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Theory of the lattice Boltzmann method: Two-fluid model for binary mixtures

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A two-fluid lattice Boltzmann model for binary mixtures is developed. The model is derived formally from kinetic theory by discretizing two-fluid Boltzmann equations in which mutual collisions and self-collisions are treated independently. In the resulting lattice Boltzmann model, viscosity and diffusion coefficients can be varied independently by a suitable choice of mutual- and self-collision relaxation-time scales. Further, the proposed model can simulate miscible and immiscible fluids by changing the sign of the mutual-collision term. This is in contrast to most existing single-fluid lattice Boltzmann models that employ a single-relaxation-time scale and hence are restricted to unity Prandtl and Schmidt numbers. The extension of binary mixing model to multiscalar mixing is quite straightforward.

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I. INTRODUCTION

In many practical flows involving pollutant dispersion, chemical processing, and combustor mixing and reaction, mass and momentum transport within multispecies fluids plays an important role. For these flows, it can be difficult to construct continuum-based models from first principles. Further, these flows typically involve complex geometry and/or multiple phases making computation with continuum-based models quite complicated. Therefore, for these flows, there is a growing interest in using the lattice Boltzmann equation [1–7].

In general, for computing fluid flow of any type, the lattice Boltzmann equation (LBE) [1–7] offers several computational advantages over continuum-based methods while retaining the flow physics intact. Although the origins of the modern LBE can be traced back to lattice-gas automata [8–10], the new LBE models are free of some well-known defects associated with their predecessors. Recent works have unequivocally established that the lattice Boltzmann equation is in fact connected to kinetic theory [4–6] and completely consistent with the fundamental conservation principles governing fluid flow [11–13]. In these papers, *a priori* derivation of the lattice Boltzmann equation from the parent continuous Boltzmann equation is developed [4–6]. The Navier-Stokes equation also has its basis in the Boltzmann equation—the former can be derived from the latter through the Chapman-Enskog analysis [14]. That very same Chapman-Enskog analysis can be used to show that the lattice Boltzmann methodology can be applied to solve any conservation law of the continuous Boltzmann equation including the Navier-Stokes equations. It has also been proved that the lattice Boltzmann equation tantamounts to an explicit finite difference scheme of the Navier-Stokes equations with second-order spatial accuracy and first-order temporal accuracy with a nonzero compressibility [11–13]. The present day lattice Boltzmann equation, with its high-fidelity physics and computation-efficient formulation, is a viable alternative to the continuum methods for simulating fluid flows. In fact, it can be argued that for many complex problems involving multifluid phenomena, the physics can be more naturally captured by the Boltzmann-equation based methods rather than Navier-Stokes equation based methods. Recently LBE method has been extended to multiphase flows [15–18] and multicomponent flows [19–31], flows through porous media [32,33], and particulate suspensions in fluids [34–37]. Most existing LBE models for multicomponent fluids [19–27] tend to be somewhat heuristic and make the single-fluid assumption. The single-relaxation-time or Bhatnagar-Gross-Krook (BGK) approximation [38] is used in most existing models [21–27] restricting applicability to unity Prandtl and Schmidt numbers.

A rigorous mathematical development of multifluid lattice Boltzmann equation for multicomponent fluids is still in its infancy and such is the object of the present work. As a first step, in this work we develop a two-fluid lattice Boltzmann model which is based on kinetic theory for binary mixtures. Such a model would be capable of (i) simulating arbitrary Schmidt and Prandtl numbers, and (ii) accurately modeling the interaction between miscible and immiscible fluids. We follow a general approach within the framework of kinetic theory for developing the lattice Boltzmann models for multifluid mixtures. This work is a part of our ongoing effort to set the lattice Boltzmann equation on a more rigorous theoretical foundation and extend its use to more complex flows. We derive a discretized version of the continuum Boltzmann equations for binary mixtures. The extension of this methodology to multifluid mixtures is relatively straightforward.

Kinetic theory of gas mixtures has received much attention in literature [14,39–51]. Many of the kinetic models for gas mixtures are based on the linearized Boltzmann equation [38,52,53], especially the single-relaxation-time model due to Bhatnagar, Gross, and Krook—the celebrated BGK model [38]. The kinetic-theory mixtures model employed in this work was proposed by Sirrovich [43], which is also linear in nature.

This paper is a detailed follow-up to our previous work

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published as a Rapid Communication in Physical Review E [31] and it is organized as follows. Section II provides a brief review for some of the existing kinetic models of mixtures that form the theoretical basis of the present work. Section III contains the derivation of the lattice Boltzmann model for binary mixtures from the corresponding continuous Boltzmann equations. In Sec. IV, the hydrodynamic equations of the lattice Boltzmann model are determined. Section V contains the derivation of the diffusion force and the mutual diffusion coefficient in the lattice Boltzmann model, and the diffusion-advection equations for the mass and molar concentrations of the system. Section VI discusses the short and long time behaviors of the model. Section VII concludes the paper with a summary of the present work and possible directions of future work. The three appendixes contain the details of: (a) the iterative procedure to solve the Boltzmann equation; (b) the discretized equilibrium distribution function; and, (c) the Chapman-Enskog analysis of the lattice Boltzmann model for binary mixtures.

II. KINETIC THEORY OF GAS MIXTURES

Following a procedure similar to the derivation of the Boltzmann equation for a pure system of single species, one can derive N simultaneous equations for a system of N species by reducing the appropriate Liouville equation. For the sake of simplicity without loss of generality, we shall only discuss the Boltzmann equations for a binary system here. The simultaneous Boltzmann equations for a binary system are

$$\partial_t f^A + \boldsymbol{\xi} \cdot \boldsymbol{\nabla} f^A + \boldsymbol{a}_A \cdot \boldsymbol{\nabla}_\xi f^A = Q^{AA} + Q^{AB}, \tag{1a}$$

$$\partial_t f^B + \boldsymbol{\xi} \cdot \boldsymbol{\nabla} f^B + \boldsymbol{a}_B \cdot \boldsymbol{\nabla}_\xi f^B = Q^{BA} + Q^{BB}, \tag{1b}$$

where $Q^{AB} = Q^{BA}$ is the collision term due to the interaction among two different species A and B. Obviously, for an N-component system, there will be N such equations, each containing N-collision terms on the right-hand side. In general, the collision term is

$$Q^{AB} = \int d\boldsymbol{\xi}_B d\Omega\, \sigma_{AB} \| \boldsymbol{\xi}_B - \boldsymbol{\xi}_A \| [f'^A f'^B - f^A f^B], \tag{2}$$

where σ_{AB} is the differential cross section of A-B collision, and Ω is the solid angle, and $f'^A (f'^B)$ and f^A (f^B) denote the post-collision and precollision states of the particle A (B), respectively. Obviously, the equations for a system of multiple species are much more formidable to analyze than the comparable Boltzmann equation for a pure system of single species. The first modeling objective is to find a suitable approximation for the collision term given by Eq. (2) that would substantially simplify computation without compromising the essential physics.

The justified approximation of the collision terms must rely on our clear understanding of the underlying physical system. In a system of multiple species, there are a number of competing equilibration processes occurring simultaneously. The approach to equilibrium in the system can be roughly divided into two stages. First, each individual species equilibrates within itself so that its local distribution function approaches a local Maxwellian distribution. This process of individual equilibration is also referred to as Maxwellization. Second, the entire system equilibrates so that the velocity and temperature differences among different species vanishes eventually. There are many different time scales in the equilibrating process of a multicomponent system. In addition, the Maxwellization itself can take place in many scenarios depending on the molecular weights and mass fractions of the participating species. Consider two mixtures, each consisting of a light and heavy gas. In mixture 1, the total mass of each species is the same, implying smaller number density for the heavier gas. In mixture 2, the number densities of the two species is the same, implying the mass density (or mass fraction) of the heavier species is larger. In mixture 1, the Maxwellization of light species is mostly due to self-collision, whereas the equilibration of the heavier species is predominantly due to cross collisions. This is due to the fact that the number of molecules of heavy species available for collisions is small. In mixture 2, where the number densities of the two species are comparable, Maxwellization of both species involves self-collision and cross collision. When the process of Maxwellization is complete, the stress of the corresponding species becomes isotropic, or equivalently the heat conduction relaxes. Therefore, the time scale on which the stress becomes isotropic or the heat conduction relaxes is a suitable measure of Maxwellization.

The equilibration among different species can also take place in several different manners. Velocity and temperature differences may equilibrate on the same temporal scale (as in mixture 1 above) or on vastly different scales (as in mixture 2). In addition, these equilibrating processes need not to occur sequentially but also concurrently with the Maxwellization.

There is a significant amount of literature on gas mixtures within the framework of kinetic theory [14,39–51]. In the Chapman-Enskog analysis for a simple gas, one assumes a clear separation of scales in space and time, that is, to distinguish the spatial and temporal scales, which are much larger than the mean free path and mean free time. An analogy for a mixture becomes difficult because of multiplicity of length scales. In the classic work of Chapman and Cowling [14], the full Boltzmann equations (with integral collision terms) for a binary mixture are analyzed under the assumptions that all scales are roughly of the same order, or equivalently, that the phenomenon to be examined is smooth with respect to all collisional scales. Determination of the various transport coefficients—viscosities, diffusivities, thermal diffusivities and conductivity—was the main objective of that work. However, no attempt was made to describe the dynamics of the evolution.

Direct analysis or computation of the Boltzmann equation is not generally feasible. This is due to the difficulty involved in evaluating the complex integral collision operators. To make further progress one can follow one of two approaches. The first, Grad's moment method, is to obtain the nonnormal solutions of the Boltzmann equation (i.e., the solutions beyond the hydrodynamic or conserved variables) [54]. Closure modeling would then be required to express the unclosed moments in terms of the closed moments. And the second is to derive simplified model equations from the the Boltzmann equation, which are more manageable to solve. Many model equations are influenced by Maxwell's approach to solving the Boltzmann equation by making extensive use of the properties of the Maxwell molecule [55] and the linearized Boltzmann equation. The simplest model equations for a binary mixture is that by Gross and Krook [41], which is an extension of the single-relaxation-time model for a pure system—the celebrated BGK model [38].

With the BGK approximation [38,41], the collision integrals $Q^{\sigma\varsigma}$ $[\sigma,\varsigma\in(A,B)]$ can be approximated by following linearized collision terms:

$$J^{\sigma\sigma}=-\frac{1}{\lambda_{\sigma}}[f^{\sigma}-f^{\sigma(0)}], \tag{3a}$$

$$J^{\sigma\varsigma}=-\frac{1}{\lambda_{\sigma\varsigma}}[f^{\sigma}-f^{\sigma\varsigma(0)}], \tag{3b}$$

where $f^{\sigma(0)}$ and $f^{\sigma\varsigma(0)}$ are Maxwellians

$$f^{\sigma(0)}=\frac{n_{\sigma}}{(2\pi R_{\sigma}T_{\sigma})^{D/2}}e^{-(\boldsymbol{\xi}-\boldsymbol{u}_{\sigma})^{2}/(2R_{\sigma}T_{\sigma})}, \tag{4a}$$

$$f^{\sigma\varsigma(0)}=\frac{n_{\sigma}}{(2\pi R_{\sigma}T_{\sigma\varsigma})^{D/2}}e^{-(\boldsymbol{\xi}-\boldsymbol{u}_{\sigma\varsigma})^{2}/(2R_{\sigma}T_{\sigma\varsigma})}, \tag{4b}$$

where D is the spatial dimension, $R_{\sigma}=k_{B}/m_{\sigma}$ is the gas constant of the σ species, k_{B} is the Boltzmann constant and m_{σ} is the molecular mass of the σ species. There are three adjustable relaxation parameters in the collision terms: λ_{σ}, λ_{ς}, and $\lambda_{\sigma\varsigma}=(n_{\varsigma}/n_{\sigma})\lambda_{\varsigma\sigma}$. The first Maxwellian $f^{\sigma(0)}$ is characterized by the conserved variables of each individual species: the number density n_{σ}, the mass velocity $\boldsymbol{u}_{\sigma}$, and the temperature T_{σ}; while the second Maxwellian $f^{\sigma\varsigma(0)}$ and $f^{\varsigma\sigma(0)}$ is characterized by four adjustable parameters: $\boldsymbol{u}_{\sigma\varsigma}$, $\boldsymbol{u}_{\varsigma\sigma}$, $T_{\sigma\varsigma}$, and $T_{\varsigma\sigma}$. There are several considerations in determining these arbitrary parameters: simplicity of the resulting theory, accuracy of approximation, and ease of computation. Cross-collisional terms will be symmetric only if one takes $\boldsymbol{u}_{\sigma\varsigma}=\boldsymbol{u}_{\varsigma\sigma}=\boldsymbol{u}$ and $T_{\sigma\varsigma}=T_{\varsigma\sigma}=T$, where $\boldsymbol{u}$ and T are the velocity and temperature of the mixture. This is essential in preserving a similarity to irreversible thermodynamics, especially the Onsager relation [56]. On the other hand, fewer terms in the expansion of f^{σ} about $f^{\sigma\varsigma(0)}$ would be needed in many cases if one chooses $\boldsymbol{u}_{\sigma\varsigma}=\boldsymbol{u}_{\sigma}$ and $T_{\sigma\varsigma}=T_{\sigma}$, i.e., $f^{\sigma\varsigma(0)}=f^{\sigma(0)}$. One salient difference between using $\boldsymbol{u}$ and T of the mixture in the Maxwellian $f^{\sigma\varsigma(0)}$ as opposed to using $\boldsymbol{u}_{\sigma}$ and T_{σ} for the species is that the former choice leads to a single-fluid theory, i.e., a set of hydrodynamic equations for the mixture, while the latter leads to a two-fluid theory [43,49], i.e., two sets of hydrodynamic equations for the species. Obviously, in the cases where the properties between the two species are vastly different, the two-fluid theory is preferred [56].

The cross-collision term $J^{\sigma\varsigma}$ can be better approximated by expanding f^{σ} around the Maxwellian [43]

$$J^{\sigma\varsigma}=-\frac{f^{\sigma(0)}}{n_{\sigma}k_{B}T_{\sigma}}\bigg[\mu_{D}\boldsymbol{c}_{\sigma}\cdot(\boldsymbol{u}_{\sigma}-\boldsymbol{u}_{\varsigma})+\mu_{T}\frac{3}{2}\bigg(\frac{\boldsymbol{c}_{\sigma}^{2}}{2R_{\sigma}T_{\sigma}}-1\bigg)\times(T_{\sigma}-T_{\varsigma})-M_{\sigma\varsigma}\bigg(\frac{\boldsymbol{c}_{\sigma}^{2}}{2R_{\sigma}T_{\sigma}}-1\bigg)(\boldsymbol{u}_{\sigma}-\boldsymbol{u}_{\varsigma})^{2}\bigg], \tag{5}$$

where $\boldsymbol{c}_{\sigma}=(\boldsymbol{\xi}-\boldsymbol{u}_{\sigma})$ is the peculiar (or thermal) velocity of the σ species, and

$$M_{\sigma\varsigma}=\mu_{m}\frac{\rho_{\sigma}\rho_{\varsigma}}{\rho}\bigg[\frac{1}{\rho_{\sigma}}+\mu_{m}'\frac{(n_{\sigma}-n_{\varsigma})}{n_{\sigma}n_{\varsigma}(m_{\sigma}+m_{\varsigma})}\bigg], \tag{6}$$

and $\mu_{D},\mu_{T},\mu_{m},\mu_{m}'$ are positive and at most functions of density and temperature [43], the physical significances of these parameters are to be discussed next.

We now consider the following model equations for a binary mixture due to Sirovich [43]:

$$\partial_{t}f^{\sigma}+\boldsymbol{\xi}\cdot\boldsymbol{\nabla}f^{\sigma}+\boldsymbol{a}_{\sigma}\cdot\boldsymbol{\nabla}_{\xi}f^{\sigma}=J^{\sigma\sigma}+J^{\sigma\varsigma}, \tag{7}$$

where the self-collision term $J^{\sigma\sigma}$ is approximated with the BGK model of Eq. (3a), and the cross-collision term $J^{\sigma\varsigma}$ is given by Eq. (5). When the external force is not present $(\boldsymbol{a}_{\sigma}=\boldsymbol{0})$, and if $\mu_{D},\mu_{T},\mu_{m},\mu_{m}'$ are considered as constants (for the sake of simplicity) we can immediately obtain the following moment equations from the above equations:

$$\partial_{t}(\boldsymbol{u}_{\sigma}-\boldsymbol{u}_{\varsigma})=-\mu_{D}\bigg(\frac{1}{\rho_{\sigma}}+\frac{1}{\rho_{\varsigma}}\bigg)(\boldsymbol{u}_{\sigma}-\boldsymbol{u}_{\varsigma}), \tag{8a}$$

$$\partial_{t}(T_{\sigma}-T_{\varsigma})=-\mu_{T}\frac{1}{k_{B}}\bigg(\frac{1}{n_{\sigma}}+\frac{1}{n_{\varsigma}}\bigg)(T_{\sigma}-T_{\varsigma})+\frac{2}{3k_{B}}\bigg(\frac{M_{\sigma\varsigma}}{n_{\sigma}}-\frac{M_{\varsigma\sigma}}{n_{\varsigma}}\bigg)(\boldsymbol{u}_{\sigma}-\boldsymbol{u}_{\varsigma})^{2}. \tag{8b}$$

The above equations describe the exponential decay of the velocity and temperature differences for the two species, as discussed in the earlier remarks regarding the processes of Maxwellianization and equilibration in the mixture. The physical significances of the parameters μ_{D}, μ_{T}, μ_{m}, and μ_{m}' become apparent in the above equations—these parameters determine the relaxation rates in the Maxwellization processes.

Solving Eqs. (7) by means of iteration (cf. Ref. [43] or Appendix A), one first obtains

$$\boldsymbol{u}_{\sigma}=\boldsymbol{u}_{\varsigma}=\boldsymbol{u}, \tag{9a}$$

$$T_{\sigma}=T_{\varsigma}=T, \tag{9b}$$

$$f^{\sigma(0)}=f^{\sigma\varsigma(0)}(n_{\sigma},\boldsymbol{u},T)=\frac{n_{\sigma}}{(2\pi R_{\sigma}T)^{D/2}}\exp\bigg[-\frac{(\boldsymbol{\xi}-\boldsymbol{u})^{2}}{2R_{\sigma}T}\bigg]. \tag{9c}$$

Substituting the above results into the left-hand side of Eqs. (7), one has the following equation for the second-order solution of f^{σ} [43]:

$$f^{\sigma\varsigma(0)}(n_\sigma,\boldsymbol{u},T)\left[\frac{n}{n_\sigma}\boldsymbol{c}_\sigma\cdot\boldsymbol{d}_\sigma+\left(\frac{c_{\sigma i}c_{\sigma j}}{R_\sigma T}-\delta_{ij}\right)\cdot S_{ij}\right.$$
$$\left.+\left(\frac{c_\sigma^2}{2R_\sigma T}-\frac{5}{2}\right)\boldsymbol{c}_\sigma\cdot\boldsymbol{\nabla}\ln T\right]$$
$$=-\frac{1}{\lambda_\sigma}[f^\sigma-f^{\sigma(0)}]-\frac{f^{\sigma(0)}}{n_\sigma k_B T_\sigma}\left[\mu_D\boldsymbol{c}_\sigma\cdot(\boldsymbol{u}_\sigma-\boldsymbol{u}_\varsigma)\right.$$
$$+\mu_T\left(\frac{c_\sigma^2}{3R_\sigma T_\sigma}-1\right)(T_\sigma-T_\varsigma)$$
$$\left.-M_{\sigma\varsigma}\left(\frac{c_\sigma^2}{3R_\sigma T_\sigma}-1\right)(\boldsymbol{u}_\sigma-\boldsymbol{u}_\varsigma)^2\right], \tag{10}$$

where $\boldsymbol{c}_\sigma=(\boldsymbol{\xi}-\boldsymbol{u}_\sigma)$, and the diffusion force $\boldsymbol{d}_\sigma$ and the rate-of-shear tensor S_{ij} are given by

$$\boldsymbol{d}_\sigma=\boldsymbol{\nabla}\left(\frac{n_\sigma}{n}\right)+\frac{n_\sigma n_\varsigma}{n\rho}(m_\varsigma-m_\sigma)\boldsymbol{\nabla}\ln p-\frac{\rho_\sigma\rho_\varsigma}{\rho p}(\boldsymbol{a}_\sigma-\boldsymbol{a}_\varsigma),$$
$$=-\boldsymbol{d}_\varsigma, \tag{11a}$$

$$S_{ij}=\frac{1}{2}(\partial_i u_j+\partial_j u_i-\tfrac{2}{3}\boldsymbol{\nabla}\cdot\boldsymbol{u}\delta_{ij}), \tag{11b}$$

and $p=nk_BT$ is the total pressure of the mixture. From the above solution of f^σ, one can compute for the relative velocity $(\boldsymbol{u}_\sigma-\boldsymbol{u}_\varsigma)$, the temperature difference $(T_\sigma-T_\varsigma)$, the (traceless) stress tensor p_{ij}, and the heat flux $\boldsymbol{q}$ [43]:

$$(\boldsymbol{u}_\sigma-\boldsymbol{u}_\varsigma)=-\frac{nk_BT}{\mu_D}\boldsymbol{d}'_\sigma=-\frac{n^2}{n_\sigma n_\varsigma}D_{\sigma\varsigma}\boldsymbol{d}'_\sigma, \tag{12a}$$

$$(T_\sigma-T_\varsigma)=\frac{\mu_D\mu'_m}{\mu_T}\,\frac{m_\sigma m_\varsigma(n_\sigma-n_\varsigma)}{\rho(m_\sigma+m_\varsigma)}(\boldsymbol{u}_\sigma-\boldsymbol{u}_\varsigma)^2, \tag{12b}$$

$$p_{ij}=-2k_BT\left(\frac{n_\sigma}{\lambda_\sigma}+\frac{n_\varsigma}{\lambda_\varsigma}\right)S_{ij}-\frac{2\mu_D}{\rho}\left(\frac{\rho_\varsigma}{\lambda_\sigma}+\frac{\rho_\sigma}{\lambda_\varsigma}\right)$$
$$\times[(\boldsymbol{u}_\sigma-\boldsymbol{u}_\varsigma)_i(\boldsymbol{u}_\sigma-\boldsymbol{u}_\varsigma)_j-\tfrac{1}{3}(\boldsymbol{u}_\sigma-\boldsymbol{u}_\varsigma)^2\delta_{ij}], \tag{12c}$$

$$\boldsymbol{q}=\frac{5}{2}k_BT[n_\sigma(\boldsymbol{u}_\sigma-\boldsymbol{u})+n_\varsigma(\boldsymbol{u}_\varsigma-\boldsymbol{u})]$$
$$-\frac{5}{2}k_BT\left(\frac{\rho_\sigma}{\lambda_\sigma}+\frac{\rho_\varsigma}{\lambda_\varsigma}\right)k_B\boldsymbol{\nabla}T, \tag{12d}$$

where the diffusion force

$$\boldsymbol{d}'_\sigma=\boldsymbol{\nabla}\left(\frac{n_\sigma}{n}\right)+\frac{T_\sigma}{T}\,\frac{n_\sigma n_\varsigma}{n\rho}(m_\varsigma-m_\sigma)\boldsymbol{\nabla}\ln p-\frac{\rho_\sigma\rho_\varsigma}{\rho p}(\boldsymbol{a}_\sigma-\boldsymbol{a}_\varsigma), \tag{13}$$

and the binary diffusion coefficient [53] of the model

$$D_{\sigma\varsigma}=\frac{n_\sigma n_\varsigma k_BT}{n\mu_D}. \tag{14}$$

The self-diffusion coefficient $D_{\sigma\sigma}$ is a special case of the above formula when $n=n_\varsigma=n_\sigma$. The viscosity ν and thermal conductivity κ can also be read from the above formulas for p_{ij} and $\boldsymbol{q}$ as the following:

$$\nu=k_BT\left(\frac{n_\sigma}{\lambda_\sigma}+\frac{n_\varsigma}{\lambda_\varsigma}\right), \tag{15a}$$

$$\kappa=\frac{5}{2}k_B^2T\left(\frac{\rho_\sigma}{\lambda_\sigma}+\frac{\rho_\varsigma}{\lambda_\varsigma}\right). \tag{15b}$$

The above transport coefficients are determined by the parameters λ_σ and μ_D, μ_T, μ_m, and μ'_m: λ_σ determines the viscosity and the thermal conductivity of the σ species and the combination of λ_σ's determines that of the mixture; μ_D determines the diffusion coefficients in the model; and $\mu_D\mu'_m/\mu_T$ determines the diffusion of the temperature difference due to velocity difference.

Two salient features of the model described by Eqs. (7) should be addressed. First, the cross-collision term $J^{\sigma\varsigma}$ of Eq. (5) is exact for the Maxwell molecules obeying the inverse fifth-power interaction potential. Equation (7), therefore, can be considered to be a model for the Maxwell gas [43]. One immediate consequence of this approximation is that the diffusion force of Eq. (12a) does not contain a thermal diffusion term, as it should. Second, the BGK approximation of the self-collision term $J^{\sigma\sigma}$ of Eq. (3a) imposes the limitation of a unity Prandtl number. However, both these limitations of the model can be overcome by using the linearized Boltzmann equation [57–59] with multiple relaxation times and a nonlinear approximation of the collision terms [43,49,60].

III. THE LATTICE BOLTZMANN MODEL FOR BINARY MIXTURE

We shall construct a lattice Boltzmann model for binary mixtures based on the model given by Eq. (7). In the present work we only consider the isothermal case such that $T_\sigma=T_\varsigma=T_{\sigma\varsigma}=T=\text{const}$. Consequently, we can also ignore the terms related to thermal effects in $J^{\sigma\varsigma}$ of Eq. (5) by setting $\mu_T=\mu_m=\mu'_m=0$, i.e.,

$$J^{\sigma\varsigma}=-\frac{1}{\tau_D}\,\frac{\rho_\varsigma}{\rho}\,\frac{f^{\sigma(0)}}{R_\sigma T}(\boldsymbol{\xi}-\boldsymbol{u})\cdot(\boldsymbol{u}_\sigma-\boldsymbol{u}_\varsigma), \tag{16}$$

where the equilibrium function $f^{\sigma(0)}$ for the σ species is chosen to be the Maxwellian equilibrium distribution depending on the mass velocity of the σ species $\boldsymbol{u}_\sigma$ as the following:

$$f^{\sigma(0)}=\frac{\rho_\sigma}{(2\pi R_\sigma T)^{D/2}}\exp\left[-\frac{(\boldsymbol{\xi}-\boldsymbol{u}_\sigma)^2}{2R_\sigma T}\right]. \tag{17}$$

Note that from hereafter $f^{\sigma(0)}$ and f^σ are the single particle mass density distribution functions, as opposed to the single particle number density distribution functions. We can derive the lattice Boltzmann equation by discretizing the model equation (7), following the procedure described in Refs. [4,5]:

$$f^{\sigma}_{\alpha}(\boldsymbol{x}_i+\boldsymbol{e}_{\alpha}\delta_t, t+\delta_t)-f^{\sigma}_{\alpha}(\boldsymbol{x}_i, t)=J^{\sigma\sigma}_{\alpha}+J^{\sigma\varsigma}_{\alpha}-F^{\sigma}_{\alpha}\delta_t, \tag{18}$$

where the self-collision term $J^{\sigma\sigma}_{\alpha}$, the cross-collision term $J^{\sigma\varsigma}_{\alpha}$, and the forcing term F_{α} are given by

$$J^{\sigma\sigma}_{\alpha}=-\frac{1}{\tau_{\sigma}}[f^{\sigma}_{\alpha}-f^{\sigma(0)}_{\alpha}], \tag{19a}$$

$$J^{\sigma\varsigma}_{\alpha}=-\frac{1}{\tau_D}\frac{\rho_{\varsigma}}{\rho}\frac{f^{\sigma(\mathrm{eq})}}{R_{\sigma}T}(\boldsymbol{e}_{\alpha}-\boldsymbol{u})\cdot(\boldsymbol{u}_{\sigma}-\boldsymbol{u}_{\varsigma}), \tag{19b}$$

$$F^{\sigma}_{\alpha}=-w_{\alpha}\rho_{\sigma}\frac{\boldsymbol{e}_{\alpha}\cdot\boldsymbol{a}_{\sigma}}{R_{\sigma}T}, \tag{19c}$$

where ρ_{σ} and ρ_{ς}, and $\boldsymbol{u}_{\sigma}$ and $\boldsymbol{u}_{\varsigma}$ are the mass densities and flow velocities for species σ and ς, they are the moments of the distribution functions:

$$\rho_{\sigma}=\sum_{\alpha} f^{\sigma}_{\alpha}=\sum_{\alpha} f^{\sigma(0)}_{\alpha}, \tag{20a}$$

$$\rho_{\sigma}\boldsymbol{u}_{\sigma}=\sum_{\alpha} f^{\sigma}_{\alpha}\boldsymbol{e}_{\alpha}=\sum_{\alpha} f^{\sigma(0)}_{\alpha}\boldsymbol{e}_{\alpha}, \tag{20b}$$

and ρ and $\boldsymbol{u}$ are respectively, the mass density and the barycentric velocity of the mixture:

$$\rho=\rho_{\sigma}+\rho_{\varsigma}, \tag{21a}$$

$$\rho\boldsymbol{u}=\rho_{\sigma}\boldsymbol{u}_{\sigma}+\rho_{\varsigma}\boldsymbol{u}_{\varsigma}. \tag{21b}$$

The collision terms $J^{\sigma\sigma}_{\alpha}$ and $J^{\sigma\varsigma}_{\alpha}$ are constructed in such a way to respect the mass and momentum conservation laws. (The derivation of the forcing term F^{σ}_{α} is given in Refs. [17,18].)

The equilibrium distribution function $f^{\sigma(0)}_{\alpha}$ has the following form in general (cf. Appendix B):

$$f^{\sigma(0)}_{\alpha}=\left[1+\frac{1}{R_{\sigma}T}(\boldsymbol{e}_{\alpha}-\boldsymbol{u})\cdot(\boldsymbol{u}_{\sigma}-\boldsymbol{u})\right]f^{\sigma(\mathrm{eq})}_{\alpha}, \tag{22a}$$

$$f^{\sigma(\mathrm{eq})}_{\alpha}=w_{\alpha}\rho_{\sigma}\left[1+\frac{(\boldsymbol{e}_{\alpha}-\boldsymbol{u})\cdot\boldsymbol{u}}{R_{\sigma}T}+\frac{(\boldsymbol{e}_{\alpha}\cdot\boldsymbol{u})^2}{2(R_{\sigma}T)^2}\right], \tag{22b}$$

where coefficients $\{w_{\alpha}\}$ depend on the discrete velocity set $\{\boldsymbol{e}_{\alpha}\}$. For the sake of concreteness and simplicity without losing generality, we shall restrict ourselves to a nine-velocity model on a two-dimensional square lattice (D2Q9 model). In this case,

$$w_{\alpha}=\begin{cases} 4/9, & \alpha=0\\ 1/9, & \alpha=1-4\\ 1/36, & \alpha=5-8. \end{cases} \tag{23}$$

IV. HYDRODYNAMICS

The left-hand side of Eq. (18) can be expanded in a Taylor series in δ_t (up to second order in δ_t) and the equation can be rewritten as

$$\delta_t D_{\alpha} f^{\sigma}_{\alpha}+\frac{1}{2}\delta_t^2 D^2_{\alpha} f^{\sigma}_{\alpha}=J^{\sigma\sigma}_{\alpha}+J^{\sigma\varsigma}_{\alpha}-F^{\sigma}_{\alpha}\delta_t, \tag{24}$$

where $D_{\alpha}=\partial_t+\boldsymbol{e}_{\alpha}\cdot\boldsymbol{\nabla}$. Obviously,

$$\sum_{\alpha} J^{\sigma\sigma}_{\alpha}=\sum_{\alpha} J^{\varsigma\sigma}_{\alpha}=\sum_{\alpha} F^{\sigma}_{\alpha}=0, \tag{25a}$$

$$\sum_{\alpha} J^{\sigma\sigma}_{\alpha}\boldsymbol{e}_{\alpha}=0, \tag{25b}$$

$$\sum_{\alpha} J^{\varsigma\sigma}_{\alpha}\boldsymbol{e}_{\alpha}=-\frac{1}{\tau_D}\frac{\rho_{\sigma}\rho_{\varsigma}}{\rho}(\boldsymbol{u}_{\sigma}-\boldsymbol{u}_{\varsigma}), \tag{25c}$$

$$\sum_{\alpha} F^{\sigma}_{\alpha}\boldsymbol{e}_{\alpha}=-\rho_{\sigma}\boldsymbol{a}_{\sigma}. \tag{25d}$$

By means of the Chapman-Enskog analysis (multiple-scale expansion), we can derive the hydrodynamic equations for the mixture from Eq. (24) (see details in Appendix C).

The mass conservation laws for each species and the mixture can be derived immediately from Eq. (24):

$$\partial_t\rho_{\sigma}+\boldsymbol{\nabla}\cdot(\rho_{\sigma}\boldsymbol{u}_{\sigma})=\frac{1}{2}\boldsymbol{\nabla}\cdot\left[\frac{\rho_{\sigma}\rho_{\varsigma}}{\tau_D\rho}(\boldsymbol{u}_{\sigma}-\boldsymbol{u}_{\varsigma})\right], \tag{26}$$

$$\partial_t\rho+\boldsymbol{\nabla}\cdot(\rho\boldsymbol{u})=0. \tag{27}$$

Note that the mass conservation does not hold for each individual species at the Navier-Stokes level, although it does at the Euler level. However, the mass conservation law does apply to the mixture as a whole. The right-hand side of Eq. (26) reflects the mass flux due to diffusion.

We can also derive the Euler equation for each species:

$$\rho_{\sigma}\partial_{t_0}\boldsymbol{u}_{\sigma}+\rho_{\sigma}\boldsymbol{u}_{\sigma}\cdot\boldsymbol{\nabla}\boldsymbol{u}_{\sigma}=-\boldsymbol{\nabla}p_{\sigma}+\rho_{\sigma}\boldsymbol{a}_{\sigma}-\frac{1}{\tau_D\delta_t}\frac{\rho_{\sigma}\rho_{\varsigma}}{\rho}\times(\boldsymbol{u}_{\sigma}-\boldsymbol{u}_{\varsigma}), \tag{28}$$

where $p_{\sigma}=n_{\sigma}k_BT=\rho_{\sigma}R_{\sigma}T$ is the partial pressure of the σ species, and the Navier-Stokes equation:

$$\rho_{\sigma}\partial_t\boldsymbol{u}_{\sigma}+\rho_{\sigma}\boldsymbol{u}_{\sigma}\cdot\boldsymbol{\nabla}\boldsymbol{u}_{\sigma}=-\boldsymbol{\nabla}p_{\sigma}+\rho_{\sigma}\nu_{\sigma}\nabla^2\boldsymbol{u}_{\sigma}+\rho_{\sigma}\boldsymbol{a}_{\sigma}-\frac{1}{\tau_D\delta_t}\frac{\rho_{\sigma}\rho_{\varsigma}}{\rho}(\boldsymbol{u}_{\sigma}-\boldsymbol{u}_{\varsigma}), \tag{29}$$

where the viscosity of the σ species is

$$\nu_{\sigma}=R_{\sigma}T(\tau_{\sigma}-\tfrac{1}{2})\delta_t. \tag{30}$$

Equation (29) is consistent with the results in Ref. [49].

V. DIFFUSION IN ISOTHERMAL MIXTURES

The difference between the two Navier-Stokes equations for individual species (σ and ς) leads to the following equation:

$$\frac{1}{\tau_D\delta_t}(\boldsymbol{u}_\sigma-\boldsymbol{u}_\varsigma)=-\frac{\rho p}{\rho_\sigma\rho_\varsigma}\boldsymbol{d}_\sigma-\{\partial_t\delta\boldsymbol{u}+\boldsymbol{\nabla}\cdot(\overline{\boldsymbol{u}}\delta\boldsymbol{u}) +\nabla^2(\nu_\sigma\boldsymbol{u}_\sigma-\nu_\varsigma\boldsymbol{u}_\varsigma)\}, \tag{31}$$

where $\delta\boldsymbol{u}=(\boldsymbol{u}_\sigma-\boldsymbol{u}_\varsigma)$, $\overline{\boldsymbol{u}}=\frac{1}{2}(\boldsymbol{u}_\sigma+\boldsymbol{u}_\varsigma)$, $\boldsymbol{\nabla}\cdot(\overline{\boldsymbol{u}}\delta\boldsymbol{u})=\overline{\boldsymbol{u}}\cdot\boldsymbol{\nabla}\delta\boldsymbol{u}+\delta\boldsymbol{u}\cdot\boldsymbol{\nabla}\overline{\boldsymbol{u}}$, and the diffusion force

$$\begin{aligned}\boldsymbol{d}_\sigma&=\frac{\rho_\sigma\rho_\varsigma}{\rho p}\left[\left(\frac{1}{\rho_\sigma}\boldsymbol{\nabla}p_\sigma-\frac{1}{\rho_\varsigma}\boldsymbol{\nabla}p_\varsigma\right)-(\boldsymbol{a}_\sigma-\boldsymbol{a}_\varsigma)\right]\\ &=\boldsymbol{\nabla}\left(\frac{n_\sigma}{n}\right)+\left(\frac{n_\sigma}{n}-\frac{\rho_\sigma}{\rho}\right)\boldsymbol{\nabla}\ln p+\frac{\rho_\sigma\rho_\varsigma}{\rho p}(\boldsymbol{a}_\varsigma-\boldsymbol{a}_\sigma)\\ &=\boldsymbol{\nabla}\left(\frac{n_\sigma}{n}\right)+\frac{n_\sigma n_\varsigma}{n\rho}(m_\varsigma-m_\sigma)\boldsymbol{\nabla}\ln p+\frac{\rho_\sigma\rho_\varsigma}{\rho p}(\boldsymbol{a}_\varsigma-\boldsymbol{a}_\sigma)\\ &=-\boldsymbol{d}_\varsigma,\end{aligned} \tag{32}$$

where $p=nk_BT$ is the total pressure of the mixture, and the total number density n is

$$n=n_\sigma+n_\varsigma=\frac{\rho_\sigma}{m_\sigma}+\frac{\rho_\varsigma}{m_\varsigma}. \tag{33}$$

The diffusion force includes the effects due to the molar concentration gradient $\boldsymbol{\nabla}(n_\sigma/n)$, the total pressure gradient and the particle mass difference $(m_\sigma-m_\varsigma)\boldsymbol{\nabla}\ln p$, and the external force $(\boldsymbol{a}_\sigma-\boldsymbol{a}_\varsigma)$.

It has already been assumed in the derivation of the two-fluid equations that derivatives are slowly varying on the time scale of Maxwellization [49]. Therefore, the terms inside the curly brackets Eq. (31) can be neglected in the diffusion time scale. Thus, to the leading order, we have

$$(\boldsymbol{u}_\sigma-\boldsymbol{u}_\varsigma)=-\tau_D\delta_t\frac{\rho p}{\rho_\sigma\rho_\varsigma}\boldsymbol{d}_\sigma. \tag{34}$$

Also, by definition [53], [Eqs. (6.5–7a)], we have

$$(\boldsymbol{u}_\sigma-\boldsymbol{u}_\varsigma)=-\frac{n^2}{n_\sigma n_\varsigma}D_{\sigma\varsigma}\boldsymbol{d}_\sigma, \tag{35}$$

thus the mutual diffusion coefficient in the mixture is

$$D_{\sigma\varsigma}=\frac{\rho k_BT}{nm_\sigma m_\varsigma}\tau_D\delta_t. \tag{36}$$

Note that the difference between the above formula and Eq. (14) is due to the difference of a factor $\rho_\sigma\rho_\varsigma/\rho$ between $J_\alpha^{\sigma\varsigma}$ of Eq. (19b) and $J^{\sigma\varsigma}$ of Eq. (5), i.e.,

$$\tau_D\delta_t=\frac{\rho_\sigma\rho_\varsigma}{\rho}\frac{1}{\mu_D}.$$

The mass flux of the σ species, by definition, is

$$\boldsymbol{j}_\sigma=\rho_\sigma(\boldsymbol{u}_\sigma-\boldsymbol{u})=-\tau_D\delta_tp\boldsymbol{d}_\sigma,$$

where we have used the identity that $\rho(\boldsymbol{u}_\sigma-\boldsymbol{u})=\rho_\varsigma(\boldsymbol{u}_\sigma-\boldsymbol{u}_\varsigma)$. The continuity equation (26) for the σ species can be written as

$$D_t\rho_\sigma+\rho_\sigma\boldsymbol{\nabla}\cdot\boldsymbol{u}+\boldsymbol{\nabla}\cdot\boldsymbol{j}_\sigma=-\frac{1}{2}\delta_t\boldsymbol{\nabla}\cdot(p\boldsymbol{d}_\sigma), \tag{37}$$

where $D_t=(\partial_t+\boldsymbol{u}\cdot\boldsymbol{\nabla})$. By assuming the incompressibility of the fluid (i.e., $\boldsymbol{\nabla}\cdot\boldsymbol{u}=0$), we obtain the following advection-diffusion equation for an isothermal mixture:

$$\partial_t\rho_\sigma+\boldsymbol{u}\cdot\boldsymbol{\nabla}\rho_\sigma=\boldsymbol{\nabla}\cdot\tau_D^*\delta_tp\boldsymbol{d}_\sigma,\quad \tau_D^*\equiv(\tau_D-\tfrac{1}{2}). \tag{38}$$

Similar to the Hénon correction for the viscosity [61], the diffusivity is modified by the second-order discrete effect:

$$D_{\sigma\varsigma}^*=\frac{\rho k_BT}{nm_\sigma m_\varsigma}\left(\tau_D-\frac{1}{2}\right)\delta_t=\frac{\rho k_BT}{nm_\sigma m_\varsigma}\tau_D^*\delta_t. \tag{39}$$

Obviously, the self-diffusion coefficient in the lattice Boltzmann model for the σ species is

$$D_{\sigma\sigma}^*=\frac{k_BT}{m_\sigma}\tau_D^*\delta_t. \tag{40}$$

The mass concentration ϕ and molar concentration φ (dimensionless order parameters) are defined as

$$\phi=\frac{(\rho_\sigma-\rho_\varsigma)}{(\rho_\sigma+\rho_\varsigma)},\qquad \varphi=\frac{(n_\sigma-n_\varsigma)}{(n_\sigma+n_\varsigma)}, \tag{41}$$

and they related to each other

$$\phi=\frac{(m_\sigma-m_\varsigma)+(m_\sigma+m_\varsigma)\varphi}{(m_\sigma+m_\varsigma)+(m_\sigma-m_\varsigma)\varphi}, \tag{42a}$$

$$\varphi=\frac{(m_\varsigma-m_\sigma)+(m_\varsigma+m_\sigma)\phi}{(m_\varsigma+m_\sigma)+(m_\varsigma-m_\sigma)\phi}. \tag{42b}$$

The diffusion force can be written in terms of ϕ and φ:

$$\boldsymbol{d}_\sigma=\frac{1}{2}\left[\boldsymbol{\nabla}\varphi+(\varphi-\phi)\boldsymbol{\nabla}\ln p+\frac{\rho(1-\phi^2)}{2p}(\boldsymbol{a}_\varsigma-\boldsymbol{a}_\sigma)\right].$$

The nonlinear diffusion-advection equations satisfied by ϕ and φ can be derived from Eq. (38):

$$\partial_t\phi+\boldsymbol{u}\cdot\boldsymbol{\nabla}\phi=\frac{1}{\rho}\boldsymbol{\nabla}\cdot(D_\phi^*\boldsymbol{\nabla}\phi+\boldsymbol{F}), \tag{43a}$$

$$\partial_t\varphi+\boldsymbol{u}\cdot\boldsymbol{\nabla}\varphi=\frac{A}{n}\boldsymbol{\nabla}\cdot(D_\varphi^*\boldsymbol{\nabla}\varphi+\boldsymbol{F}), \tag{43b}$$

where

$$D_\phi^*=\frac{p\tau_D^*\delta_t}{m_\sigma m_\varsigma}\left(\frac{\rho}{n}\right)^2=\frac{k_BT}{m_\sigma m_\varsigma}\frac{\rho^2}{n}\tau_D^*\delta_t, \tag{44a}$$

$$D^{*}_{\varphi} = p\,\tau^{*}_{D}\,\delta_t, \tag{44b}$$

$$\boldsymbol{F} = [(\varphi-\phi)\boldsymbol{\nabla} p + \tfrac{1}{2}\rho(1-\phi^2)(\boldsymbol{a}_\varsigma - \boldsymbol{a}_\sigma)]\,\tau^{*}_{D}\,\delta_t, \tag{44c}$$

$$A = \frac{1}{2}\left[\frac{(1-\varphi)}{m_\sigma} + \frac{(1+\varphi)}{m_\varsigma}\right]. \tag{44d}$$

Obviously, when $m_\sigma = m_\varsigma$, $\varphi = \phi$, and then Eq. (43a) and Eq. (43b) become identical.

VI. SHORT AND LONG TIME BEHAVIORS

As discussed in Sec. II, short and long time behaviors of a binary mixture involve different Maxwellization and equilibration processes. This is reflected in the macroscopic equations derived in preceding sections. In an initial stage (short time), the diffusion velocity $(\boldsymbol{u}_\sigma - \boldsymbol{u}_\varsigma)$ is significant, thus the system is described $2(D+1)$ equations, i.e., two sets of mass and momentum conservation laws for each species given by Eqs. (26) and (29). As the system equilibrates so that the diffusion velocity $(\boldsymbol{u}_\sigma - \boldsymbol{u}_\varsigma)$ is diminishing, the physics is then described by $(D+2)$ equations: the continuity equation of the mixture, Eq. (27), the Navier-Stokes equation for the barycentric velocity of the mixture $\boldsymbol{u}$, and the diffusion-advection equation for the mass [Eq. (43a)] or molar [Eq. (43b)] concentrations. Only in very late stage of equilibration, the concentration behaves more or less as a passive scalar.

The Navier-Stokes equation for $\boldsymbol{u}$ is

$$\rho\partial_t \boldsymbol{u} + \rho\boldsymbol{u}\cdot\boldsymbol{\nabla}\boldsymbol{u} = -\boldsymbol{\nabla} p + \boldsymbol{\nabla}\cdot\Pi + \rho\boldsymbol{a}, \tag{45}$$

where $p = k_B T(n_\sigma + n_\varsigma)$, $\rho\boldsymbol{a} = \rho_\sigma\boldsymbol{a}_\sigma + \rho_\varsigma\boldsymbol{a}_\varsigma$, and

$$\Pi = \sum_{\sigma} \rho_\sigma \nu_\sigma [(\boldsymbol{\nabla}\boldsymbol{u}_\sigma) + (\boldsymbol{\nabla}\boldsymbol{u}_\sigma)^\dagger]$$
$$\approx (\rho_\sigma\nu_\sigma + \rho_\varsigma\nu_\varsigma)[(\boldsymbol{\nabla}\boldsymbol{u}) + (\boldsymbol{\nabla}\boldsymbol{u})^\dagger]. \tag{46}$$

In the derivation of Eq. (45), we ignore two terms: one is $(\tau_D D^{*}_{\sigma\varsigma} n/R_\sigma T n_\sigma n_\varsigma)\boldsymbol{\nabla}(p\boldsymbol{d}_\sigma)^2$, and the other is in proportion of $\boldsymbol{u}(\boldsymbol{u}_\sigma - \boldsymbol{u}_\varsigma)$ due to $J^{\sigma\varsigma}_{\alpha}$ [cf. Eq. (C7)]. These terms are negligible when the mixture is more or less homogeneous. Also, when the diffusion velocity $(\boldsymbol{u}_\sigma - \boldsymbol{u}_\varsigma)$ vanishes, the approximation in Eq. (46) becomes exact.

Some remarks to place the present work in perspective are in order. Unlike most existing lattice Boltzmann models for binary mixtures [22–27], the mutual diffusion and self-diffusion coefficients of the present model are independent of the viscosity. (We note that an existing model proposed by Flekkøy [20] already has this feature.) The diffusion coefficients depend on the relaxation parameter τ_D and relevant physical properties of the mixture, such as the molecular masses of each species m_σ and m_ς, etc. The present model is therefore capable of incorporating more general physics. In addition, the present model can simulate both miscible and immiscible binary mixtures by changing the sign of $(\tau_D - 1/2)$, i.e., for positive $(\tau_D - 1/2)$, the mixture is miscible, and for negative $(\tau_D - 1/2)$, the mixture is immiscible. We have performed numerical simulations to verify that: (a) the diffusion coefficient $D^{*}_{\sigma\varsigma}$ depends on τ_D as given by Eq. (39) and it is independent of τ_σ and τ_ς; and (b) when $(\tau_D - 1/2) < 0$, spinodal decomposition or separation (antidiffusion) between different species in the mixture occurs. (The details of the numerical results will be reported elsewhere.)

VII. DISCUSSION AND CONCLUSIONS

We have constructed a lattice Boltzmann model for binary mixtures with several important features. All the modeling issues are addressed at the continuum level within the framework of extended kinetic theory. The lattice Boltzmann model is then directly derived from the continuous kinetic model equations using a formal discretization procedure. The lattice model thus inherits the sound physics and mathematical rigor incumbent in kinetic theory. This is in contrast to previous lattice Boltzmann models for mixtures [22–27], which are not directly based on the fundamental physics of continuum kinetic equations. These models rely on fictitious interactions [22,23] or heuristic free energies [24–27] to produce the requisite mixing. (Many defects of the free-energy models [24–27] are due to the incorrectly defined equilibria [18].) These nonphysical effects present a further problem since they are not easily amenable to mathematical analysis [17,18]. The heuristic elements of the previous lattice Boltzmann models [22–27] have been eliminated, resulting in a physically justifiable model that is simple to compute. Further, due to the close connection to kinetic theory, the derivation of the hydrodynamic equations associated with the lattice Boltzmann model is significantly simplified and rendered mathematically more rigorous. The derivation of the hydrodynamic equations from the previous lattice models are much less rigorous [22–27].

The second important feature of the present work is that the model is based upon a two-fluid theory of binary mixtures. The previous models [19–27,51], on the other hand, are derived from a seemingly simpler, but highly restrictive, one-fluid theory. In the single-fluid models with BGK approximation one is constrained to use the *ad hoc* "equilibrium velocity" [22,23,51]

$$\boldsymbol{u}^{(\mathrm{eq})} = \frac{(\tau_\varsigma\rho_\sigma\boldsymbol{u}_\sigma + \tau_\sigma\rho_\varsigma\boldsymbol{u}_\varsigma)}{(\tau_\varsigma\rho_\sigma + \tau_\sigma\rho_\varsigma)}$$

in the equilibrium distribution function $f^{\sigma(0)}_{\alpha}$ in order to satisfy the local conservation laws. As a result, the viscous relaxation process and the diffusion process are inseparable. The analysis of these models therefore becomes unnecessarily tedious and cumbersome [22,23]. The models with free energies [24–27] do not yield correct hydrodynamic equations [17,18], mostly due to the incorrectly defined equilibrium distribution functions used in these models [18,62]. Furthermore, single-fluid models cannot be applied to mixtures of species with vastly different properties. In the present two-fluid model, the diffusion behavior is decoupled from viscous relaxation. The diffusivity is determined by the parameter τ_D and the physical properties of the mixture. The model is capable of simulating either miscible or immiscible fluids by changing the sign of $(\tau_D - 1/2)$.

The proposed LBE model for binary mixtures simulates diffusion by considering a mutual interaction term leading to the diffusion velocity $(\boldsymbol{u}_\sigma - \boldsymbol{u}_\varsigma)$, which is directly related to the diffusion driving force in binary mixtures. The diffusion velocity $(\boldsymbol{u}_\sigma - \boldsymbol{u}_\varsigma)$ is of the first order in the density gradient $\boldsymbol{\nabla}\rho$. This suggests that the proposed model, however, does not include any higher-order terms of the density gradient. This in turn implies that the proposed model does not have a surface tension, which is related to the density gradient square $|\boldsymbol{\nabla}\rho|^2$. To include the effect due surface tension, the terms related to $|\boldsymbol{\nabla}\rho|^2$ must be explicitly considered.

To further improve the proposed lattice Boltzmann model for binary mixing, efforts are currently underway are: (a) development of a multiple-relaxation-time model for the self-collision term [57–59] that will significantly enhance the numerical stability of the scheme [58,59]; (b) consideration of models with surface tension; (c) inclusion of thermal diffusion effects which may be important in combustion applications; and, (d) development of a model for non-Maxwellian molecules. It should be noted that all the existing lattice Boltzmann models are only applicable to the Maxwell molecules as a direct consequence of the linearization of the Boltzmann equation. This limitation can be overcome by either using a different expansion of the distribution function, or by including the non-Maxwellian effects in the collision terms.

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APPENDIX A: ITERATIVE SOLUTION OF THE BOLTZMANN EQUATION

We present a short description of the iterative procedure [43] to solve the Boltzmann equation

$$D_t f = Q[f], \tag{A1}$$

where $D_t = \partial_t + \boldsymbol{\xi}\cdot\boldsymbol{\nabla}$ and $Q[f]$ is the collision term. The $(n+1)$th iteration solution $f^{(n+1)}$ is obtained from the nth iteration solution $f^{(n)}$ by solving the equation

$$Q[f^{(n+1)}] = D_t f^{(n)}, \tag{A2}$$

subject to the following conservation constraints

$$\int d\boldsymbol{\xi} f^{(n+1)} \begin{bmatrix} 1 \\ \boldsymbol{\xi} \\ \boldsymbol{\xi}^2 \end{bmatrix} = \int d\boldsymbol{\xi} f^{(0)} \begin{bmatrix} 1 \\ \boldsymbol{\xi} \\ \boldsymbol{\xi}^2 \end{bmatrix}, \tag{A3}$$

where $f^{(0)} = f^{(0)}(\rho^{(n+1)}, \boldsymbol{u}^{(n+1)}, T^{(n+1)})$ is the equilibrium distribution function that depends on the hydrodynamical moments ρ, $\boldsymbol{u}$, and T computed from $f^{(n+1)}$. Following the Chapman-Enskog procedure, the temporal derivatives are removed by using the conservation equations

$$\partial_t \rho^{(n+1)} = -\boldsymbol{\nabla}\cdot(\rho\boldsymbol{u}),$$

$$\partial_t \boldsymbol{u}^{(n+1)} = -\boldsymbol{u}\cdot\boldsymbol{\nabla}\boldsymbol{u} + \boldsymbol{a} - \frac{1}{\rho}\boldsymbol{\nabla}\cdot\mathsf{P}^{(n)},$$

$$\partial_t e^{(n+1)} = -\boldsymbol{\nabla}\cdot(e\boldsymbol{u}) + \rho\boldsymbol{a}\cdot\boldsymbol{u} - \boldsymbol{\nabla}\cdot\boldsymbol{q}^{(n)} + \mathsf{P}^{(n)}:\boldsymbol{\nabla}\boldsymbol{u},$$

where e is the internal energy and $e = (D/2)nk_BT$ for ideal gases, and P and $\boldsymbol{q}$ are the stress tenor and the heat flux, respectively. On right-hand side of the above equations, the hydrodynamical moments ρ, $\boldsymbol{u}$, and e are computed from the $(n+1)$th iteration solution $f^{(n+1)}$ but their superscript $(n+1)$ are omitted since they are conserved quantities. On the other hand, the stress tensor P and the heat flux $\boldsymbol{q}$, which are not conserved, are denoted with the superscript n as they are obtained from the solution of the previous iteration $f^{(n)}$.

In general, the iterative procedure described above is expected to converge more rapidly than a procedure of successive approximation, such as the Chapman-Enskog procedure for the Boltzmann equation. The reason is that in the iterative procedure the (nonlinear) integral equation must be solved at each step as given by Eq. (A2), whereas in the Chapman-Enskog procedure the integral equation is only solved at the initial step and at all approximation of higher order only the linearized integral equation is solved.

APPENDIX B: THE EQUILIBRIUM DISTRIBUTION FUNCTION

We consider the equilibrium distribution function of Eq. (17) for σ species based on its mass velocity $\boldsymbol{u}_\sigma$. The distribution function can be written in terms of the mixture velocity $\boldsymbol{u}$ as follows:

$$\begin{aligned} &\exp\left[-\frac{1}{2}\frac{(\boldsymbol{\xi}-\boldsymbol{u}_\sigma)^2}{R_\sigma T}\right] \\ &\quad = \exp\left[-\frac{1}{2}\frac{(\boldsymbol{\xi}-\boldsymbol{u})^2}{R_\sigma T}\right]\exp\left[\frac{(\boldsymbol{\xi}-\boldsymbol{u})\cdot(\boldsymbol{u}-\boldsymbol{u}_\sigma)}{R_\sigma T}\right] \\ &\qquad \times\exp\left[-\frac{1}{2}\frac{(\boldsymbol{u}-\boldsymbol{u}_\sigma)^2}{R_\sigma T}\right]. \end{aligned} \tag{B1}$$

With the procedure described in Refs. [4,5], $f_\alpha^{\sigma(\mathrm{eq})}$ of Eq. (22b) is the second-order Taylor expansion of the first exponential in the right-hand side of the above equality. Because the velocity difference $(\boldsymbol{u}-\boldsymbol{u}_\sigma)$ is considered to be a small quantity [49], the second exponential in the right-hand side of the above equality can be approximated by its first-order Taylor expansion:

$$\exp[\beta_\sigma(\boldsymbol{\xi}-\boldsymbol{u})\cdot(\boldsymbol{u}-\boldsymbol{u}_\sigma)] \approx 1 + \beta_\sigma(\boldsymbol{\xi}-\boldsymbol{u})\cdot(\boldsymbol{u}_\sigma-\boldsymbol{u}), \tag{B2}$$

where $\beta_\sigma = 1/R_\sigma T$. Since the velocity difference $(\boldsymbol{u}-\boldsymbol{u}_\sigma)$ is small, the third exponential in the right-hand side that depends on $(\boldsymbol{u}-\boldsymbol{u}_\sigma)^2$ can be neglected in a first-order approximation. It should be noted that this term affects the temperature difference [cf. Eqs. (8b) and (12b)], and must be included if thermal diffusion is important. For the case of isothermal mixing considered here the simplifications yield the final result of the equilibrium distribution function for $f_{\sigma}^{\sigma(0)}$ given by Eq. (22a) after the velocity space $\boldsymbol{\xi}$ is properly discretized [4,5].

APPENDIX C: CHAPMAN-ENSKOG ANALYSIS

In the Chapman-Enskog expansion, we introduce a small parameter ε (which is the Knudsen number), and

$$f_{\alpha}^{\sigma}=f_{\alpha}^{\sigma(0)}+\varepsilon f_{\alpha}^{\sigma(1)}+\cdots, \tag{C1a}$$

$$\partial_t=\varepsilon\partial_{t_0}+\varepsilon^2\partial_{t_1}+\cdots, \tag{C1b}$$

$$\boldsymbol{\nabla}\rightarrow\varepsilon\boldsymbol{\nabla}. \tag{C1c}$$

The left-hand side of Eq. (18) is expanded in a Taylor series in δ_t up to second-order first, and then is substituted with the expansions of ε:

$$\begin{aligned}f_{\alpha}^{\sigma}(\boldsymbol{x}_i+\boldsymbol{e}_{\alpha}\delta_t,t+\delta_t)-f_{\alpha}^{\sigma}(\boldsymbol{x}_i,t)&\\=\varepsilon\delta_t D_{\alpha}f_{\alpha}^{\sigma}+\tfrac{1}{2}\varepsilon^2\delta_t^2 D_{\alpha}D_{\alpha}f_{\alpha}^{\sigma}+\cdots&\\=\varepsilon\delta_t D_{\alpha}^{(0)}f_{\alpha}^{\sigma(0)}+\varepsilon^2\delta_t(\partial_{t_1}f_{\alpha}^{\sigma(0)}+D_{\alpha}^{(0)}f_{\alpha}^{\sigma(1)}&\\+\tfrac{1}{2}\delta_t D_{\alpha}^{(0)}D_{\alpha}^{(0)}f_{\alpha}^{\sigma(0)})+\cdots,&\end{aligned}$$

where Eqs. (C1) have been substituted, and

$$D_{\alpha}\equiv\partial_t+\boldsymbol{e}_{\alpha}\cdot\boldsymbol{\nabla},\quad\text{and}\quad D_{\alpha}^{(0)}\equiv\partial_{t_0}+\boldsymbol{e}_{\alpha}\cdot\boldsymbol{\nabla}.$$

The first few equations of a set of successive equations in the order of ε obtained from the lattice Boltzmann equation (18) are

$$\varepsilon^0\colon f_{\alpha}^{\sigma(0)}=f_{\alpha}^{\sigma(\mathrm{eq})}\left[1+\frac{(\boldsymbol{e}_{\alpha}-\boldsymbol{u})\cdot(\boldsymbol{u}_{\sigma}-\boldsymbol{u})}{R_{\sigma}T}\right], \tag{C2a}$$

$$\varepsilon^1\colon \delta_t D_{\alpha}^{(0)}f_{\alpha}^{\sigma(0)}=-\frac{1}{\tau_{\sigma}}f_{\alpha}^{\sigma(1)}+J_{\alpha}^{\sigma\varsigma}-\delta_t F_{\alpha}, \tag{C2b}$$

$$\varepsilon^2\colon \delta_t(\partial_{t_1}f_{\alpha}^{\sigma(0)}+D_{\alpha}^{(0)}f_{\alpha}^{\sigma(1)}+\tfrac{1}{2}\delta_t D_{\alpha}^{(0)}D_{\alpha}^{(0)}f_{\alpha}^{\sigma(0)})=-\frac{1}{\tau_{\sigma}}f_{\alpha}^{\sigma(2)}. \tag{C2c}$$

In the Chapman-Enskog analysis, it is assumed that the mutual interaction term $J_{\alpha}^{\sigma\varsigma}$ as well as the forcing term F_{α} are of the first order in ε [17,18,63,64].

The first few moments of the equilibrium distribution function $f_{\alpha}^{\sigma(0)}$ can be easily computed:

$$\sum_{\alpha} f_{\alpha}^{\sigma(0)}=\rho_{\sigma}, \tag{C3a}$$

$$\sum_{\alpha} f_{\alpha}^{\sigma(0)}\boldsymbol{e}_{\alpha}=\rho_{\sigma}\boldsymbol{u}_{\sigma}, \tag{C3b}$$

$$\begin{aligned}\sum_{\alpha} f_{\alpha}^{\sigma(0)}e_{\alpha i}e_{\alpha j}&=R_{\sigma}T\rho_{\sigma}\delta_{ij}+\rho_{\sigma}u_iu_j+\rho_{\sigma}\bigg[(u_{\sigma}-u)_iu_j\\&\quad+(u_{\sigma}-u)_ju_i-\frac{u_iu_j\boldsymbol{u}\cdot(\boldsymbol{u}_{\sigma}-\boldsymbol{u})}{R_{\sigma}T}\bigg]\\&\approx R_{\sigma}T\rho_{\sigma}\delta_{ij}+\rho_{\sigma}u_iu_j,\end{aligned} \tag{C3c}$$

$$\begin{aligned}\sum_{\alpha} f_{\alpha}^{\sigma(0)}e_{\alpha i}e_{\alpha j}\boldsymbol{e}_{\alpha k}&=R_{\sigma}T\rho_{\sigma}\Delta_{ijkl}u_l\left[1-\frac{\boldsymbol{u}\cdot(\boldsymbol{u}_{\sigma}-\boldsymbol{u})}{R_{\sigma}T}\right]\\&\quad+R_{\sigma}T\rho_{\sigma}\Delta_{ijkl}(u_{\sigma}-u)_l+\cdots\\&\approx R_{\sigma}T\rho_{\sigma}(\delta_{ij}u_{\sigma k}+\delta_{jk}u_{\sigma i}+\delta_{ki}u_{\sigma j}),\end{aligned} \tag{C3d}$$

where $e_{\alpha i}$ is the ith Cartesian component of the vector $\boldsymbol{e}_{\alpha}$ and

$$\Delta_{ijkl}=\delta_{ij}\delta_{kl}+\delta_{ik}\delta_{jl}+\delta_{il}\delta_{jk}.$$

In the second- and third-order moments of $f_{\alpha}^{\sigma(0)}$ in Eqs. (C3c) and (C3d), respectively, the terms involving products of the velocity and the velocity difference are neglected.

With the substitution of Eq. (C2b), Eq. (C2c) becomes

$$\begin{aligned}&\delta_t[\partial_{t_1}f_{\alpha}^{\sigma(0)}+D_{\alpha}^{(0)}f_{\alpha}^{\sigma(1)}+\tfrac{1}{2}D_{\alpha}^{(0)}(J_{\alpha}^{\sigma\varsigma}-\delta_t F_{\alpha})]\\&\quad=\frac{1}{\tau_{\sigma}}[\delta_t D_{\alpha}^{(0)}f_{\alpha}^{\sigma(1)}-f_{\alpha}^{\sigma(2)}].\end{aligned} \tag{C4}$$

Therefore, the second-order (in ε) equation for the mass conservation is

$$\partial_{t_1}\rho_{\sigma}=\frac{1}{2}\boldsymbol{\nabla}\cdot\left[\frac{1}{\tau_D}\frac{\rho_{\sigma}\rho_{\varsigma}}{\rho}(\boldsymbol{u}_{\sigma}-\boldsymbol{u}_{\varsigma})\right]=-\frac{1}{2}\delta_t\boldsymbol{\nabla}\cdot p\boldsymbol{d}_{\sigma}, \tag{C5}$$

where the spatial variation of the external force is neglected because it is canceled out by properly setting the velocity change due to the external forcing to $\delta\boldsymbol{u}=\frac{1}{2}\boldsymbol{a}\delta_t$ in the equilibrium distribution function [63,64].

By construction of the forcing term F_{α} [given by Eq. (19c)], we have

$$\sum_{\alpha} F_{\alpha}e_{\alpha i}e_{\alpha j}=0. \tag{C6}$$

Therefore, the forcing term has no contribution to the pressure tensor. However, the partial pressure tensor for each individual species is affected by the cross-interaction term $J_{\alpha}^{\sigma\varsigma}$ because

$$\sum_{\alpha} J_{\alpha}^{\sigma\varsigma}e_{\alpha i}e_{\alpha j}=-\frac{1}{\tau_D}\frac{\rho_{\sigma}\rho_{\varsigma}}{\rho}\left[(u_i\delta u_j+u_j\delta u_i)-\frac{1}{R_{\sigma}T}u_iu_j\boldsymbol{u}\cdot\delta\boldsymbol{u}\right], \tag{C7}$$

where $\delta \boldsymbol{u} \equiv (\boldsymbol{u}_\sigma - \boldsymbol{u}_\varsigma)$, and $\delta u_i \equiv (u_{\sigma i} - u_{\varsigma i})$. The effect due to $J_\alpha^{\sigma\varsigma}$ is of higher order in $\boldsymbol{u}$ and $\boldsymbol{u}_\sigma$, and thus can be neglected in the pressure. Consequently, the partial pressure tensor can be approximated as the following:

$$\begin{aligned}
\mathsf{P}_{ij}^{\sigma(1)} &= \sum_\alpha f_\alpha^{\sigma(1)} e_{\alpha i} e_{\alpha j} \\
&= -\tau_\sigma \delta_t \sum_\alpha D_\alpha^{(0)} f_\alpha^{\sigma(0)} e_{\alpha i} e_{\alpha j} \\
&= -\tau_\sigma \delta_t \left[\partial_{t_0} \sum_\alpha f_\alpha^{\sigma(0)} e_{\alpha i} e_{\alpha j} + \sum_\alpha \boldsymbol{\nabla} \cdot \boldsymbol{e}_\alpha f_\alpha^{\sigma(0)} e_{\alpha i} e_{\alpha j} \right] \\
&= -\tau_\sigma \delta_t [\partial_{t_0} (R_\sigma T \rho_\sigma \delta_{ij} + \rho_\sigma u_i u_j) + R_\sigma T (\partial_j \rho_\sigma u_{\sigma i} \\
&\qquad + \partial_i \rho_\sigma u_{\sigma j} + \delta_{ij} \boldsymbol{\nabla} \cdot \rho_\sigma \boldsymbol{u}_\sigma)] + \cdots \\
&= -\tau_\sigma \delta_t [R_\sigma T (\partial_{t_0} \rho_\sigma + \boldsymbol{\nabla} \cdot \rho_\sigma \boldsymbol{u}_\sigma) \delta_{ij} + \partial_{t_0} (\rho_\sigma u_i u_j) \\
&\qquad + R_\sigma T (\partial_j \rho_\sigma u_{\sigma i} + \partial_i \rho_\sigma u_{\sigma j})] + \cdots \\
&\approx -\tau_\sigma \delta_t R_\sigma T (\partial_j \rho_\sigma u_{\sigma i} + \partial_i \rho_\sigma u_{\sigma j}), \qquad \text{(C8)}
\end{aligned}$$

where the terms smaller than $O(M^2)$ (M is the mach number) are dropped as usual [65]. Therefore,

$$\partial_j \mathsf{P}_{ij}^{\sigma(1)} \approx -\tau_\sigma \delta_t R_\sigma T \rho_\sigma \nabla^2 u_{\sigma i}. \qquad \text{(C9)}$$

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