + \sum \Delta_i^{\text{int}} c_{ia} + \sum \Delta_i^{\text{corr}} c_{ia}.
\]
Abbreviating
\[
\sum n_{i\alpha} c_{ia} = : j_{0\alpha},
\]
\[
\Delta_i^{\text{int}} + \Delta_i^{\text{corr}} = : \Delta_i',
\]
we thus find
\[
h f_{\alpha} = (\gamma - 1)(j_{0\alpha} - j_{\alpha}) + \sum \Delta_i' c_{ia}. \tag{59}
\]
On the other hand, the momentum density \( j_{\alpha} \) is defined via the prescription
\[
\sum \Delta_i^{\text{corr}} c_{ia} = \frac{1}{2} \sum (n_{i\alpha} + n_{i\alpha}^0) c_{ia} + \tilde{j}_{\alpha}, \tag{60}
\]
where the first term involves the arithmetic mean of the pre- \((n_{i\alpha})\) and post-collisional \((n_{i\alpha}^0 = n_{i\alpha} + \Delta_i)\) populations, while the second is the correction current. We can therefore write
\[
\sum \Delta_i^{\text{int}} c_{ia} = \frac{1}{2} h f_{\alpha} + (\gamma - 1)\tilde{j}_{\alpha}. \tag{62}
\]
Combining Eqs. 59 and 61, we can eliminate \( j_{0\alpha} - j_{\alpha} \) to find
\[
\sum \Delta_i^{\text{int}} c_{ia} = \frac{1 + \gamma}{2} h f_{\alpha} + (\gamma - 1)\tilde{j}_{\alpha}.
\]
Since \( \Delta_i' = \Delta_i^{\text{int}} + \Delta_i^{\text{corr}} \), it is most natural to require that
\[
\sum \Delta_i^{\text{int}} c_{ia} = \frac{1 + \gamma}{2} h f_{\alpha}, \tag{63}
\]
\[
\sum \Delta_i^{\text{corr}} c_{ia} = (\gamma - 1)\tilde{j}_{\alpha}; \tag{64}
\]
note that \( \Delta_i^{\text{int}} \) should be a result of the interface force, while both \( \Delta_i^{\text{corr}} \) and \( \tilde{j}_{\alpha} \) are correction terms derived within the framework of the CE analysis. Actually, the two terms correspond to different CE orders — \( f_{\alpha} \) is of third order, while \( \tilde{j}_{\alpha} \) is of second order.

For the momentum transfer of the BGK part we thus obtain
\[
\sum \Delta_i^{\text{bulk}} c_{ia} = \frac{1 - \gamma}{2} h f_{\alpha} + (1 - \gamma)\tilde{j}_{\alpha}. \tag{65}
\]

VI. INTERFACE FORCE COLLISION OPERATOR

On the continuum level, the interface force density is given by (cf. Eq. 7):
\[
f_{\alpha} = \kappa \rho \partial_a \partial_b \partial_c \rho.
\]
According to the results of Sec. IV E, \( f_{\alpha} \) can hence be approximated on the lattice as
\[
f_{\alpha} = \frac{\kappa \rho}{\alpha^2} \sum \tau d_{ia} \rho_i. \tag{66}
\]
Furthermore, from Sec. V we know that the interface collision operator should be mass-conserving, and have a first velocity moment
\[ \sum_i \Delta_i^{int} c_{i\alpha} = \frac{1 + \gamma}{2} h f_\alpha. \] (68)

It is easy to show that these conditions are met by the operator
\[ \Delta_i^{int} = \frac{1 + \gamma}{2} h w_i(\rho_0) c_{i\alpha} f_\alpha; \] (69)

here \( \rho_0 \) is some arbitrarily chosen reference density. The second velocity moment of this operator is evidently zero, while its third moment is easily evaluated as
\[ \sum_i \Delta_i^{int} c_{i\alpha} c_{i\beta} c_{i\gamma} = \frac{1 + \gamma}{2} h c_2(\rho_0) (f_{\alpha\beta\gamma} + f_{\beta\alpha\gamma} + f_{\gamma\alpha\beta}). \] (70)

Since the evaluation of \( f_\alpha \) involves a third-order derivative, \( f_\alpha \) is of third order in the CE expansion, and this is true as well for \( \Delta_i^{int} \) and all of its moments.

Finally, we note that the procedure does not conserve the momentum on the single lattice site; nevertheless, the global momentum is strictly conserved. This is so because of the relation
\[ \sum_r f(r) = \frac{K}{a^3} \sum_i \gamma_i \sum_r \rho(r) d_i \rho(r + ad_i) = 0. \] (71)

The vanishing of the total force is due to the fact that in the inner sum each pair of densities occurs twice, with weighting vectors \( +d \) and \( -d \), respectively. These terms therefore exactly cancel. A prerequisite is however that the system is translationally invariant, which is the case for periodic boundary conditions.

**VII. CHAPMAN-ENSKOG ANALYSIS**

**A. CE hierarchy**

After these preliminary considerations, we are prepared for the CE analysis of the algorithm, which will allow us to derive \( \Delta_i^{corr} \). We start from the LB equation (cf. Eq. 15)
\[ n_i(r_1 + \varepsilon c_i h, t_1 + \varepsilon h, t_2 + \varepsilon^2 h, t_3 + \varepsilon^3 h) - n_i(r_1, t_1, t_2, t_3) = \Delta_i. \] (72)

Introducing the differential operator
\[ D_i = \varepsilon h c_{i\alpha} \partial_{\alpha} + \varepsilon h \partial_t + \varepsilon^2 h \partial_t + \varepsilon^3 h \partial_t, \] (73)

the LB equation can be re-written exactly as
\[ (\exp(D_i) - 1) n_i = \Delta_i, \] (74)
or
\[ D_i n_i = D_i (\exp(D_i) - 1)^{-1} \Delta_i, \] (75)

and the right hand side can be expanded as a series involving the Bernoulli numbers:
\[ D_i n_i = \left[ 1 - \frac{D_i}{2} + \frac{D_i^2}{12} + \ldots \right] \Delta_i. \] (76)

Furthermore, \( n_i \) and \( \Delta_i \) are also expanded in terms of the parameter \( \varepsilon \) up to third order:
\[ n_i = n_i^{(0)} + \varepsilon n_i^{(1)} + \varepsilon^2 n_i^{(2)} + \varepsilon^3 n_i^{(3)} + \ldots, \] (77)
\[ \Delta_i = \Delta_i^{(0)} + \varepsilon \Delta_i^{(1)} + \varepsilon^2 \Delta_i^{(2)} + \varepsilon^3 \Delta_i^{(3)} + \ldots. \] (78)

Inserting Eqs. 77, 78 and 73 into Eq. 76 we get a systematic expansion in \( \varepsilon \), which has to be satisfied at each order separately:

- \( \varepsilon^0: \)
  \[ \Delta_i^{(0)} = 0. \] (79)

- \( \varepsilon^1: \)
  \[ (c_{i\alpha} \partial_{\alpha} + \partial_t) n_i^{(0)} = \frac{1}{h} \Delta_i^{(1)}. \] (80)

- \( \varepsilon^2: \)
  \[ (c_{i\alpha} \partial_{\alpha} + \partial_t) n_i^{(1)} + \partial_t n_i^{(0)} = \frac{1}{h} \Delta_i^{(2)} - \frac{1}{2} (c_{i\alpha} \partial_{\alpha} + \partial_t) \Delta_i^{(1)}. \] (81)

- \( \varepsilon^3: \)
  \[ (c_{i\alpha} \partial_{\alpha} + \partial_t) n_i^{(2)} + \partial_t n_i^{(1)} + \partial_t n_i^{(0)} = \frac{1}{h} \Delta_i^{(3)} - \frac{1}{2} (c_{i\alpha} \partial_{\alpha} + \partial_t) \Delta_i^{(2)} \]
  \[ - \frac{1}{2} \partial_t \Delta_i^{(1)} + \frac{1}{12} h (c_{i\alpha} \partial_{\alpha} + \partial_t)^2 \Delta_i^{(1)}. \] (82)

Since both \( \Delta_i^{int} \) and \( \Delta_i^{corr} \) are of higher order, the zeroth-order equation is only of importance for the BGK operator. This however means that we can identify the equilibrium populations with the zeroth order: \( n_i^{(0)} \equiv n_i^{eq} \), and we can use the notations “(0)” and “\( ecq \)” interchangeably, both for the populations and their moments. The next step will involve taking velocity moments of the CE hierarchy.

**B. Velocity moments**

We hence define:
• zeroth moment: mass density
\[ \rho = \sum_i n_i. \]  
(83)

• first moment: momentum density
\[ j_\alpha = \sum_i n_i c_{i\alpha} + \frac{h}{2} f_\alpha + j_\alpha. \]  
(84)

• second moment: stress
\[ \pi_{\alpha\beta} = \sum_i n_i c_{i\alpha} c_{i\beta}. \]  
(85)

• third moment:
\[ \phi_{\alpha\beta\gamma} = \sum_i n_i c_{i\alpha} c_{i\beta} c_{i\gamma}. \]  
(86)

• fourth moment:
\[ \psi_{\alpha\beta\gamma\delta} = \sum_i n_i c_{i\alpha} c_{i\beta} c_{i\gamma} c_{i\delta}. \]  
(87)

By replacing \( n_i \) with \( n_i^{(k)} \), we obtain the corresponding moments at \( k \)th order of the CE expansion, such that, e. g., \( \pi_{\alpha\beta}^{(1)} \) denotes the first-order stress, i. e. the second velocity moment of \( n_i^{(1)} \). For \( k = 0 \) (equilibrium populations) these moments have already been derived at the end of Sec. III. For \( \rho \) and \( j \) it should be noted that these are the hydrodynamic variables for which there are no higher-order contributions, i. e. \( \rho^{(1)} = \rho^{(2)} = \ldots = 0 \), \( j^{(1)} = j^{(2)} = \ldots = 0 \).

The momentum density in Eq. 84 is defined as a mean value between pre- and post-collisional momentum density plus an additional term: \( j = (1/2) \sum_i (n_i + n_i^\neq) c_i + \bar{j} \), see also Sec. V. The additional term \( \bar{j} \) is needed to guarantee the continuity equation to be consistent up to the third order. Because of this an implicit algorithm for the calculation of the fluid velocity \( u = j/\rho \) is required. This will be discussed in more detail later.

We can re-write Eq. 84 as
\[ \sum_i n_i c_{i\alpha} = j_\alpha - \frac{h}{2} f_\alpha - j_\alpha \]  
(88)

and specify for each order (note that \( f \) is of third order, \( f = \varepsilon^3 f^{(3)} + O(\varepsilon^4) \), and \( \bar{j} \) cannot have a zeroth-order contribution):
\[ \sum_i n_i^{(0)} c_{i\alpha} = j_\alpha, \]  
(89)
\[ \sum_i n_i^{(1)} c_{i\alpha} = -\bar{j}_\alpha^{(1)}, \]  
(90)
\[ \sum_i n_i^{(2)} c_{i\alpha} = -\bar{j}_\alpha^{(2)}, \]  
(91)
\[ \sum_i n_i^{(3)} c_{i\alpha} = -\frac{h}{2} f_\alpha^{(3)} - \bar{j}_\alpha^{(3)}. \]  
(92)

We will find later that \( \bar{j}_\alpha^{(1)} = \bar{j}_\alpha^{(3)} = 0 \), i. e. that \( \bar{j} \) is a pure second-order contribution.

The analogous relation for the zeroth moment is
\[ \sum_i n_i^{(0)} = \rho, \]  
(93)
\[ \sum_i n_i^{(k)} = 0 \text{ for } k \geq 1. \]  
(94)

Similarly, we need to discuss moments of the collision operator. For the stress we make use of the fact that \( \Delta_i = n_i^\neq - n_i^\eq \), and hence we can write
\[ \sum_i \Delta_i^{(k)} c_{i\alpha} c_{i\beta} = \pi_{\alpha\beta}^{(k)} - \pi_{\alpha\beta}, \]  
(95)
and we will make use of similar expressions for the higher-order moments as well. For the zeroth moment we note the mass conservation condition \( \sum_i \Delta_i = 0 \), and hence
\[ \sum_i \Delta_i^{(k)} = 0 \text{ for all } k. \]  
(96)

For the first moment, we know the momentum-transfer condition \( \sum_i \Delta_i c_i = h f \), and that the rhs is of third order. Hence
\[ \sum_i \Delta_i^{(k)} c_i = 0 \text{ for } k = 0, 1, 2, \]  
(97)
\[ \sum_i \Delta_i^{(3)} c_i = h f^{(3)}. \]  
(98)

C. Pre- and post-collisional moments

For the later development, it will be useful to know some relations between pre- and post-collisional moments. The collision operator is
\[ \Delta_i = (\gamma - 1)(n_i - n_i^\eq) + \Delta_i^{\text{int}} + \Delta_i^{\text{corr}}. \]  
(99)

Introducing the notation \( n_i^\neq = n_i - n_i^\eq \) for the non-equilibrium populations, the update rule is
\[ n_i^{\neq} = \gamma n_i^{\neq} + \Delta_i^{\text{int}} + \Delta_i^{\text{corr}}. \]  
(100)

In view of later results, it will be useful to re-write this as
\[ \frac{1}{\gamma} \left( n_i^{\neq} + n_i^\eq \right) = \frac{1}{\gamma} \left( n_i^{\neq} - n_i^\eq \right) - \frac{1}{1 - \gamma} \left( \Delta_i^{\text{int}} + \Delta_i^{\text{corr}} \right). \]  
(101)

We now consider the second and third velocity moment of this relation, at first and second order of the CE expansion. Since \( \Delta_i^{\text{int}} \) is of third order, it does not contribute. For the moments of \( \Delta_i^{\text{corr}} \) we introduce the abbreviations
\[ \sum_i \Delta_i^{\text{corr}} c_{i\alpha} c_{i\beta} =: \Sigma_{\alpha\beta}, \]  
(102)
\[ \sum_i \Delta_i^{\text{corr}} c_{i\alpha} c_{i\beta} c_{i\gamma} =: \Xi_{\alpha\beta\gamma}, \]  
(103)
and hence we have
\[
\frac{1}{2} \left( \pi_{\alpha\beta}^{*(1,2)} + \pi_{\alpha\beta}^{(1,2)} \right) = \frac{1}{2} \gamma - 1 \left( \pi_{\alpha\beta}^{*(1,2)} - \pi_{\alpha\beta}^{(1,2)} \right) + \frac{1}{2} \gamma \pi_{\alpha\beta}^{(1,2)},
\]
\[
\frac{1}{2} \left( \phi_{\alpha\beta\gamma}^{*(1,2)} + \phi_{\alpha\beta\gamma}^{(1,2)} \right) = \frac{1}{2} \gamma - 1 \left( \phi_{\alpha\beta\gamma}^{*(1,2)} - \phi_{\alpha\beta\gamma}^{(1,2)} \right) + \frac{1}{2} \gamma \pi_{\alpha\beta\gamma}^{(1,2)},
\]
where \( (1, 2) \) means that either first or second order can be expressed this way.

D. Mass conservation equation

By taking the zeroth velocity moment of the CE hierarchy, we find

- at order \( \varepsilon^1 \):
  \[
  \partial_1 \rho + \partial_1 j_\alpha = 0.
  \]

- at order \( \varepsilon^2 \):
  \[
  \partial_2 \rho - \partial_1 \tilde{j}_\alpha^{(1)} = 0.
  \]

- at order \( \varepsilon^3 \):
  \[
  \partial_3 \rho - \partial_1 \tilde{j}_\alpha^{(2)} = \frac{h}{12} \partial_1 \partial_1 \left( \pi_{\alpha\beta}^{*(1)} - \pi_{\alpha\beta}^{(1)} \right).
  \]

At this point, it becomes clear why the correction current \( \tilde{j} \) is needed — its purpose is to compensate the rhs of Eq. 108. We therefore require
\[
\tilde{j}_\alpha^{(1)} = 0 \quad \text{and} \quad \tilde{j}_\alpha^{(2)} = -\frac{h}{12} \partial_1 \left( \pi_{\alpha\beta}^{*(1)} - \pi_{\alpha\beta}^{(1)} \right). \tag{109}
\]

We then multiply each equation of motion with its corresponding power of \( \varepsilon \) and add up the resulting equations. This gives rise to the continuity equation
\[
\partial_t \rho + \partial_\alpha j_\alpha = 0, \tag{110}
\]
which is therefore, by construction, accurate up to third order. We will later see that \( \tilde{j}_\alpha^{(3)} \) does not appear in the equations, and therefore we may assume that this order vanishes. We will discuss later how to actually determine \( \tilde{j} \).

E. Momentum conservation equation

Taking the first velocity moment of the CE hierarchy, we obtain

- at order \( \varepsilon^1 \):
  \[
  \partial_1 j_\alpha + \partial_\beta \pi_{\alpha\beta}^{(0)} = 0.
  \]

F. Dynamics of higher-order moments, and closure

By taking moments of the CE hierarchy, we derive the following equations of motion:
where $d$ is the spatial dimension. It should be noted that the condition $\eta_\nu > 0$ places a further restriction on the admissible equation of state:

$$\frac{\partial p}{\partial \rho} < \left(1 + \frac{2}{d}\right) \frac{p}{\rho}.$$  

(124)

This result may also be used directly to determine the correction current $\tilde{j}$. From Eq. 109 and its discussion we find

$$\tilde{j}_\alpha = -\varepsilon^2 \frac{h^2}{12} \frac{\partial \rho}{\partial \rho} \left(\frac{\pi^{(1)}_{\alpha\beta} - \pi^{(1)}_{\alpha\beta}}{\eta_\nu} - \delta_{\alpha\beta}\right),$$  

(125)

$$= -\frac{h^2}{12} \frac{\partial \rho}{\partial \rho} \left[p - \frac{\partial p}{\partial \rho}\right] \delta_{\alpha\beta} \partial_\gamma u_\gamma + p \left(\partial_\alpha u_\alpha + \partial_\beta u_\beta\right).$$  

We now use Eq. 119 to eliminate the second-order stress. Inserting this into Eq. 115, we find

$$\partial_t J_\alpha + \partial_\beta \pi^{(0)}_{\alpha\beta} + \frac{\varepsilon}{\gamma - 1} \partial_\beta \left(\pi^{(1)}_{\alpha\beta} - \pi^{(1)}_{\alpha\beta}\right) = f_\alpha$$  

(126)

$$+ \frac{\varepsilon}{\gamma - 1} \partial_\beta \Sigma^{(1)}_{\alpha\beta} + \frac{\varepsilon^2 h}{(\gamma - 1)^2} \frac{\partial_\beta \partial_\gamma \Sigma^{(1)}_{\alpha\beta}}{\partial_\beta \partial_\gamma}$$  

$$+ \frac{\varepsilon^2}{\gamma - 1} \partial_\beta \Sigma^{(2)}_{\alpha\beta} + \frac{\varepsilon h}{\gamma - 1} \partial_\beta \partial_\gamma \Sigma^{(1)}_{\alpha\beta}$$  

$$- \frac{\varepsilon^2 h}{\gamma - 1} \frac{\partial_\beta \partial_\gamma \pi^{(0)}_{\alpha\beta}}{\partial_\beta \partial_\gamma}$$  

$$- \frac{\varepsilon^2 h}{\gamma - 1} \frac{\partial_\beta \partial_\gamma \pi^{(1)}_{\alpha\beta}}{\partial_\beta \partial_\gamma}$$  

$$+ \frac{\varepsilon h}{\gamma - 1} \partial_\beta \partial_\gamma \left(\pi^{(1)}_{\alpha\beta} - \pi^{(1)}_{\alpha\beta}\right)$$  

It is now easy to see that all the spurious terms can be eliminated by proper construction of the second and third moments of the correction collision operator (see Eqs. 102 and 103), which have been up to now completely arbitrary. By matching powers of $\varepsilon$ and tensor ranks, one finds that the following conditions have to be fulfilled:

$$\Sigma^{(1)}_{\alpha\beta} = 0,$$  

(127)

$$\Xi^{(1)}_{\alpha\beta} = \frac{\gamma^2 + 4\gamma + 1}{3(\gamma + 1)} \left(\phi^{(1)}_{\alpha\beta} - \phi^{(1)}_{\alpha\beta}\right),$$  

(128)

$$\Sigma^{(2)}_{\alpha\beta} = h \frac{\gamma + 1}{2} \partial_\beta \pi^{(0)}_{\alpha\beta}$$  

$$+ h \frac{\gamma^2 + 4\gamma + 1}{6(\gamma - 1)} \partial_\beta \left(\pi^{(1)}_{\alpha\beta} - \pi^{(1)}_{\alpha\beta}\right).$$  

(129)
We proceed by further closing the equations, i.e. by replacing the rhs sides of Eqs. 128 and 129 with the corresponding spatial derivatives of the hydrodynamic variables. This is done in App. C, see Eqs. C11, C16 and C20 (we do not repeat the lengthy expressions derived there). The thus-derived derivatives then need to be discretized in a consistent fashion, i.e. correctly up to third order in $\varepsilon$. How to do this has already been discussed in Sec. IV.

VIII. CORRECTION COLLISION OPERATOR

The correction collision operator $\Delta_i^{\text{corr}}$ is still unknown. However, we now know all the conditions that it has to satisfy. Let us collect them here again:

- Mass conservation:
  \[ \sum_i \Delta_i^{\text{corr}} = 0. \]  
  (130)

- Consistent momentum transfer:
  \[ \sum_i \Delta_i^{\text{corr}} c_{i\alpha} = (\gamma - 1) \dot{j}_\alpha \]  
  (131)

(see Eq. 64), where $\dot{j}$ is given by Eq. 125.

- Second moment:
  \[ \sum_i \Delta_i^{\text{corr}} c_{i\alpha} c_{i\beta} = \Sigma_{\alpha\beta} = \varepsilon^2 \Sigma^{(2)}_{\alpha\beta}, \]  
  (132)

where the rhs is given by Eqs. 129, C16 and C20.

- Third moment:
  \[ \sum_i \Delta_i^{\text{corr}} c_{i\alpha} c_{i\beta} c_{i\gamma} = \Xi_{\alpha\beta\gamma} = \varepsilon \Xi^{(1)}_{\alpha\beta\gamma}, \]  
  (133)

where the rhs is given by Eqs. 128 and C11.

A collision operator that yields these desired moments is

\[ \Delta_i^{\text{corr}} = (\gamma - 1) \frac{w_i}{c_s^2} \dot{j}_\alpha c_{i\alpha} \]  
(134)

\[ + \frac{w_i}{2 c_s^2} \Sigma_{\alpha\beta} (c_{i\alpha} c_{i\beta} - c_s^2 \delta_{\alpha\beta}) \]

\[ + \frac{w_i}{6 c_s^6} \Xi_{\alpha\beta\gamma} (c_{i\alpha} c_{i\beta} c_{i\gamma} - c_s^2 \delta_{\alpha\beta} c_{i\gamma}) \]

\[ - c_s^2 \delta_{\alpha\gamma} c_{i\beta} - c_s^2 \delta_{\beta\gamma} c_{i\alpha} \]

with

\[ \Xi^{(1)}_{\alpha\beta\gamma} = \Xi_{\alpha\beta\gamma} - (\gamma - 1) c_s^2 \left( \delta_{\alpha\beta} \dot{\gamma} + \delta_{\alpha\gamma} \dot{j}_\beta + \delta_{\beta\gamma} j_{\alpha} \right). \]  
(135)

As in the case of the interface force operator, the weights $w_i$ and the pressure $\rho_0 c_s^2$ need to be evaluated at one fixed reference density $\rho_0$, in order to avoid the occurrence of additional unwanted gradients.

IX. IMPLICIT ALGORITHM

The correction current $\dot{j}$ is given by Eq. 125. However, the rhs of this equation depends on the streaming velocity $\dot{u} = \dot{j}/\rho$, plus its gradients, while $\dot{u}$ in turn depends on $\dot{j}$, see Eq. 20. This means that $\dot{j}$ and $\dot{u}$ are defined implicitly. These observations suggest the following iterative procedure to calculate the correction current:

1. On each lattice site, determine the density $\rho$.
2. On each lattice site, determine the force density $f$, using the outlined finite-difference procedure.
3. On each lattice site, initialize $\dot{j}$ by setting it to zero, or by taking the value from the previous time step.
4. On each lattice site, calculate $\dot{j}$ and $\dot{u}$ from Eq. 20.
5. On each lattice site, calculate $\dot{j}$ from Eq. 125, again using a finite-difference procedure.
6. Go to step 4, unless the iteration has converged.

It should be noted that this problem is essentially a linear system of equations, and hence it should in principle be amenable to more sophisticated iterative solvers as well.

As soon as the iteration has converged, the values of all hydrodynamic variables are available on all lattice sites. One may then proceed to evaluate the equilibrium populations and the three contributions to the collision operator. The collision is followed by a streaming step, after which the procedure starts again.

X. CONCLUSIONS

The present paper has dealt with the attempt to construct an isothermal LB algorithm for gas-liquid coexistence, with the goal to obtain a procedure that is (in the limit of sufficiently slow flows) fully consistent with both hydrodynamics and thermodynamics. Motivated by the success of LB methods for the ideal gas, we constructed the method in close analogy to what is known from there. Central to our approach is the observation that bulk and interfacial free energies should enter the analysis at very different orders of the CE expansion: The bulk free energy (or the bulk pressure) should be encoded in the zeroth order, or the equilibrium populations, while the interfacial force density, involving a third-order gradient, should enter at third order. The present paper therefore directly builds upon this observation, and constructs an algorithm that is systematically shown to be consistent up to and including the third order, since this is a necessary condition for consistency of the method as a whole. Up to now, to the best of our knowledge, has neither a CE analysis of multiphase LB ever been done up to third order, nor has any LB algorithm for a gas-liquid system been constructed that would satisfy that consistency criterion. It is therefore hardly surprising that so
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APPENDIX A: INTERFACE FORCE FROM CAHN-HILLIARD FREE ENERGY

1. Dissipation-free bulk hydrodynamics

The continuity equation reads as

\[ \frac{\partial}{\partial t} \rho + \partial_{\alpha} (\rho u_{\alpha}) = 0. \]  

(A1)

Introducing the convective derivative

\[ \frac{D}{Dt} = \frac{\partial}{\partial t} + u_{\alpha} \partial_{\alpha}, \]  

(A2)

this is rewritten as

\[ \frac{D}{Dt} \rho + \rho \partial_{\alpha} u_{\alpha} = 0. \]  

(A3)

From this, one can easily show the identity

\[ \rho \frac{D}{Dt} \left( \frac{\phi}{\rho} \right) = \frac{\partial \phi}{\partial t} + \partial_{\alpha} (u_{\alpha} \phi) \]  

(A4)

for an arbitrary function \( \phi \). Introducing the momentum density \( j = \rho u \), the Euler equation in the presence of a force density \( f \) is written as

\[ \frac{\partial}{\partial t} j_{\alpha} + \partial_{\beta} (u_{\beta} j_{\alpha}) + \partial_{\alpha} p = f_{\alpha}. \]  

(A5)

Using Eq. A4, this is rewritten as

\[ \rho \frac{D}{Dt} u_{\alpha} + \partial_{\alpha} p = f_{\alpha}. \]  

(A6)

Therefore, we find for the kinetic energy, again using Eq. A4

\[ \frac{\partial}{\partial t} \left( \frac{1}{2} \rho u_{\alpha} u_{\alpha} \right) + \partial_{\beta} \left( \frac{1}{2} \rho u_{\beta} u_{\alpha} u_{\alpha} \right) \]  

\[ = \rho \frac{D}{Dt} \left( \frac{1}{2} u_{\alpha} u_{\alpha} \right) = \rho u_{\alpha} \frac{D}{Dt} u_{\alpha} = u_{\alpha} (f_{\alpha} - \partial_{\alpha} p). \]  

(A7)

Let \( e \) and \( s \) denote the internal energy and entropy per unit mass, respectively, such that internal energy and entropy density are given by \( pe \) and \( ps \), respectively. If \( E \) is the internal energy and \( S \) the entropy, then the first law of thermodynamics for fixed particle number \( N \) or fixed total mass \( M \) reads

\[ dE = TdS - pdV; \]  

(A8)

here \( T \) is the temperature and \( V \) the volume. Dividing this equation by the total mass \( M \), we obtain

\[ \frac{de}{dt} = Tds - pd \left( \frac{1}{\rho} \right) = Tds + \frac{p}{\rho^2} dp. \]  

(A9)

Since we are studying dissipation-free hydrodynamics, there is no entropy production, and the equation of motion for the entropy is simply

\[ \frac{D}{Dt} s = 0. \]  

(A10)
We therefore find
\[ \rho \frac{D}{Dt} \mathbf{u} = \frac{p}{\rho} \frac{D}{Dt} \rho = -p \partial_\alpha u_\alpha. \]  
(A11)

Again using Eq. A4, we can rewrite this as
\[ \frac{\partial}{\partial t} (\rho \mathbf{u}) + \partial_\alpha (u_\alpha \rho) = -p \partial_\alpha u_\alpha. \]  
(A12)

The equation of motion for the total energy density therefore results to
\[ \frac{\partial}{\partial t} \left( \frac{1}{2} \rho u^2 + \rho e \right) + \partial_\alpha \left\{ u_\alpha \left( \frac{1}{2} \rho u^2 + \rho e \right) \right\} = u_\alpha (f_\alpha - \partial_\alpha p) - p \partial_\alpha u_\alpha \]  
(A13)
or
\[ \frac{\partial}{\partial t} \left( \frac{1}{2} \rho u^2 + \rho e \right) + \partial_\alpha \left\{ u_\alpha \left( \frac{1}{2} \rho u^2 + \rho e + p \right) \right\} = u_\alpha f_\alpha. \]  
(A14)

For \( f = 0 \), this is the conservation of energy. For a system with periodic boundary conditions, we define the Hamiltonian in \( d \)-dimensional space as
\[ \mathcal{H} = \int d^d \mathbf{r} \left( \frac{1}{2} \rho \mathbf{u}^2 + \rho e \right), \]  
(A15)

and Gauss’ theorem implies that it is only changed as a result of the force density \( f \):
\[ \frac{d}{dt} \mathcal{H} = \int d^d \mathbf{r} u_\alpha f_\alpha. \]  
(A16)

2. Inclusion of interfacial energies

We now modify the Hamiltonian to also include an interfacial term, i.e.
\[ \mathcal{H} = \int d^d \mathbf{r} \left[ \frac{1}{2} \rho \mathbf{u}^2 + \rho e + \frac{\kappa}{2} (\nabla \rho)^2 \right], \]  
(A17)

with \( \kappa > 0 \). Furthermore, we require that the Hamiltonian is conserved under the non-dissipative dynamics of the previous section, i.e. that the force term \( f \) is chosen in such a way that
\[ \frac{d}{dt} \mathcal{H} = 0. \]  
(A18)

This however means
\[ \int d^d \mathbf{r} u_\alpha f_\alpha + \frac{\kappa}{2} \int d^d \mathbf{r} \frac{\partial}{\partial t} (\nabla \rho)^2 = 0. \]  
(A19)

Now, from the continuity equation we find
\[ \frac{1}{2} \frac{\partial}{\partial t} (\nabla \rho)^2 = (\partial_\beta \rho) \partial_\beta \frac{\partial}{\partial t} \rho \]
\[ = - (\partial_\beta \rho) \partial_\beta \partial_\alpha (u_\alpha \rho), \]  
(A20)

resulting in
\[ \int d^d \mathbf{r} u_\alpha f_\alpha = \kappa \int d^d \mathbf{r} (\partial_\beta \rho) \partial_\beta \partial_\alpha (u_\alpha \rho) \]
\[ = \kappa \int d^d \mathbf{r} u_\alpha \rho \partial_\alpha \partial_\beta \partial_\beta \rho, \]  
(A21)

where in the last step we have done a two-fold partial integration. Since this result must hold for any \( u \), we find for the force density \( f \) the unique result
\[ f = \kappa \rho \nabla \nabla^2 \rho. \]  
(A22)

The dynamics is therefore, by construction, energy-conserving. It is however also momentum-conserving, since the total force applied to the system vanishes:
\[ \int d^d \mathbf{r} f = 0; \]  
(A23)

this latter relation is easily shown by inserting the explicit formula Eq. A22, and doing a three-fold partial integration, which shows
\[ \int d^d \mathbf{r} f = - \int d^d \mathbf{r} f. \]  
(A24)

Alternatively, it is also possible to write \( f \) as the divergence of a stress tensor. This latter approach has been mainly pursued by Swift et al. [9, 10]; it has however the disadvantage that the stress tensor is in general not unique, while the force term definitely is, as has been shown by the present derivation.

Appendix B: Equation of state

The purpose of this appendix is to demonstrate, by explicit construction, that it is possible to find an equation of state that satisfies all the conditions for the present model.

We first recall that the equation of state is written as
\[ p(\rho) = \rho c_s^2(\rho), \]  
(B1)

and introduce the function
\[ \psi(\rho) := \frac{\partial}{\partial \rho} \ln c_s^2(\rho), \]  
(B2)

for which we note the identity
\[ \frac{\rho}{p} \frac{\partial}{\partial \rho} \ln p = \frac{\partial}{\partial \rho} \left[ (\ln \rho + \ln c_s^2) \right] = 1 + \rho \psi. \]  
(B3)

Secondly, we recall all the conditions that the equation of state has to satisfy:

- Positivity of weights:
\[ c_{s,min}^2 < c_s^2 < c_{s,max}^2; \]  
(B4)

where \( c_{s,min}^2 = 0.3510760, c_{s,max}^2 = 1.333333 \) in two dimensions, while \( c_{s,min}^2 = 0.3850612, c_{s,max}^2 = 1.1917145 \) in three dimensions (in lattice units, where lattice spacing and time step have been set to unity).
• Two-phase coexistence: There must be some density interval for which the equation of state is unstable; i.e.

\[ \frac{\partial p}{\partial \rho} < 0 \] (B5)

or

\[ \psi(\rho) < -\frac{1}{\rho} \] (B6)

• Positivity of the bulk viscosity (in \( d \) dimensions):

\[ \frac{\rho}{p} \frac{\partial p}{\partial \rho} < 1 + \frac{2}{d} \] (B7)

or (for all values of \( \rho \))

\[ \psi(\rho) < \frac{2}{d} \frac{1}{\rho} \] (B8)

Our strategy to find a valid equation of state therefore consists of first constructing a function \( \psi(\rho) \) that satisfies Eqs. B6 and B8. After picking some density value \( \rho_0 \) and its corresponding \( c_s^2 \) value, we then find, via integration

\[ c_s^2(\rho) = c_s^2(\rho_0) \exp \left( \int_{\rho_0}^\rho d\rho' \psi(\rho') \right), \] (B9)

and the final step is to verify that this function satisfies Eq. B4 for all \( \rho \) values.

In order to construct a very simple model function for \( \psi \), we choose two densities \( \rho_1 \) and \( \rho_2 \) with \( \rho_1 < \rho_2 \) and set \( \rho_3 = 2\rho_2 - \rho_1 \). Furthermore, we choose an amplitude \( A \geq 0 \) and assume

\[ \psi(\rho) = \begin{cases} 
0 & \rho \leq \rho_1 \\
A \sin \left( \frac{\pi \rho - \rho_1}{\rho_2 - \rho_1} \right) & \rho_1 < \rho < \rho_3 \\
0 & \rho_3 \leq \rho. \end{cases} \] (B10)

Figure 1 shows this function for various values of the amplitude \( A \), where we have chosen (in some arbitrary units) \( \rho_1 = 0.5, \rho_2 = 1 \). We also compare with the right hand sides of Eqs. B6 and B8, where we have picked the spatial dimension \( d = 2 \). One sees that for large values of \( A \) the condition of positive bulk viscosity is violated, while for too small amplitudes there is no two-phase coexistence. However, there is a certain window of admissible amplitudes (for example, \( A = 1.2 \) in Fig. 1) where both conditions are met. Now, choosing \( c_s^2(\rho_1) = 0.6 \), we can also plot the function \( c_s^2(\rho) \) (Fig. 2), from which we see that Eq. B4 is satisfied as well. It should be noted that we have constructed our function \( \psi \) in such a way that

\[ \int_{\rho_1}^{\rho_3} d\rho \psi(\rho) = 0, \] (B11)

which means that \( c_s^2 \) takes the same value for \( \rho < \rho_1 \) and \( \rho > \rho_3 \). This is a simplifying feature which is however not necessary for the validity of the model.

Having thus found a valid function \( c_s^2(\rho) \), we can now proceed to look at the equation of state \( p(\rho) \). This is done in Fig. 3, for the parameters of Figs. 1 and 2, where we focus attention on the valid amplitude \( A = 1.2 \). The same data are re-plotted as a function of specific volume \( 1/\rho \) in Fig. 4, focusing on the interesting coexistence region. This representation is amenable to the standard Maxwell construction (also shown) that allows us to determine the coexistence densities. The Maxwell construction was facilitated by numerical root-finding, combined with a tabulated free energy per unit mass \( f \), which we found by numerically integrating the relation \( \partial f/\partial \rho = p/\rho^2 \), and normalizing by the requirement \( f(\rho = \rho_1) = 0 \).

It should be noted that the amplitude \( A \) must be viewed as the essential parameter that controls the thermodynamics of the system. For \( A = 0 \), we recover the equation of state of an ideal gas. For larger values of \( A \), the equation of state more and more deviates from ideality, until we reach a value beyond which the equation
ψ(ρ) = −1/ρ has solutions. This is the system’s (Mean Field) critical point. From then on, the equation of state assumes a more and more pronounced unstable region (∂p/∂ρ < 0), indicative of two-phase coexistence, until finally A becomes so large that the bulk viscosity becomes negative — this corresponds to a situation where the system has been quenched so deeply into the two-phase region that the LB model with its limited set of velocities is no longer able to represent the physics in a consistent and numerically stable fashion.

By systematically solving the Maxwell construction for various values of A, we can finally find the system’s bulk phase diagram in the ρ vs. A plane. It is presented in Fig. 5.

**Appendix C: Miscellaneous expressions in the derivation of the closure**

1. \( \pi^{*}(1)_{\alpha\beta} - \pi^{(1)}_{\alpha\beta} \)

\( \pi^{*}(1)_{\alpha\beta} - \pi^{(1)}_{\alpha\beta} \) is determined via Eq. 116:

\[
\frac{1}{\hbar} \left( \pi^{*}(1)_{\alpha\beta} - \pi^{(1)}_{\alpha\beta} \right) = \partial_{\gamma_{1}} \pi^{(0)}_{\alpha\beta} + \partial_{\gamma} \phi^{(0)}_{\alpha\beta\gamma},
\]

(C1)

It should be noted that \( \pi^{(0)}_{\alpha\beta} \) and \( \phi^{(0)}_{\alpha\beta\gamma} \) are functions only of the hydrodynamic variables (see Eqs. 31 and 32). Furthermore, at the first-order level of the CE expansion, the dynamics of the hydrodynamic variables is simply given by the continuity and the Euler equations (see Eqs. 106 and 111). Now we observe that, neglecting terms of order \( u^3 \), we can write

\[
\phi^{(0)}_{\alpha\beta\gamma} = \pi^{(0)}_{\alpha\beta} u_{\gamma} + p (u_{\alpha} \delta_{\beta\gamma} + u_{\beta} \delta_{\alpha\gamma}),
\]

(C2)

i.e.

\[
\partial_{\gamma_{1}} \phi^{(0)}_{\alpha\beta\gamma} = \partial_{\gamma_{1}} \left( \pi^{(0)}_{\alpha\beta} u_{\gamma} + \partial_{\alpha_{1}} (pu_{\beta}) + \partial_{\beta_{1}} (pu_{\alpha}) \right).
\]

(C3)

Therefore we may write (cf. also App. A):

\[
\frac{1}{\hbar} \left( \pi^{*}(1)_{\alpha\beta} - \pi^{(1)}_{\alpha\beta} \right) = \rho \frac{D}{Dt} \left( \frac{1}{\rho} \pi^{(0)}_{\alpha\beta} \right) + \partial_{\alpha_{1}} (pu_{\beta}) + \partial_{\beta_{1}} (pu_{\alpha})
\]

(C4)

= \rho \frac{D}{Dt} \left( \frac{p}{\rho} \delta_{\alpha\beta} + u_{\alpha} u_{\beta} \right) + \partial_{\alpha_{1}} (pu_{\beta}) + \partial_{\beta_{1}} (pu_{\alpha})

= \rho \frac{D}{Dt} \left( p \rho \delta_{\alpha\beta} \right) - p \rho \delta_{\alpha\beta} \partial_{\gamma_{1}} u_{\gamma} - u_{\beta} \partial_{\alpha_{1}} p - u_{\alpha} \partial_{\beta_{1}} p + \partial_{\alpha_{1}} (pu_{\beta}) + \partial_{\beta_{1}} (pu_{\alpha})

= \left( p - \rho \frac{D}{Dt} \right) \delta_{\alpha\beta} \partial_{\gamma_{1}} u_{\gamma} + p (\partial_{\alpha_{1}} u_{\beta} + \partial_{\beta_{1}} u_{\alpha} ).
2. $\phi^{*\dagger}_{\alpha \beta \gamma} - \phi^{\dagger}_{\alpha \beta \gamma}$

This expression is derived from Eq. 117,
\[
\frac{1}{\hbar} \left( \phi^{*\dagger}_{\alpha \beta \gamma} - \phi^{\dagger}_{\alpha \beta \gamma} \right) = \partial_{\gamma} \phi^{0}_{\alpha \beta \gamma} + \partial_{\delta} \psi^{0}_{\alpha \beta \gamma \delta},
\]
and we proceed in a quite analogous fashion. First, from Eqs. 31 – 33 we conclude
\[
\psi^{(0)}_{\alpha \beta \gamma \delta} = \phi^{0}_{\alpha \beta \gamma \delta} u_{\delta} + \frac{\rho}{\rho} \left( \pi^{(0)}_{\alpha \beta \gamma \delta} + \pi^{(0)}_{\alpha \delta \delta} + \pi^{(0)}_{\beta \gamma \delta} \right)
\]
and
\[
\partial_{\beta_{i}} \psi^{(0)}_{\alpha \beta \gamma \delta} = \partial_{\beta_{i}} \left( \phi^{(0)}_{\alpha \beta \gamma \delta} u_{\delta} \right) + \partial_{\gamma_{i}} \left( \frac{\rho}{\rho} \pi^{(0)}_{\alpha \beta \gamma} \right) + \partial_{\delta_{i}} \left( \frac{\rho}{\rho} \pi^{(0)}_{\alpha \beta \gamma \delta} \right) \quad \text{(C7)}
\]
From this we conclude (note that again $\partial_{\gamma_{i}}$ implies simple Euler dynamics)
\[
\frac{1}{\hbar} \left( \phi^{*\dagger}_{\alpha \beta \gamma} - \phi^{\dagger}_{\alpha \beta \gamma} \right) = \rho \frac{D}{Dt_{1}} \left( \frac{\rho}{\rho} \right) u_{\gamma_{i}} + \partial_{\gamma_{i}} \left( \frac{\rho}{\rho} \pi^{(0)}_{\alpha \beta \gamma} \right) + \partial_{\beta_{i}} \left( \frac{\rho}{\rho} \pi^{(0)}_{\alpha \beta \gamma \delta} \right) \quad \text{(C8)}
\]

Now,
\[
\rho \frac{D}{Dt_{1}} \left( \frac{p}{\rho} u_{\gamma_{i}} \right) = -\rho \frac{D}{Dt_{1}} \left( \frac{p}{\rho} u_{\gamma_{i}} \right) + \partial_{\gamma_{i}} \left( \frac{p}{\rho} \right) u_{\gamma_{i}} u_{\delta_{i}} \quad \text{(C9)}
\]
and
\[
\partial_{\gamma_{i}} \left( \frac{p}{\rho} \right) = -\partial_{\gamma_{i}} \left( \frac{p}{\rho} \right) u_{\gamma_{i}} \partial_{\delta_{i}} u_{\delta_{i}} + \partial_{\gamma_{i}} \left( pu_{\beta_{i}} \right) \quad \text{(C10)}
\]

Hence,
\[
\frac{1}{\hbar} \left( \phi^{*\dagger}_{\alpha \beta \gamma} - \phi^{\dagger}_{\alpha \beta \gamma} \right) = \partial_{\gamma_{i}} \left( pu_{\beta_{i}} \right) + \delta_{\beta_{i}} \left( \rho \frac{D}{Dt_{1}} \left( \frac{p}{\rho} \right) u_{\gamma_{i}} \partial_{\delta_{i}} u_{\delta_{i}} \right) + \text{perm.}
\]

3. $\partial_{\gamma_{i}} (\pi^{*\dagger}_{\alpha \beta} - \pi^{\dagger}_{\alpha \beta})$

From Eq. C4 we find
\[
\frac{1}{\hbar} \partial_{\gamma_{i}} \left( \pi^{*\dagger}_{\alpha \beta} - \pi^{\dagger}_{\alpha \beta} \right) = \partial_{\gamma_{i}} \left[ p \left( \partial_{\alpha_{i}} u_{\beta_{i}} + \partial_{\beta_{i}} u_{\alpha_{i}} \right) + \left( p - \frac{\partial p}{\partial \rho} \right) \delta_{\alpha_{i}} \partial_{\gamma_{i}} u_{\alpha_{i}} \right] \quad \text{(C12)}
\]
and
\[
\delta_{\alpha_{i}} \partial_{\gamma_{i}} u_{\alpha_{i}} = -\frac{1}{p} \partial_{\alpha_{i}} u_{\alpha_{i}} - u_{\alpha_{i}} \partial_{\gamma_{i}} u_{\alpha_{i}} \quad \text{(C14)}
\]

Furthermore, we have
\[
\partial_{\gamma_{i}} u_{\alpha_{i}} = -\partial_{\gamma_{i}} u_{\alpha_{i}} \quad \text{(C15)}
\]

Inserting these results, we find
\[
\frac{1}{\hbar} \left( \pi^{*\dagger}_{\alpha \beta} - \pi^{\dagger}_{\alpha \beta} \right) = -\frac{\partial p}{\partial \rho} \delta_{\alpha_{i}} \partial_{\gamma_{i}} \left( \partial_{\alpha_{i}} u_{\beta_{i}} + \partial_{\beta_{i}} u_{\alpha_{i}} \right) - p \delta_{\alpha_{i}} \partial_{\gamma_{i}} \left( \frac{1}{\rho} \partial_{\alpha_{i}} p + \frac{1}{\rho} \partial_{\alpha_{i}} p \right) + \frac{\partial^{2} p}{\partial \rho^{2}} \delta_{\alpha_{i}} \partial_{\gamma_{i}} \left( \frac{1}{\rho} \partial_{\alpha_{i}} p + \frac{1}{\rho} \partial_{\alpha_{i}} p \right) \quad \text{(C16)}
\]

4. $\partial_{\beta_{i}} \pi^{(0)}_{\alpha \beta}$

On the $t_{2}$ time scale the dynamics is simply given by
\[
(cf. Eqs. 107 and 112, taking into account that $j^{(1)}$ vanishes)
vanishes, plus of Eq. C4. Therefore,
\[
\partial_{t_2} \pi^{(0)}_{\alpha \beta} = \partial_{t_2} \left( \rho \delta_{\alpha \beta} + \rho u_\alpha u_\beta \right) = \rho \left( u_\alpha \partial_{t_2} u_\beta + u_\beta \partial_{t_2} u_\alpha \right)
\]
From this, we conclude
\[
\partial_{t_2} \rho = 0,
\]
\[
\partial_{t_2} j_\alpha = -\frac{1}{2} \partial_{t_1} \left( \pi^{(1)}_{\alpha \beta} + \pi^{(1)}_{\beta \alpha} \right)
\]
\[
= \frac{1}{2} \frac{\gamma + 3}{2} \gamma - 1 \partial_{t_1} \left( \pi^{(1)}_{\alpha \beta} - \pi^{(1)}_{\beta \alpha} \right)
\]
\[
= -\frac{h + 1}{2} \gamma - 1 \partial_{t_1} \left[ \left( p - \frac{\partial p}{\partial \rho} \right) \partial_{t_1} u_\gamma \right]
\]
\[
- \frac{h + 1}{2} \gamma - 1 \partial_{t_1} \left[ \rho \left( \partial_{t_2} u_\beta + \partial_{t_2} u_\alpha \right) \right],
\]
where in the transformations of $\partial_{t_2} j_\alpha$ we have made use of the results of Sec. VII C, taking into account that $\Sigma^{(1)}_{\alpha \beta}$

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