

Practical Kinetic Models for Dense Fluids

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A novel approach to constructing kinetic models is proposed on the basis of the separation of local and nonlocal contributions to particle interaction. The method results in a generic kinetic equation for complex fluid systems, amenable to efficient numerical realization. The main obstruction to causality caused by a coupling between the nonuniform temperature field and the nonideal equation of state is resolved based on gauge invariance of the hydrodynamic limit. A new lattice Boltzmann model is derived for compressible nonideal fluid simulation based on the Enskog-Vlasov kinetic theory as a case of practical importance.

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The lattice Boltzmann method (LBM) [1–5] is a modern approach to the simulation of complex flows such as turbulence [6], surface and multiphase phenomena [7–10], soft-glassy materials [11], relativistic hydrodynamics [12], hemodynamics [13], and *ab initio* electronic structure calculations [14]. LBM is a recast of continuum fluid mechanics in the form of a fully discrete kinetic equation for the populations of designer particles $f_i(\mathbf{r}, t)$, with the simplest rule of propagation in a space-filling lattice formed by discrete velocities $\mathbf{c}_i$, in discrete-time steps, and relaxation at the nodes $\mathbf{r}$ to a local equilibrium $f_i^{\text{eq}}(\mathbf{r}, t)$. So far, most of the LBM success remains in the realm of isothermal simulations, including nonideal fluids [15–17]. More recent progress for compressible ideal gas [18–20] enables extension to combustion [21]. However, the interplay between compressibility and the nonideal equation of state remains a challenge [22–28].

In this Letter, we derive a consistent lattice Boltzmann model for compressible nonideal fluids. Our approach is based on a new representation of local and nonlocal contributions of the interaction of particles. The analysis of the resulting generic kinetic equation demonstrates implicit coupling between a nonuniform temperature field and the nonideal equation of state. This obstruction is resolved by introducing a gauge LBM formulation. Realization on the standard discrete velocity set leads to a simple and fully explicit and thermodynamically consistent LBM for compressible nonideal fluid, enabling supersonic flow simulations in sub- and supercritical regimes.

A formal starting point is the Bogoliubov-Born-Green-Kirkwood-Yvon equation (BBGKY), $\partial_t f + \mathbf{v} \cdot \nabla f = \mathcal{J}$, with the interaction integral $\mathcal{J}$ [29],

$$\mathcal{J} = \frac{1}{m} \int \nabla V(|\mathbf{r} - \mathbf{r}'|) \cdot \frac{\partial}{\partial \mathbf{v}} f_2(\mathbf{r}, \mathbf{v}, \mathbf{r}', \mathbf{v}', t) d\mathbf{v}' d\mathbf{r}'. \quad (1)$$

Here m is the molecular mass, $f(\mathbf{r}, \mathbf{v}, t)$ and $f_2(\mathbf{r}, \mathbf{v}, \mathbf{r}', \mathbf{v}', t)$ are the one- and two-particle distribution functions, respectively, $\mathbf{r}, \mathbf{r}'$ and $\mathbf{v}, \mathbf{v}'$ are the position and velocity of the particles, and V is a pair interaction potential. Although the BBGKY equation satisfies local mass conservation $\int \mathcal{J} d\mathbf{v} = 0$, momentum and energy are not locally conserved due to a nonlocal force $\mathbf{F}$ and a nonlocal kinetic energy rate of change $\tilde{Q}$,

$$\mathbf{F}(\mathcal{J}) = \int m \mathbf{v} \mathcal{J} d\mathbf{v}, \quad (2)$$

$$\tilde{Q}(\mathcal{J}) = \int \frac{m(\mathbf{v} - \mathbf{u})^2}{2} \mathcal{J} d\mathbf{v}, \quad (3)$$

where $\mathbf{u}$ is the flow velocity. Our first step is to formally split the interaction into local and nonlocal parts relative to the local Maxwellian,

$$f^{\text{eq}}(\mathbf{u}, T) = \frac{n}{(2\pi RT)^{3/2}} \exp \left[-\frac{(\mathbf{v} - \mathbf{u})^2}{2RT} \right], \quad (4)$$

where n is the number density, R is the specific gas constant, and T is the temperature. To this end, we consider the following operation applied to the interaction integral (1):

$$\mathcal{K}_\lambda(\mathcal{J}) = \frac{1}{\lambda} [f^{\text{eq}}(\mathbf{u}_\lambda^*, T_\lambda^*) - f^{\text{eq}}(\mathbf{u}, T)], \quad (5)$$

where the *shifted flow velocity* $\mathbf{u}_\lambda^*$ and the *shifted temperature* T_λ^* include the nonlocality (2) and (3),

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$$\mathbf{u}_\lambda^* = \mathbf{u} + \frac{\lambda}{\rho} \mathbf{F}, \quad (6)$$

$$T_\lambda^* = T + \frac{2\lambda}{3R\rho} \tilde{Q} - \frac{\lambda^2}{3R\rho^2} \mathbf{F} \cdot \mathbf{F}. \quad (7)$$

Here, $\rho = mn$ is the density of the fluid, and $\lambda > 0$ is a parameter with the dimension of time, which will be specified in the following. Direct computation verifies that operator (5) is a projector (albeit nonlinear), $\mathcal{K}_\lambda[\mathcal{K}_\lambda(\mathcal{J})] = \mathcal{K}_\lambda(\mathcal{J})$, and satisfies the orthogonality condition, $\mathcal{K}_\lambda[\mathcal{J} - \mathcal{K}_\lambda(\mathcal{J})] = 0$. Therefore, operation (5) can be used to formally split the interaction $\mathcal{J}$ into local and nonlocal parts, $\mathcal{J} = \mathcal{J}_{\text{loc}} + \mathcal{J}_{\text{nloc}}$, where $\mathcal{J}_{\text{loc}} = \mathcal{J} - \mathcal{K}_\lambda(\mathcal{J})$ and $\mathcal{J}_{\text{nloc}} = \mathcal{K}_\lambda(\mathcal{J})$. The local part $\mathcal{J}_{\text{loc}}$ satisfies the local conservation of mass, momentum, and kinetic energy, $\int \{1, \mathbf{v}, v^2\} \mathcal{J}_{\text{loc}} d\mathbf{v} = 0$. Hence, a rationale for $\mathcal{J}_{\text{loc}}$ is the Bhatnagar-Gross-Krook (BGK) approximation, $\mathcal{J}_{\text{loc}} \rightarrow -(1/\tau)(f - f^{\text{eq}})$, where τ is the relaxation time, and the formal kinetic model becomes

$$\partial_t f + \mathbf{v} \cdot \nabla f = -\frac{1}{\tau}(f - f^{\text{eq}}) + \frac{1}{\lambda}(f^* - f^{\text{eq}}), \quad (8)$$

where $f^* = f^{\text{eq}}(\mathbf{u}_\lambda^*, T_\lambda^*)$ is the *shifted equilibrium* in (5). The kinetic equation (8) has a special advantage for space-time discretization. To that end, the standard method of integration along characteristics and evaluation of quadratures by a trapezoidal rule [30,31] results in a second-order accurate formal scheme (see details in Supplemental Material [32]),

$$f(\mathbf{r} + \mathbf{v}\delta t, t + \delta t) = f + 2\beta(f^{\text{eq}} - f) + \frac{\delta t}{\lambda}(1 - \beta)(f^* - f^{\text{eq}}), \quad (9)$$

where the right-hand side is evaluated at time t and position $\mathbf{r}$, where δt is the time step, and where $\beta \in [0, 1]$ is related to the relaxation time, $\beta = \delta t/(2\tau + \delta t)$. Moreover, the flow velocity $\mathbf{u}$ and the temperature T entering the equilibrium and the shifted equilibrium distributions elsewhere in (9) are computed according to a transformation (which is key to second-order accuracy),

$$\rho \mathbf{u} = \int m \mathbf{v} f d\mathbf{v} + \frac{\delta t}{2} \mathbf{F}, \quad (10)$$

$$\frac{3}{2} \rho RT = \int \frac{m(\mathbf{v} - \mathbf{u})^2}{2} f d\mathbf{v} + \frac{\delta t}{2} \tilde{Q}. \quad (11)$$

This concludes the formal part of the analysis. Our approach to splitting the local and nonlocal contributions is similar to that of [33] but instead uses a nonlinear projection (5). The outcome is a new kinetic model (8) tailored to achieve an

efficient scheme (9), provided that the transformations (10) and (11) are solvable.

To address the latter point, we substantiate the non-locality (2) and (3) using the Enskog-Vlasov kinetic theory [34–37] where hard-sphere collisions and a long-range attraction potential contribute to nonlocal momentum and energy transfer. Evaluation with the Enskog-Vlasov interaction integral results in a closed-form approximation (see details in Supplemental Material [32]),

$$\mathbf{F} = -\nabla(P_E - P_0) - \rho \nabla V_V, \quad (12)$$

$$\tilde{Q} = -(P_E - P_0)(\nabla \cdot \mathbf{u}). \quad (13)$$

Here, P_E is the Enskog pressure that we assume in van der Waals form, $P_E = \rho RT/(1 - b\rho)$, with b the excluded volume parameter and $P_0 = \rho RT$ the ideal gas pressure. Furthermore, $V_V = \int V(|\mathbf{r} - \mathbf{r}'|) \rho(\mathbf{r}', t) d\mathbf{r}'$ is Vlasov's mean field potential producing the force, $\mathbf{F}_V = -\rho \nabla V_V \approx -\nabla P_V + \kappa \rho \nabla \nabla \cdot \nabla \rho$, where $P_V = -a\rho^2$ is Vlasov's pressure contribution, while κ is the capillarity coefficient. The Enskog and Vlasov pressure contributions sum up to the van der Waals equation of state, $P = P_E + P_V$. The rate of kinetic energy change due to collisional transport is manifested in the corresponding compression work (13).

Inspection of the Enskog-Vlasov force (12) reveals a coupling between momentum and temperature “hidden” in the transformations (10) and (11), triggered by nonuniformity of temperature in Enskog's contribution to the force (12). This makes the transformations (10) and (11) *implicit*. Such a coupling vanishes for the compressible ideal gas, which arguably characterizes the nonideal compressible fluid as one of the hardest cases. We will resolve this obstruction below.

Discrete-in-time equations (9) become particularly useful when the velocity is also rendered discrete. This is the case of the lattice Boltzmann realization using the standard discrete velocity set, $\mathbf{c}_i = (c_{ix}, c_{iy}, c_{iz})$, $c_{i\alpha} \in \{-1, 0, 1\}$, and we only need to specify the corresponding equilibrium and the shifted equilibrium. We first address mass and momentum transport. Let us define functions in two variables, ξ_α and $\zeta_{\alpha\alpha}$, $\alpha \in \{x, y, z\}$,

$$\Psi_{i\alpha}(\xi_\alpha, \zeta_{\alpha\alpha}) = 1 - c_{i\alpha}^2 + \frac{1}{2}[(3c_{i\alpha}^2 - 2)\zeta_{\alpha\alpha} + c_{i\alpha}\xi_\alpha]. \quad (14)$$

All pertinent populations are constructed using products of functions (14) by assigning certain values to place holders ξ_α and $\zeta_{\alpha\alpha}$. For equilibrium populations, we set $\xi_\alpha^{\text{eq}} = u_\alpha$ and $\zeta_{\alpha\alpha}^{\text{eq}} = P_0/\rho + u_\alpha^2$ to obtain

$$f_i^{\text{eq}} = \rho \prod_\alpha \Psi_{i\alpha}\left(u_\alpha, \frac{P_0}{\rho} + u_\alpha^2\right). \quad (15)$$

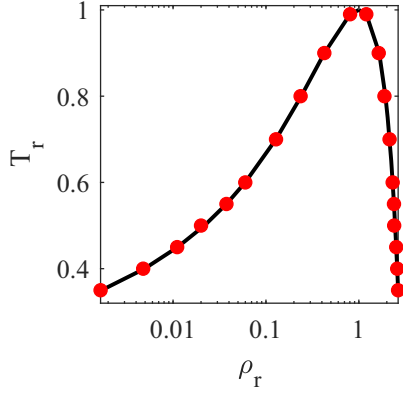


FIG. 1. Liquid-vapor coexistence for the van der Waals equation of state, with the reference pressure $P_0 = P_E$. Axes are reduced by critical values of density and temperature. Line: saturated liquid and vapor lines by Maxwell's equal-area construction; Symbol: simulation. Data available at [39].

Note that a *reference pressure* P_0 is not yet specified and is left as a parameter. For the shifted equilibrium, the velocity is set in accordance with (7): $\xi_\alpha^* = u_\alpha + (\lambda/\rho)F_\alpha$ and $\zeta_{\alpha\alpha}^* = P_0/\rho + (\xi_\alpha^*)^2 + (\lambda/\rho)\Phi_{\alpha\alpha}$, with the nonequilibrium momentum flux correction as

$$\Phi_{\alpha\alpha} = -\partial_\alpha \left[\rho u_\alpha \left(u_\alpha^2 + \frac{3P_0}{\rho} - 3\zeta^2 \right) \right] + \Phi'. \quad (16)$$

Here $\zeta = 1/\sqrt{3}$ is the lattice speed of sound, while the second term reads

$$\Phi' = P_0 \left(\frac{5}{3} - \gamma_0 \right) (\nabla \cdot \mathbf{u}), \quad (17)$$

where γ_0 is a pseudoadiabatic parameter,

$$\gamma_0 = \left(\frac{\partial \ln P_0}{\partial \ln \rho} \right) + \frac{2}{3} \left(\frac{P_E}{\rho RT} \right) \left(\frac{\partial \ln P_0}{\partial \ln T} \right). \quad (18)$$

The resulting lattice Boltzmann equation follows the pattern of Eq. (9) upon replacement $f \rightarrow f_i$, whereby the left-hand side becomes exact on-lattice propagation while the flow velocity elsewhere in the equilibrium and shifted equilibrium populations is computed as in (11), with summation over discrete velocities replacing integration. The proposed lattice Boltzmann model adopts the necessary adjustments due to the minimalist discrete velocity setting: the first term in (16) is a correction of viscous stress [19], while the second correction term (17) maintains the vanishing bulk viscosity of the Enskog–Vlasov fluid. Finally, since the reference pressure was left as a parameter in the equilibrium (15), it is also used consistently as a parameter in the force (12). With this, the continuity and the momentum equations are recovered as follows (Supplemental Material [32] provides details of the Chapman–Enskog analysis [38]):

$$\partial_t \rho + \nabla \cdot \rho \mathbf{u} = 0, \quad (19)$$

$$\partial_t \rho \mathbf{u} + \nabla \cdot \rho \mathbf{u} \otimes \mathbf{u} + \nabla P_E + \rho \nabla V_V + \nabla \cdot \mathbf{T}_{NS} = 0, \quad (20)$$

where $\mathbf{T}_{NS} = -\mu \mathbf{S}$ is the Navier-Stokes viscous stress tensor, with $\mathbf{S} = \nabla \mathbf{u} + \nabla \mathbf{u}^\dagger - (2/3)(\nabla \cdot \mathbf{u})\mathbf{I}$ being the trace-free rate-of-strain tensor, while $\mu = (1/2)(1/\beta - 1)P_0\delta t$ is the viscosity. Note that the momentum equation (20) is *form invariant* with respect to the reference pressure P_0 and is independent of the relaxation parameter λ . As an interim example, simulation with the current lattice Boltzmann model at a fixed temperature in Fig. 1 shows the liquid-vapor coexistence diagram for $P_0 = P_E$ (this choice of reference pressure will be explained shortly).

Moving on to the energy dynamics, we adopt a double-population path, following [19,20,40]. With this, the *total kinetic energy*, ρE , is the locally conserved moment of the populations g_i , and E is a specific kinetic energy,

$$E(\mathbf{u}, T) = \frac{3}{2}RT + \frac{1}{2}u^2. \quad (21)$$

Equilibrium and shifted equilibrium g populations are constructed as in [19,20,40]. To that end, the operators $\mathcal{O}_\alpha$ acting on the function (21) are introduced as [19,20,40]

$$\mathcal{O}_\alpha E(\mathbf{u}, T) = \left(\frac{P_0}{\rho} \right) \frac{\partial E(\mathbf{u}, T)}{\partial u_\alpha} + u_\alpha E(\mathbf{u}, T). \quad (22)$$

By setting symbols $\xi_\alpha = \mathcal{O}_\alpha$, $\zeta_{\alpha\alpha} = \mathcal{O}_\alpha^2$ in the functions (14), equilibrium populations g_i^{eq} are defined by the action of the product operators on the generating function (21),

$$g_i^{\text{eq}}(\mathbf{u}, T) = \rho \prod_\alpha \Psi_{i\alpha}(\mathcal{O}_\alpha, \mathcal{O}_\alpha^2) E(\mathbf{u}, T). \quad (23)$$

It is easily verified that equilibrium populations (23) satisfy all the constraints required to recover the target energy balance equation (see the details in Supplemental Material [32]). Furthermore, the shifted equilibrium g populations are defined by replacing the arguments in (23) according to the flow velocity and temperature shift (6) and (7), and adding a correction,

$$g_i^* = g_i^{\text{eq}}(\mathbf{u}_\lambda^*, T_\lambda^*) + \begin{cases} \frac{1}{2} \mathbf{c}_i \cdot \mathbf{q}', & \text{if } c_i^2 = 1, \\ 0, & \text{otherwise,} \end{cases} \quad (24)$$

where the nonequilibrium energy flux correction is the vector $\mathbf{q}'$ with the components,

$$q'_\alpha = \lambda P_0 \partial_\alpha \left(\frac{P_0}{\rho} - RT \right) + \lambda u_\alpha \Phi'. \quad (25)$$

The first term in (25) provides a correction to the Fourier heat flux, while the second term is required for consistency

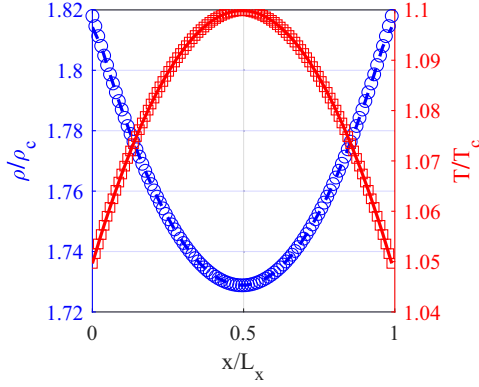


FIG. 2. Distribution of density and temperature along the channel. Red filled and blue dashed lines are numerical results for temperature and density. Red square marker and blue circular marker are corresponding analytical solutions. Data available at [39].

with the above bulk viscosity correction (17). The lattice Boltzmann equation for the g populations follows again the pattern of (9), and the total kinetic energy balance equation is recovered as

$$\partial_t \rho E + \nabla \cdot \rho \mathbf{u} E + \mathbf{u} \cdot (\nabla P_E + \rho \nabla V_V) + P_E (\nabla \cdot \mathbf{u}) + \nabla \cdot (\mathbf{u} \cdot \mathbf{T}_{NS}) - \nabla \cdot (k \nabla T) = 0, \quad (26)$$

where $k = (5/2)R\mu$ is the thermal conductivity.

Since also the energy balance equation (26) is form invariant with respect to the reference pressure P_0 , this allows us to choose the *gauge* P_0 based on convenience. At first glance, it seems natural to apply the ideal gas gauge, $P_0 = \rho RT$, as this annihilates the Fourier flux correction in (25). However, a closer look reveals that the transformation relations (10) and (11) remain strongly coupled, as discussed above, and result in an implicit problem of elliptic type. Instead, a gauge that renders the transformations (10) and (11) explicitly solvable is Enskog's gauge,

$$P_0 = P_E. \quad (27)$$

In fact, with (27), the kinetic energy rate cancels in (13), while the temperature dependence cancels in the nonlocal force (12). To avoid possible confusion, the “cancellation” of the mentioned terms in the force and kinetic energy rate does not imply a “neglect” of the corresponding fluxes in the hydrodynamic balance equations. Rather, the corresponding contributions are delegated to the local equilibrium (15) evaluated at the chosen reference pressure P_0 . This features the gauge invariance of the hydrodynamic limit.

With the choice of the gauge (27), the fully explicit lattice Boltzmann model of the Enskog-Vlasov fluid is complete, and we set $\lambda = \delta t$. For a numerical validation, we consider a supersonic thermal Couette flow in a two-dimensional

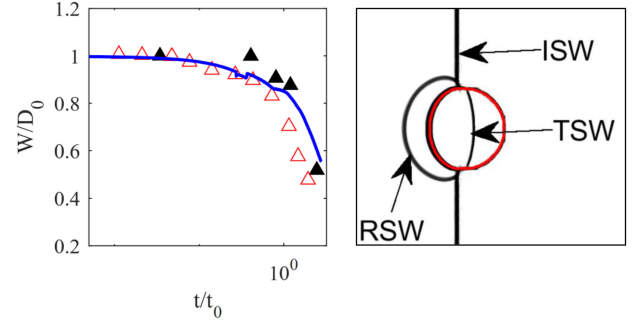


FIG. 3. Right: Schlieren image of shock interaction with liquid drop at $t/t_0 = 0.3$. ISW, incident shock wave; TSW, transmitted shock wave; RSW, reflected shock wave. Left: evolution of drop size along x axis W . Solid line, present simulation; filled black triangles, experiment [41]; red triangles, simulation of [23]. Data available at [39].

channel of width L_x between a static and a moving wall, defined via the following boundary conditions, where T_w is the temperature of both walls and U_w is the speed of the moving wall. In steady state, once energy loss at the walls is balanced by energy production due to viscous heating, a steady state solution is reached. The simulation was performed for $Ma = 1.2$ on the moving wall. The results are compared to the analytical solution in Fig. 2 and point to excellent agreement.

Finally, we present a two-dimensional simulation of the early stage interaction of a planar shock wave with a cylindrical water column at $Ma = 1.3$, at the initial subcritical temperature $T_r = 0.9$. A snapshot of the shock propagating through and reflecting from the liquid at $t/t_0 = 0.3$ is shown in Fig. 3. The time was reduced by $t_0 = (D_0/u_s)(\zeta^v/\zeta^l)\sqrt{\rho^l/\rho^v}$, where D_0 is the initial diameter of the liquid cylinder, u_s is the shock speed, $\rho^{v,l}$ are the saturated vapor and liquid densities, respectively, and $\zeta^{v,l}$ are the corresponding speed of sound. The evolution of the size of the liquid column cross section in Fig. 3 shows a good comparison to the experiment and the reference simulation data.

In summary, we have proposed a novel strategy to derive kinetic models of practical utility. This is achieved by using a formalism of shifted equilibrium to split the local and nonlocal contributions of the particles' interaction and to produce a formal kinetic model amenable to efficient time discretization. The generic formalism was substantiated with the classical Enskog-Vlasov theory to derive a new lattice Boltzmann model for a compressible nonideal fluid. Targeting the hydrodynamic limit, we revealed the gauge invariance of the theory with respect to a reference pressure. Analysis shows that in the nonideal compressible case the requirement of explicitness (and thus causality) of the lattice Boltzmann method is critical constraint, which led to selecting Enskog's excluded volume pressure as the reference.

In a retrospective, we note that the low density limit of the present theory yields $P_E \rightarrow \rho RT$ and returns the conventional lattice Boltzmann models for compressible ideal gas [19,20,40]. We also note that, while Enskog's gauge (27) emerges as a natural reference pressure, this choice is not unique. Another gauge that makes the transformations (10) and (11) explicit is the total thermodynamic pressure, $P_0 = P = P_E + P_V$. The latter gauge was first suggested on phenomenological grounds in the aforementioned work by Reyhanian *et al.* [23]. However, the authors used a different off-lattice propagation, based on the particle-on-demand approach [42], and a different force representation. In contrast, the present lattice Boltzmann system for compressible nonideal fluids on the standard minimalist lattice is essentially as simple and efficient as the classical; the (unoptimized) realization used to perform the above simulations demonstrated a 50% computation overhead compared to a single-population (standard) LBM and only a negligible overhead compared to a two-population LBM for compressible ideal gas [19]. Because of its simplicity, there are many directions in which the present model can be further developed. Since we here used the simplest local BGK relaxation, the usual restriction on the Prandtl number can be removed by applying Shakhov [43] or quasiequilibrium [19] kinetic models. The proposed formalism for implementing complex flow physics in a lattice Boltzmann setting is based on the availability of the quadratures (2) and (3), and it can be extended to other kinetic theories, such as the revised Enskog theory [44] or the variational kinetic theory [37]. Moreover, our approach can be applied to nonideal multicomponent fluids, following the recent LBM for compressible ideal gas mixtures [20] and extending the latter with the nonlocal force (2) and the energy rate of change (3). The method of shifted equilibrium can also be applied to other cases of model equations of state that are not related to particles. For example, this method was used in [45] to produce a consistent lattice Boltzmann model for shallow water systems, corresponding to a quadratic pressure function, $P \sim \rho^2$. In general, the present new approach opens the door to the development of efficient kinetic models beyond the standard lattice Boltzmann model.

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Data availability—The data that support the findings of this article are openly available [39].

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