

Lattice Boltzmann study of pattern formation in reaction-diffusion systems

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(Received 19 July 2010; revised manuscript received 24 October 2010; published 26 January 2011)

Pattern formation in reaction-diffusion systems is of great importance in surface micropatterning [Grzybowski *et al.*, *Soft Matter* **1**, 114 (2005)], self-organization of cellular micro-organisms [Schulz *et al.*, *Annu. Rev. Microbiol.* **55**, 105 (2001)], and in developmental biology [Barkai *et al.*, *FEBS Journal* **276**, 1196 (2009)]. In this work, we apply the lattice Boltzmann method to study pattern formation in reaction-diffusion systems. As a first methodological step, we consider the case of a single species undergoing transformation reaction and diffusion. In this case, we perform a third-order Chapman-Enskog multiscale expansion and study the dependence of the lattice Boltzmann truncation error on the diffusion coefficient and the reaction rate. These findings are in good agreement with numerical simulations. Furthermore, taking the Gray-Scott model as a prominent example, we provide evidence for the maturity of the lattice Boltzmann method in studying pattern formation in nonlinear reaction-diffusion systems. For this purpose, we perform linear stability analysis of the Gray-Scott model and determine the relevant parameter range for pattern formation. Lattice Boltzmann simulations allow us not only to test the validity of the linear stability phase diagram including Turing and Hopf instabilities, but also permit going beyond the linear stability regime, where large perturbations give rise to interesting dynamical behavior such as the so-called self-replicating spots. We also show that the length scale of the patterns may be tuned by rescaling all relevant diffusion coefficients in the system with the same factor while leaving all the reaction constants unchanged.

DOI: [10.1103/PhysRevE.83.016702](https://doi.org/10.1103/PhysRevE.83.016702)

PACS number(s): 47.11.-j, 05.10.-a, 82.40.Ck, 82.20.-w

I. INTRODUCTION

Spatially and/or temporally varying patterns have been observed in a variety of physical [1,2], chemical [3–5], and biological [6–11] systems operating far from equilibrium. The interest in understanding the physics of pattern formation in these systems has been increasing steadily over the last few years, especially after the experimental verification of Turing's idea [12]. In chemical and biological systems, for instance, macroscopic reaction-diffusion equations have been proposed as models for morphogenesis [13], pattern formation [6,7], and self-organization [14,15]. This class of equations usually includes the following two features: (i) a nonlinear reaction between chemical species describing local production or consumption of the species, and (ii) the diffusive transport of these species due to density gradients. The simple form of the reaction-diffusion equation for a system of N species is described by the following set of equations:

$$\frac{\partial \rho_s(\mathbf{x}, t)}{\partial t} = D_s \Delta \rho_s(\mathbf{x}, t) + R_s, \quad 1 \leq s \leq N, \quad (1)$$

where $\rho_s(\mathbf{x}, t)$ is the mass density or concentration of species s at time t and location \mathbf{x} , Δ is the Laplacian operator with respect to spatial coordinate \mathbf{x} , and D_s is the diffusion coefficient of individual species s . In this work, we assume that D_s is isotropic and independent of \mathbf{x} . The last term on the right hand side, R_s , is the reaction term. This term depends on the local density or concentration of the individual reacting species and the reaction mechanism governing the system. In most pattern forming systems, R_s usually contains nonlinear or autocatalytic reaction terms with the product of the densities of the reacting species.

Due to their great importance both in biology, environmental science, and industry, there has been growing interest in a

study of these systems both experimentally, by numerical integration of the governing equations, and via well-tuned analytic theories (see, e.g., [16–22] and references therein). However, solving problems with complex geometry (as is sometimes the case in biological systems) often requires a more efficient and robust method. The lattice Boltzmann method has met significant success in simulating a wide range of phenomena in complex geometries over the last decades [23–27]. In contrast with other traditional numerical techniques which only focus on the solution of the governing macroscopic equation, the lattice Boltzmann (LB) method is based on kinetic theory. In cell-scale modeling of micro-organisms [28–30], for instance, the kinetic nature of the lattice Boltzmann method makes the approach computationally less demanding and allows for a relatively simple implementation of microbial interactions between cells. Furthermore, for problems involving large domain sizes, the local nature of LB operations allows easier implementation on parallel computational platforms thus enabling fast and large scale computations. In addition to the above features, the inherent capability of the LB approach in dealing with irregular boundaries makes it suitable for studying reaction-diffusion phenomena in porous media [31] at the pore scale. However, the accuracy and efficiency of a numerical method are often evaluated in terms of the smallest truncation error within the method. In previous lattice Boltzmann studies of reaction-diffusion equation [32,33], it is rather unclear as to how the truncation error varies with the system parameters such as reaction rate and diffusion constant. These parameters become important in pattern forming systems where nonlinear reaction terms are present and reaction rate as well as diffusion constant may vary over a wide range. Thus for a better performance and accuracy, it is important to find out whether there is a range of optimal parameters that leads to the smallest truncation error and a better convergence of the method. Such

a study is performed in this work for the case of a single species reaction-diffusion systems. Performing a third-order Chapman-Enskog multiscale expansion, we investigate the dependence of the truncation error on the system parameters. Indeed, for this simple case, while the truncation error linearly varies with the reaction rate, it exhibits a pronounced minimum as a function of the diffusion coefficient.

In order to extend the investigation to a pattern-forming multispecies reaction-diffusion model, we have selected the Gray-Scott model [34], which serves as a standard paradigm for studying reaction-diffusion systems. The Gray-Scott model, though simple, exhibits a wide range of interesting dynamical features including spots [35], spiral waves [36], stationary waves [37], and spatiotemporal chaos [38]. A particular feature of this model which makes it different from the other models is the existence of the so-called self-replicating spots [39]. Spatially localized cell-like structures grow, deform, and make replicas of themselves. This act of “cell division” resembling DNA and RNA replication in cells or the replication growth of biological cells as seen in developmental biology makes it an ideal model for studying these biological systems with regard to pattern formation. In this reaction-diffusion system, generation of patterns comes usually from the instability of an initially uniform state to spatially inhomogeneous perturbations over a certain range of wavelengths. The possible range of wavelengths, as determined by a fixed set of system parameters, is usually invariant against a change of the system size. A change in system size often leads to a corresponding change in the number of spots, stripes, or segments observed in the system. Hence the number of segments or stripes is not invariant, but proportional to the system size. In contrast, for some biological systems, the pattern-forming wavelength is often proportional to the system size, while the number of stripes or segments is invariant against the change of system size. For instance, some mammalian coat markings have been shown to enlarge in proportion to system size [40], patterns in some micro-organisms like *Hydra* and *Dictyostelium discoideum* have also been observed to show proportionality with size [41]. Modeling this type of biological systems with Turing-type reaction-diffusion therefore requires rendering the governing equations dimensionless and adjusting the system parameters in a proper way [42,43]. One such approach involves using diffusion constants which depend on the concentration of a system-size-dependent auxiliary chemical factor [44–47] or using the possibility that the concentration of some chemical species changes with some power of the system size [48]. Interestingly, it is possible to change the length scale of the patterns in the Gray-Scott model via a simple rescaling of all the involved diffusion coefficients by the same factor, while keeping all the reaction constants unchanged. We provide a test of the validity of this simple approach with lattice Boltzmann simulations.

The paper is organized as follows. In the following section, we briefly introduce the lattice Boltzmann simulation scheme for the reaction-diffusion equation. We then provide some benchmark tests for our LB simulation by comparing our results with analytical solutions for the transformation reaction and diffusion of a point source in a domain with periodic boundary conditions. Excellent agreement with the analytical

solutions is found. We also carry out a truncation error analysis of the model via a third-order multiscale expansion. Results obtained from this analysis are in agreement with our numerical simulations. In Sec. III, we present the Gray-Scott model and, using linear stability analysis, determine the parameter range for the existence of unstable solutions which we identify as a necessary condition for pattern formation. Our numerical simulations show good agreement with the predictions obtained from linear stability analysis. In Sec. IV we present a detailed study of the patterns which may be obtained via large amplitude perturbations of a linearly stable state. This case comprises the self-replicating spots.

II. NUMERICAL MODEL AND ITS VALIDATION

A. Lattice Boltzmann method

The lattice Boltzmann method [49–52] can be regarded as a mesoscopic particle based numerical approach allowing us to solve fluid-dynamical equations in a certain approximation, which (within, e.g., the so-called diffusive scaling, i.e., by choosing $\Delta t = \Delta x^2$) becomes exact as the grid resolution is progressively increased. The density of the fluid at each lattice site is accounted for by a one particle probability distribution $f_i(\mathbf{x}, t)$, where \mathbf{x} is the lattice site, t is the time, and the subscript i represents one of the finite velocity vectors \mathbf{e}_i at each lattice node. The number and direction of the velocities are chosen such that the resulting lattice is symmetric so as to easily reproduce the isotropy of the fluid [53]. During each time step, particles stream along velocity vectors \mathbf{e}_i to the corresponding neighboring lattice site and collide locally, conserving mass and momentum in the process. The LB equation describing propagation and collision of the particles is given by

$$f_i(\mathbf{x} + \mathbf{e}_i, t + 1) - f_i(\mathbf{x}, t) = \Omega_i f_i(\mathbf{x}, t), \quad (2)$$

where Ω_i is the collision operator.

The most widely used variant of LB is the lattice Bhatnagar-Gross-Krook (BGK) model [51], which approximates the collision step by a single time relaxation toward a local equilibrium distribution f_i^{eq} . The lattice BGK model is written as

$$f_i(\mathbf{x} + \mathbf{e}_i, t + 1) - f_i(\mathbf{x}, t) = \frac{f_i^{\text{eq}}(\mathbf{x}, t) - f_i(\mathbf{x}, t)}{\tau}, \quad (3)$$

where τ is the relaxation time and the equilibrium distribution f_i^{eq} is closely related to the low Mach number expansion of the Maxwell velocity distribution given as [54]

$$f_i^{\text{eq}}(\mathbf{x}, t) = w_i \rho \left[1 + \frac{1}{c_s^2} (\mathbf{e}_i \cdot \mathbf{u}) + \frac{1}{2c_s^4} (\mathbf{e}_i \cdot \mathbf{u})^2 - \frac{1}{2c_s^2} u^2 \right]. \quad (4)$$

In Eq. (4), c_s is the sound speed on the lattice and w_i is a set of weights normalized to unity. The weights w_i and speed c_s depend in general on the dimension and the type of the lattice used. In this work, we use the two dimensional nine velocity (D2Q9) model with the sound speed c_s given as $c_s^2 = c^2/3$, where $c = \Delta x/\Delta t$ is the lattice speed. The lattice weights w_i for the D2Q9 model are given as

$$w_i = \begin{cases} 4/9 & \mathbf{e}_i = (0,0), \quad i = 0; \\ 1/9 & \mathbf{e}_i = (\pm 1,0), (0, \pm 1), \quad i = 1 \dots 4; \\ 1/36 & \mathbf{e}_i = (\pm 1, \pm 1), \quad i = 5 \dots 8. \end{cases} \quad (5)$$

In order to model the reaction-diffusion equations in the framework of a lattice Boltzmann BGK model, we introduce a multispecies distribution function $f_{i,s}$ where the subscript s runs over the number of species $s = 1 \dots N$. Here we assume that the diffusion of a given species does not depend on the concentration of other species. In other words, the species in our model do not interact among each other, except through the chemical reaction term. This assumption is physically justified since many pattern-forming systems are studied in the form of dilute solutions. At higher concentrations, however, the mutual interactions of different species shall be taken into account [55,56]. In addition, we focus on chemical reactions with diffusion as the only transport mechanism, i.e., with no effect of the solvent velocity field. Experimentally, this is a good approximation to pattern formation in a gel [57,58], where the effect of advection is largely suppressed. Thus the flow velocity \mathbf{u} in Eq. (4) can be set to zero. This leads to

$$f_{i,s}^{\text{eq}}(\mathbf{x}, t) = w_i \rho_s. \quad (6)$$

Equation (6) satisfies the requirement $\sum_{i=0}^N f_{i,s}^{\text{eq}} = \rho_s$. The chemical reaction is modeled by including a source term, R_s , in the collision step. This leads to

$$f_{i,s}(\mathbf{x} + \mathbf{e}_i, t + 1) - f_{i,s}(\mathbf{x}, t) = \frac{f_{i,s}^{\text{eq}}(\mathbf{x}, t) - f_{i,s}(\mathbf{x}, t)}{\tau_s} + w_i R_s, \quad (7)$$

where τ_s is the relaxation time for species s . The source term R_s represents the rate of change of density of the species s with regard to reaction kinetics. The exact form of the relation between the reaction rate R_s and the density (concentration) of each species depends on the type of reaction being modeled. The density of the species s , ρ_s is then computed from the distribution function using $\rho_s = \sum_{i=0}^N f_{i,s}$.

Near equilibrium and in the limit of small Knudsen number (=mean free path/characteristic length of problem) the macroscopic reaction-diffusion equation can be recovered using Chapman-Enskog multiscale analysis. The detailed analysis leading to the macroscopic equation is outlined in the Appendix. The relaxation time τ_s is then found to be related to the diffusion coefficient as $D_s = c_s^2 \Delta t (\tau_s - 0.5)$.

B. A transformation reaction

We provide here a simple test of our lattice Boltzmann approach for reaction-diffusion systems and characterize the truncation error obtained with regard to system parameters. We compute the problem of diffusion of a species A undergoing an irreversible transformation or decay reaction to a species B ,



The reaction-diffusion equation describing the dynamics of species A can be written as

$$\frac{\partial \rho_A(x, y, t)}{\partial t} = D_A \Delta \rho_A(x, y, t) - \kappa_B \rho_A(x, y, t), \quad (9)$$

where $\rho_A(x, y, t)$ is the density of species A at point (x, y) and time t , D_A is the diffusion coefficient of A , κ_B is the rate of the transformation reaction, and Δ is the Laplacian operator with respect to spatial coordinates x, y . Taking ρ_{A0} as a reference

concentration, Eq. (9) can be made dimensionless by introducing the characteristic time $t_d = L^2/D_A$, which is the time for the diffusion over the dimension of the simulation box L . This choice is reasonable since L is the only characteristic length in this problem ($L_x = L_y = L$). This yields

$$\frac{\partial \tilde{\rho}_A(\tilde{x}, \tilde{y}, \tilde{t})}{\partial \tilde{t}} = \Delta \tilde{\rho}_A(\tilde{x}, \tilde{y}, \tilde{t}) - \phi^2 \tilde{\rho}_A(\tilde{x}, \tilde{y}, \tilde{t}), \quad (10)$$

where $\tilde{\rho}_A = \rho_A/\rho_{A0}$, $(\tilde{x}, \tilde{y}) = (x, y)/L$, $\tilde{t} = t/t_d$, and $\phi^2 = t_d \kappa_B = L^2 \kappa_B/D_A$. The Thiele modulus ϕ^2 compares the rate of reaction to the rate of diffusion. Transport by diffusion dominates the reaction kinetics when $\phi^2 \ll 1$, while reaction dominates when $\phi^2 \gg 1$. We proceed now to solve Eq. (10) by Fourier transformation. Using the initial condition $\tilde{\rho}_A(\tilde{x}, \tilde{y}, \tilde{t} = 0) = \delta(\tilde{x} - \tilde{x}_0)\delta(\tilde{y} - \tilde{y}_0)$, the Fourier transformation of Eq. (10) yields

$$\frac{d\hat{\rho}_A(\tilde{q}, \tilde{t})}{d\tilde{t}} = \tilde{q}^2 \hat{\rho}_A(\tilde{q}, \tilde{t}) - \phi^2 \hat{\rho}_A(\tilde{q}, \tilde{t}), \quad \hat{\rho}_A(\tilde{q}, 0) = 1, \quad (11)$$

where $\hat{\rho}_A(\tilde{q}, \tilde{t})$ is the Fourier transform of $\tilde{\rho}_A(\tilde{x}, \tilde{y}, \tilde{t})$. Integrating Eq. (11), taking the inverse Fourier transform, and slightly re-arranging the terms, one obtains

$$\tilde{\rho}_A(\tilde{x}, \tilde{y}, \tilde{t}) = \frac{1}{(4\pi\tilde{t})} \exp\left(\frac{-(\tilde{x} - \tilde{x}_0)^2 - (\tilde{y} - \tilde{y}_0)^2}{4\tilde{t}}\right) \exp(-\phi^2\tilde{t}). \quad (12)$$

Equation (12) is the analytical solution of the problem posed by Eq. (10) on a region infinitely extended in space. In particular, it does not contain effects of periodic images of the simulation box [59]. It thus provides a satisfactory approximation to the simulated problem as long as the width of the Gaussian is small compared to the linear dimension of the simulation box, i.e., $D_A t \ll L^2$ or, equivalently, $\tilde{t} \ll 1$. Using Eq. (12), we perform a test of the present lattice Boltzmann (LB) model. For this purpose, we set up a two dimensional domain with $L = 200$ lattice units and $\rho_A(t = 0) = 1$ at the center of the simulation box ($x = x_0 = L/2, y = y_0 = L/2$), while $\rho_A(t = 0) = 0$ on all other points in the system. For the whole region of the domain, we initialize the density of the species B to zero. Periodic boundary condition is imposed along both the x and y directions. The lattice Boltzmann relaxation parameter of the species A and B is set to $\tau_A = \tau_B = 0.56$, corresponding to $D_A = D_B = 0.02$ (in LB units). The reaction rate, on the other hand, is $\kappa_B = 0.01$. These values correspond to a Thiele modulus of $\phi^2 = 2 \times 10^4$.

Results thus obtained are collected in Fig. 1(a). The plot compares, for two different times, the density profiles of species A along the x direction ($y = y_0 = L/2$) obtained from simulations to the analytical solution, Eq. (12).

In order to quantify the numerical error, we introduce the relative error E_ρ via the definition

$$E_\rho = \sqrt{\frac{\sum_{x,y} |\rho_{A,\text{an}}(x, y) - \rho_{s,\text{sim}}(x, y)|^2}{\sum_{x,y} |\rho_{A,\text{an}}(x, y)|^2}}. \quad (13)$$

Here, $\rho_{A,\text{an}}(x, y)$ is the density field obtained from the analytical solution in Eq. (12) and $\rho_{s,\text{sim}}(x, y)$ is the density field obtained from the simulation. The summation is taken over all lattice points in the domain.

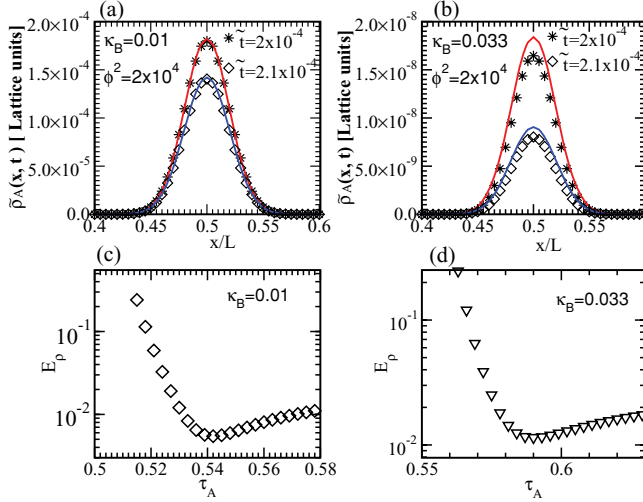


FIG. 1. (Color online) Comparison of the lattice Boltzmann simulations (symbols) with the analytical solution (solid lines) given in Eq. (12). (a) Density profiles of species A along the x direction at two different dimensionless times as indicated for $\kappa_B = 0.01$ and $\tau_A = \tau_B = 0.56$ (corresponding to $D_A = D_B = 0.02$) along the line $y = y_0$. (b) The same data as in (a) but for $\kappa_B = 0.033$. (c) Behavior of the relative error in the density of A , E_ρ [see Eq. (13)], with the relaxation parameter τ_A at $\kappa_B = 0.01$. (d) The same data as in (c) but for $\kappa_B = 0.033$. The Thiele modulus is kept constant for all the data shown here, $\phi^2 = 2 \times 10^4$. Note that a larger τ_A corresponds to a higher grid resolution, via Eq. (14).

As seen from Fig. 1(a), we obtain a good agreement between the LB simulation data and the analytical solution in Eq. (12). Using Eq. (13), this corresponds to a relative error of less than 1%. We note, however, that increasing the value of the reaction rate κ_B leads to an increase in the relative error as evident by comparing Figs. 1(a) and 1(b). We will return to this point further below.

To proceed further, we express the Thiele modulus in terms of dimensionless lattice Boltzmann parameters. For this purpose, we recall the relation between relevant physical and LB quantities, $D_A^{\text{phys}} = c_s^2 \Delta t (\tau_A - 0.5) = (\tau_A - 0.5) \Delta x^2 / (3 \Delta t)$, $\kappa_B^{\text{phys}} = \kappa_B / \Delta t$, and $L^{\text{phys}} = L \Delta x$. Note that, since all our simulation results are expressed in LB units, we have dropped—for the sake of the simplicity of notation—the superscript “LB” in all the lattice Boltzmann variables. Inserting these relations into the definition of the Thiele number, it is easy to see that

$$\phi^2 = \frac{L^{\text{phys}2} \kappa_B^{\text{phys}}}{D_A^{\text{phys}}} = \frac{3L^2 \kappa_B}{(\tau_A - 0.5)}. \quad (14)$$

A nice property of Eq. (14) is that it provides a prescription for how to change the grid resolution without changing the physical problem under investigation ($\phi = \text{const}$). In particular, in the case of the present transformation reaction and for a fixed lattice reaction rate κ_B , an increase of the grid resolution (i.e., an increase of L) must be accompanied by a corresponding increase of the LB relaxation parameter τ_A .

The reader may have noticed that the sound speed enters the Thiele modulus via a factor of 3 only. This is related to the fact that the scale of the sound speed, $\Delta x / \Delta t$, drops out

when constructing this dimensionless quantity. Indeed, effects of the sound speed on numerical accuracy are not expected to show up at this level of description, where the flow velocity is assumed to be identically zero. In the presence of flow (generated, e.g., via buoyancy forces), on the other hand, this issue may become important. A more detailed discussion of the effects of sound speed on the LB truncation error can be found in [60].

Next we systematically investigate how the relative error in the density, E_ρ , varies with the relaxation time τ_A and the reaction rate κ_B for a given physical situation. For this purpose, we first fix the Thiele modulus to $\phi^2 = 2 \times 10^4$ as in the case of Figs. 1(a) and 1(b). We then vary, for each value of the reaction rate κ_B , the relaxation parameter τ_A and determine the corresponding relative error. Note that, by doing so, the lattice resolution is automatically adapted according to Eq. (14). Results of these studies are shown in Figs. 1(c) and 1(d) for $\kappa_B = 0.01$ and $\kappa_B = 0.033$, respectively. The first observation from both curves in Figs. 1(c) and 1(d) is the existence of a minimum in the magnitude of the relative error. It is interesting that a similar minimum in the LB truncation error is also observed in the computation of shear stress [61]. Another observation is the shift in the position of the minimum of these curves with increasing reaction rate. Figure 1(c) shows that the minimum occurs at $\tau_{A,\text{min}} \approx 0.536$, for $\kappa_B = 0.01$, while increasing κ_B to 0.033 leads to $\tau_{A,\text{min}} \approx 0.59$ [Fig. 1(d)].

In order to better understand this minimum in this model and characterize the shift observed for an increasing reaction rate, we perform a third-order Chapman-Enskog expansion of the lattice Boltzmann BGK model for the reaction-diffusion equation (see the Appendix) and obtain an expression for the truncation error up to the third order in the expansion parameter ϵ . Using $R_A = -\kappa_B \rho_A$ in Eq. (A23) and rearranging the terms in powers of τ_A leads to

$$E = 3 \Delta x^2 \partial_t \partial_{x_\alpha}^2 \rho_A \left[\tau_A^2 - \left(\frac{\kappa_B \partial_t \rho_A}{3c_s^2 \partial_t \partial_{x_\alpha}^2 \rho_A} + 1 \right) \tau_A + \frac{1}{6} \right]. \quad (15)$$

The third-order LB truncation error E is thus the product of time and spatial derivative of density with a quadratic polynomial in τ_A . This polynomial has a minimum at

$$\tau_{A,\text{min}} = \left(\frac{\kappa_B \partial_t \rho_A}{6c_s^2 \partial_t \partial_{x_\alpha}^2 \rho_A} + 0.5 \right). \quad (16)$$

Strictly speaking, the value of $\tau_{A,\text{min}}$ not only depends on the reaction rate κ_B but also on space and time variables (through ρ_A). The fact that a minimum does indeed occur in E_ρ as a function of τ_A is therefore not at all a trivial consequence of Eq. (15). Indeed, the shape of $E_\rho(\tau_A)$ significantly deviates from a parabola suggesting that the nontrivial effects related to time and spatial derivatives of ρ_A are present.

Nevertheless, it is worth testing to which extent useful information on the behavior of LB truncation error can be gained via the above analysis. For this purpose, we note two important features, which can be extracted from Eqs. (15) and (16). The first one is that E_ρ could be a linear function of the reaction rate κ_B [see Eq. (15)]. The second observation is that also the position of the minimum in $E_\rho(\tau_A)$, i.e. the value of $\tau_{A,\text{min}}$ could be a linearly increasing function of κ_B .

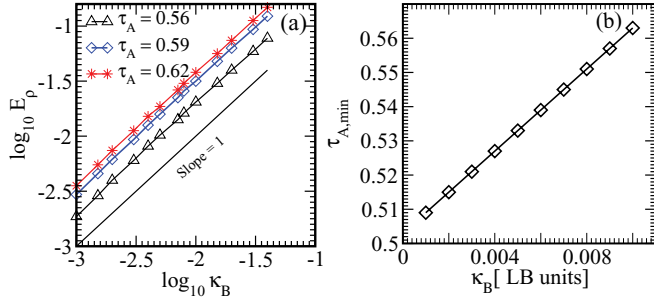
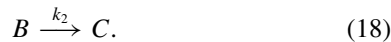
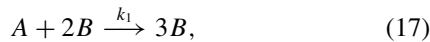


FIG. 2. (Color online) (a) Log-log plot of E_ρ vs κ_B for different values of τ_A . The curves are parallel to the solid black line with a slope of 1. (b) Plot of $\tau_{A,\min}$ vs κ_B . The curve is in line with Eq. (16).

As illustrated in Fig. 2, both these predictions are confirmed by our lattice Boltzmann simulations. Moreover—even though not exactly—the position of the minimum error tends toward the predicted limit of $\tau_{A,\min} = 0.5$ as $\kappa_B \rightarrow 0$ [Fig. 2(b)]. In summary, given a physical system (i.e., for constant physical parameters such as reaction rate and diffusion coefficient) the above discussion may provide guidance in choosing an optimum grid resolution, when seeking a compromise between computational cost and numerical accuracy.

III. GRAY-SCOTT MODEL

We consider next the Gray-Scott model as a typical example of a two species reaction-diffusion system where the nonlinear reaction terms between the species coupled with the transport by diffusion give rise to spatiotemporal patterns. The Gray-Scott model describes the kinetics of a simple autocatalytic reaction in an unstirred homogeneous flow reactor [34]. The reactor is confined in a narrow space between two porous walls in contact with a reservoir. Substance A whose density is kept fixed at A_0 in the reservoir outside of the reactor is supplied through the walls into the reactor with the volumetric flow rate per unit volume k_f . Inside the reactor, A undergoes an autocatalytic reaction with an intermediate species B at a rate k_1 . The species B then undergoes a decay reaction to an inert product C at a rate k_2 . The product C and excess reactants A and B are then removed from the reactor at the same flow rate per unit volume k_f . The basic reaction steps are summarized as follows:



The reaction in Eq. (17) is the cubic autocatalytic reaction in which two molecules of species B produce three molecules of B through interaction with the species A . The presence of B stimulates further production of itself, while the presence of A controls the production of B . Substance A is sometimes called the inhibitor and B the activator. By constantly feeding the reactor with a uniform flow of species A while at the same time removing the product and excess reactants, far from equilibrium conditions can be maintained. Note that inside the reactor the two species A and B are assumed to interact only through the nonlinear autocatalytic reaction in Eq. (17). In

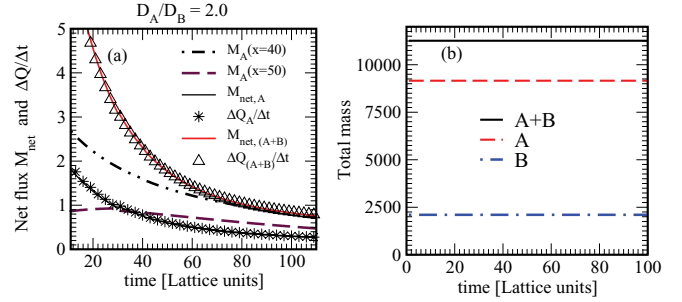


FIG. 3. (Color online) Test of the continuity equation within a stripe defined by two vertical lines at $x_1 = 40$ and $x_2 = 50$. (a) The mass fluxes across x_1 and x_2 are not identical: $M_A(x = 40) \neq M_A(x = 50)$, where $M_A(x) = \sum_{y=0}^{L_y} J_A(x, y)$ and $J_A = \sum_i f_{i,A} c_{i,x}$. This implies that the mass within the domain must change with time ($\Delta Q_A / \Delta t \neq 0$). However, as expected from the continuity equation, this rate of change is identical to the net mass flux across the boundaries of the domain, $M_{\text{net},A}(t) = M_A(x_1) - M_A(x_2)$. Note that, here, we define $M_{\text{net},A}$ as “inflow (at x_1) minus outflow (at x_2).” Similarly, the sum of the masses of the species A and B within the stripe also changes with time without violating the continuity equation. (b) The total mass within the entire simulation box vs time for the species A and B . The initial configuration is identical to that of Figs. 10 (a square box of high concentration of B within an A -rich domain). All the reaction rates are set to zero.

particular, interaction terms due to cross diffusion between the species are neglected. As stated before, this assumption is physically justified as pattern-forming systems often occur in the form of dilute solutions. Following this assumption, the equations of chemical kinetics which describe the above situations and include the spatiotemporal variations of the concentrations of A and B in the reactor take the following form:

$$\frac{\partial A}{\partial t} = k_f(A_0 - A) - k_1 B^2 A + D_A \nabla^2 A, \quad (19)$$

$$\frac{\partial B}{\partial t} = -(k_f + k_2)B + k_1 B^2 A + D_B \nabla^2 B, \quad (20)$$

where A and B are the density of species A and B , respectively, A_0 is the density of A in the reservoir, while D_A and D_B are the diffusion coefficients of species A and B , respectively. Note that the full Gray-Scott model in Eqs. (19) and (20) describe chemical reaction and mass transfer in an open system and therefore it is generally not a mass-conserving system. However, by setting all the reaction terms in Eqs. (19) and (20) to zero, one recovers a purely diffusive system. In this case, it is easy to see that the system is mass conserving. This, however, does not imply that the sum of the densities of all the species at a given point in space is constant. Rather, the rate of mass change in a given area in space is identical to the net flux across the boundary of that domain. Our numerical simulations do indeed confirm this fact (Fig. 3).

In order to understand and control the relevant time scale and length scale of patterns observed in these systems, we introduce variables in the form of time and length scales that represent the physical processes acting in the system. For A , the characteristic time scale is the time for the removal of A given

as $1/k_f$ whereas for B it is $1/(k_f + k_2)$. The characteristic time and length scales for these quantities are then

$$\tau_A = 1/k_f, \quad \tau_B = 1/(k_f + k_2), \quad l_A = (D_A \tau_A)^{1/2}, \quad (21)$$

$$l_B = (D_B \tau_B)^{1/2}.$$

Please note that in contrast to Sec. II B where τ_A and τ_B are used to denote the LB relaxation parameters for the species A and B , here they denote the times associated with the reaction rates k_f and $k_f + k_2$. Furthermore, note also that the diffusion coefficients D_A and D_B in Eqs. (19) and (20) are independent of τ_A and τ_B . Rather, in an implementation within the present D2Q9-LB model, these quantities are related to the LB relaxation parameters via $D_A = (\tau_A^{\text{LB}} - 0.5)/3$ and $D_B = (\tau_B^{\text{LB}} - 0.5)/3$. Next we introduce the dimensionless quantities

$$\tilde{A} = A/A_0, \quad \tilde{B} = B/B_0, \quad B_0 = \left(\frac{k_f}{k_1}\right)^{1/2}. \quad (22)$$

Inserting the quantities in Eqs. (21) and (22) into Eqs. (19) and (20) leads to

$$\tau_A \frac{\partial \tilde{A}}{\partial t} = -\tilde{B}^2 \tilde{A} + 1 - \tilde{A} + l_A^2 \nabla^2 \tilde{A}, \quad (23)$$

and

$$\tau_B \frac{\partial \tilde{B}}{\partial t} = +\eta \tilde{B}^2 \tilde{A} - \tilde{B} + l_B^2 \nabla^2 \tilde{B}, \quad (24)$$

where the parameter $\eta = A_0(k_1 k_f)^{1/2}/(k_f + k_2)$ is the strength of the activation process. It adjusts the strength of the nonlinear term in Eq. (24).

The number of parameters can be further reduced by rescaling the time and length scale in units of τ_A and l_A , respectively. This yields

$$\frac{\partial \tilde{A}}{\partial \tilde{t}} = -\tilde{B}^2 \tilde{A} + 1 - \tilde{A} + \tilde{\nabla}^2 \tilde{A}, \quad (25)$$

$$\frac{1}{\tau} \frac{\partial \tilde{B}}{\partial \tilde{t}} = +\eta \tilde{B}^2 \tilde{A} - \tilde{B} + \frac{1}{\varepsilon^2} \tilde{\nabla}^2 \tilde{B}, \quad (26)$$

where $\tau = \tau_A/\tau_B$ and $\varepsilon = l_A/l_B = \sqrt{\tau_A D_A/\tau_B D_B}$. The parameter τ describes the relative strength of the reaction terms.

In general, Eqs. (25) and (26) are difficult to investigate by analytic means. However, simple cases exist for which analytical solutions can be found. We start with probably the most simple situation of a spatially homogeneous distribution of \tilde{A} and \tilde{B} ($\nabla^2 \tilde{A} = 0$, $\nabla^2 \tilde{B} = 0$). In this case, the steady state solutions of Eqs. (25) and (26), denoted as \tilde{A}_e and \tilde{B}_e , obey

$$-\tilde{B}_e^2 \tilde{A}_e + 1 - \tilde{A}_e = 0, \quad (27)$$

$$\eta \tilde{B}_e^2 \tilde{A}_e - \tilde{B}_e = 0.$$

Equation (27) has three solutions. The first solution is the trivial homogeneous solution $\tilde{B}_e = 0$, $\tilde{A}_e = 1$. This state exists for all system parameters. The other two solutions exist provided that $\eta > 2$. These are given by

$$\tilde{A}_e^\pm = \frac{\eta \pm \sqrt{\eta^2 - 4}}{2\eta} \quad \text{and} \quad \tilde{B}_e^\pm = \frac{\eta \mp \sqrt{\eta^2 - 4}}{2}. \quad (28)$$

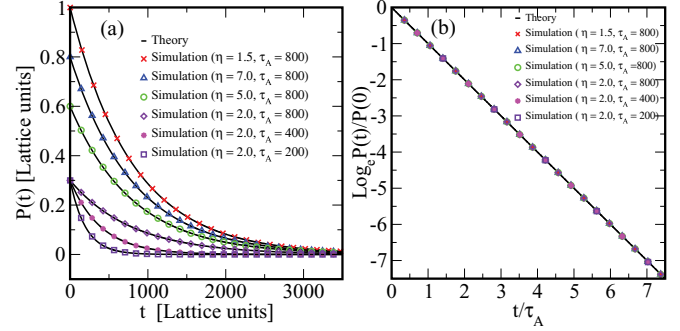


FIG. 4. (Color online) (a) A plot of $-P(t) = \eta \tilde{A}(t) + \tilde{B}(t) - \eta$ vs time for $\tau = \tau_A/\tau_B = 1$ but different values of the parameters τ_A and η as indicated. The solid black curve in each case corresponds to theoretical prediction for the parameter values used in the simulation. In all studied cases, the simulation results exponentially decay to zero and show a perfect agreement with the analytical prediction, Eq. (30). (b) Plot of $\log_e[P(t)/P(0)]$ vs t/τ_A for the same data as shown in (a).

A. Test of simulations in the case of spatially homogeneous dynamics

As a check of our simulation approach, we study the homogenized form of situations where Eqs. (25) and (26) are accessible to an analytical solution. For this purpose, we consider the case of spatially homogeneous dynamics with $\tau = 1$, implying $\tau_A = \tau_B$. In this case, multiplying Eq. (25) by η and adding the result to Eq. (26) leads to

$$\frac{d(\eta \tilde{A} + \tilde{B})}{d\tilde{t}} = \eta - (\eta \tilde{A} + \tilde{B}) \Rightarrow \frac{dP}{d\tilde{t}} = -P, \quad (29)$$

where $P = \eta \tilde{A} + \tilde{B} - \eta$ and we used the fact that $d\eta/d\tilde{t} = 0$. Equation (29) has the simple solution $P(t) = P(0) \exp(-\tilde{t}) = P(0) \exp(-t/\tau_A)$. In other words,

$$\eta \tilde{A}(t) + \tilde{B}(t) - \eta = [\eta \tilde{A}(0) + \tilde{B}(0) - \eta] \exp(-t/\tau_A). \quad (30)$$

A test of Eq. (30) is provided in Fig. 4 for $\tau = 1$ but different values of the parameters τ_A and η . In the case where $\eta < 2$, the simulation starts from a spatially homogeneous state $\tilde{A}(0) = 1$, $\tilde{B}(0) = 0$ with an additional density fluctuation $\delta A = 0.5$ and $\delta B = 0.25$ added homogeneously to A and B , respectively. This is done to break the symmetry which would keep the system at the initial state (due to the autocatalytic nature of the Gray-Scott model, without B , no reaction will take place). For all other cases where $\eta \geq 2$ we start from the nontrivial states (A_e^\pm, B_e^\pm) given by Eq. (28) with an additional small fluctuation of the form $\delta A = 0.1$ and $\delta B = 0.1$. For all values of τ_A and η investigated, we found that the error between theory and simulation is well below 0.2%.

B. Stability analysis of spatially homogeneous states

We proceed in this section to determine the stability of the stationary and homogeneous solutions obtained in Eq. (28) with regard to a spatially homogeneous perturbation. Our analysis starts by looking at the growth rate α of an infinitesimal perturbation about the steady state,

$$\tilde{A} = \tilde{A}_e + \phi_A e^{\alpha t}, \quad \tilde{B} = \tilde{B}_e + \phi_B e^{\alpha t}, \quad (31)$$

where ϕ_A and ϕ_B are the amplitude of the perturbation to the species A and B , respectively. Substituting Eq. (31) into Eqs. (25) and (26), after linearizing and re-arrangement of the terms one arrives at the eigenvalue equation

$$(\mathbf{J} - \alpha \mathbf{I}) \phi = 0, \quad (32)$$

where \mathbf{I} is the identity matrix, $\phi = (\phi_A, \phi_B)^T$, and the matrix \mathbf{J} is given as

$$\mathbf{J} = \begin{bmatrix} \tau(2\eta\tilde{A}_e^\pm\tilde{B}_e^\pm - 1) & \tau\eta\tilde{B}_e^{\pm 2} \\ -2\tilde{A}_e^\pm\tilde{B}_e^\pm & -(\tilde{B}_e^{\pm 2} + 1) \end{bmatrix}. \quad (33)$$

The eigenvalue equation in Eq. (32) has the characteristic polynomial

$$\alpha^2 - \alpha \text{tr} \mathbf{J} + |\mathbf{J}| = 0, \quad (34)$$

where $\text{tr} \mathbf{J}$ and $|\mathbf{J}|$ are the trace and determinant of matrix \mathbf{J} . The pair of solutions or eigenvalues of matrix \mathbf{J} is written

$$\alpha_{1,2} = \frac{1}{2}(\text{tr} \mathbf{J} \pm \sqrt{(\text{tr} \mathbf{J})^2 - 4|\mathbf{J}|}). \quad (35)$$

The eigenvalues $\alpha_{1,2}$ in Eq. (35), can either be real or complex conjugate depending on the relative magnitude and sign of the determinant $|\mathbf{J}|$ and trace $\text{tr} \mathbf{J}$. If the real part of at least one eigenvalue is positive, the considered solution is unstable.

For the trivial state ($A_e = 1, B_e = 0$), $\text{tr} \mathbf{J} = (\tau + 1)$ and $|\mathbf{J}| = \tau$. Using Eq. (35) and the fact that $\tau > 1$ one obtains that both eigenvalues are negative. Hence this state is linearly stable with respect to spatially homogeneous perturbations. Next we consider the nontrivial stationary homogeneous solutions. In this case, inserting the solutions of A_e^\pm and B_e^\pm given in Eq. (28) in Eq. (33), one obtains that $\text{tr} \mathbf{J} = (\tau - \eta\tilde{B}_e^\pm)$ and $|\mathbf{J}| = \tau(\eta\tilde{B}_e^\pm - 2)$. Furthermore, since in this case $\eta > 2$, and using Eq. (28), one can easily verify that $|\mathbf{J}|(\tilde{B}_e^+) = (\eta\tilde{B}_e^+ - 2) < 0$ and $|\mathbf{J}|(\tilde{B}_e^-) = (\eta\tilde{B}_e^- - 2) > 0$. Now from Eq. (35) it follows that, independent of the sign of $\text{tr} \mathbf{J}$, one of the solutions $\alpha_{1,2}$ is always positive, provided that $|\mathbf{J}| < 0$. Hence the state \tilde{B}_e^+ is always unstable. For the state \tilde{B}_e^- , on the other hand, both solutions $\alpha_{1,2}$ will have the same sign as $\text{tr} \mathbf{J}$ and thus the state \tilde{B}_e^- may be stable provided $\text{tr} \mathbf{J} < 0$ (i.e., $\tau < \eta\tilde{B}_e^-$).

Figure 5 shows a typical bifurcation diagram for the system. We plot the homogeneous steady state solutions obtained in Eq. (28) as a function of the control parameter η . For $\eta < 2$ there exists only the trivial state (1,0), while when $\eta > 2$ two additional states, $(\tilde{A}_e^-, \tilde{B}_e^-)$ and $(\tilde{A}_e^+, \tilde{B}_e^+)$, emerge. The state $(\tilde{A}_e^+, \tilde{B}_e^+)$ is always unstable (indicated by a dashed line) while the state $(\tilde{A}_e^-, \tilde{B}_e^-)$ is stable if $\tau < \eta\tilde{B}_e^-$. In the same figure we have also plotted the steady state solutions obtained from the lattice Boltzmann simulation of the spatially homogeneous solutions with small homogeneous perturbations at time $t = 0$. As seen in Fig. 5, the LB simulation well reproduces the analytically predicted stability diagram.

C. Inhomogeneous state and Turing instability

The Gray-Scott model develops a Turing instability for a range of parameters. In this region of the parameter space

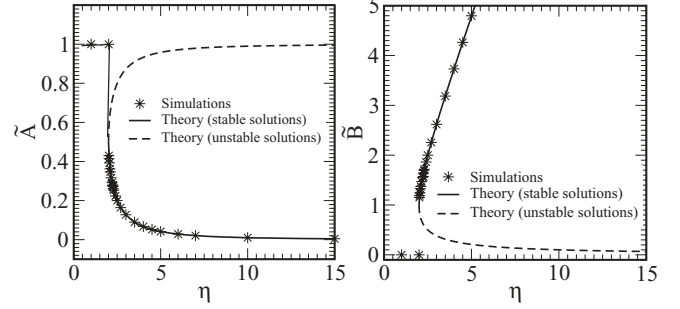


FIG. 5. Plot of the stationary homogeneous state solutions of species A and B given by Eq. (28). Above the bifurcation point ($\eta = 2.0$), two solutions exist: one is unstable to homogeneous perturbations (indicated as a dashed line) and the other may be stable (plotted as a solid line). At $\eta = 2.0$, the stable solution switches to the trivial homogeneous state (1,0) and for $\eta < 2.0$ only the trivial state exists. The lattice Boltzmann simulation (indicated by symbols) shows good agreement with the theory.

the homogeneous steady state solution becomes unstable and a new stationary but inhomogeneous state characterized by the formation of patterns becomes stable. We examine the condition for Turing instability in this system by looking at the growth rate α of an infinitesimal spatially inhomogeneous perturbation to the steady state solutions,

$$\tilde{A} = \tilde{A}_e + \phi_A e^{\alpha t} e^{i\tilde{q}x}, \quad \tilde{B} = \tilde{B}_e + \phi_B e^{\alpha t} e^{i\tilde{q}x}. \quad (36)$$

As in the case of Eq. (31), ϕ_A and ϕ_B are the amplitude of the perturbations to the species A and B respectively, and \tilde{q} is the wave number. Again, inserting Eq. (36) into the kinetic Eqs. (25) and (26) and after linearizing and slight re-arrangement one arrives at the eigenvalue equation

$$(\mathbf{M} - \alpha \mathbf{I}) \phi = 0, \quad (37)$$

where the matrix \mathbf{M} is written as

$$\mathbf{M} = \begin{bmatrix} \tau(2\eta\tilde{A}_e^\pm\tilde{B}_e^\pm - 1 - \frac{\tilde{q}^2}{\epsilon^2}) & \tau\eta\tilde{B}_e^{\pm 2} \\ -2\tilde{A}_e^\pm\tilde{B}_e^\pm & -(\tilde{q}^2 + \tilde{B}_e^{\pm 2} + 1) \end{bmatrix}. \quad (38)$$

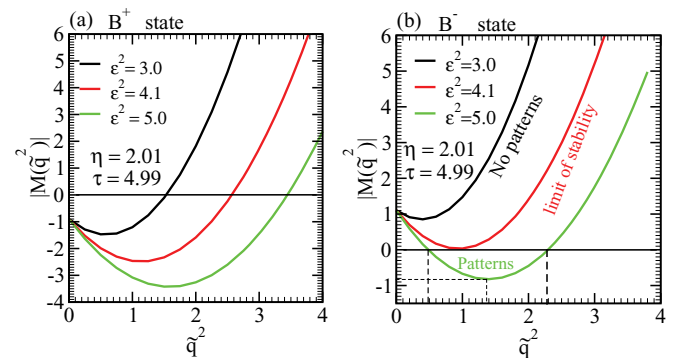


FIG. 6. (Color online) The plot of $|\mathbf{M}(\tilde{q}^2)|$ vs \tilde{q}^2 for states (a) B_e^+ (b) B_e^- . These plots show the range of growth modes \tilde{q} for which the determinant $|\mathbf{M}(\tilde{q}^2)|$ is negative with the possibility of pattern formation.

For the trivial solution ($A_e = 1, B_e = 0$) the matrix M reduces to

$$M = \begin{bmatrix} -\tau(1 + \frac{\tilde{q}^2}{\varepsilon^2}) & 0 \\ 0 & -(\tilde{q}^2 + 1) \end{bmatrix}, \quad (39)$$

and solving the eigenvalue equation in Eq. (37), we obtain the eigenvalues of the trivial state as $\alpha_1 = -\tau(1 + \tilde{q}^2/\varepsilon^2)$ and $\alpha_2 = -(\tilde{q}^2 + 1)$. Since both eigenvalues are negative, the trivial homogeneous state ($A_e = 1, B_e = 0$) is linearly stable for all system parameters, and independent of the wavelength of the applied perturbation. However, it is important to emphasize that this stability is restricted to infinitesimal perturbations. Indeed, the trivial state is found to be unstable with respect to large amplitude spatially inhomogeneous perturbations. In fact, it is in this regime that the so-called self-replicating spots are observed. The original parametrization of the Gray-Scott model by Pearson [34] is also based on the numerical simulation of spatially inhomogeneous perturbations of the trivial state.

For the remaining nontrivial solutions, we insert the homogeneous steady state solutions $\tilde{A}_e^\pm = 1/(\eta\tilde{B}_e^\pm)$ into the matrix M in Eq. (38) and solve for the eigenvalues of M with the characteristic Eq. (34) by replacing J with $M(q)$. The corresponding eigenvalues are then obtained from Eq. (35) by replacing $\text{tr}J$ and $|J|$ with $\text{tr}M(q^2) = \tau - \eta\tilde{B}_e^\pm - \tilde{q}^2(\tau/\varepsilon^2 + 1)$ and $|M(q^2)| = \tilde{q}^4\tau/\varepsilon^2 + \tilde{q}^2(\tau\eta\tilde{B}_e^\pm/\varepsilon^2 - \tau) + \tau(\eta\tilde{B}_e^\pm - 2)$, respectively.

Turing structures or patterns emerge when the system becomes unstable with respect to inhomogeneous perturbations. Again, at least one of the eigenvalues becomes positive (unstable), when $|M(q^2)| < 0$. $|M(q^2)|$ is a parabola in q^2 which attains its minimum value for

$$q_{\min}^2 = \frac{(\varepsilon^2 - \eta\tilde{B}_e^\pm)}{2}. \quad (40)$$

Since $q^2 > 0$, a minimum in $|M(q^2)|$ exists only if $\varepsilon^2 > \eta\tilde{B}_e^\pm$. This is one of the conditions for Turing instability in this system. The boundary of the instability band or range of wave number q for which $|M(q^2)| < 0$ is given by the roots of the equation $|M(q^2)| = 0$:

$$q_{1,2}^2 = \frac{-(\eta\tilde{B}_e^\pm - \varepsilon^2) \pm \sqrt{(\eta\tilde{B}_e^\pm - \varepsilon^2)^2 - 4\varepsilon^2(\eta\tilde{B}_e^\pm - 2)}}{2}. \quad (41)$$

In Eq. (41), there is an important observation concerning the state B_e^+ . Using $\eta > 2$ and Eq. (28) one can show that for the state B_e^+ , the condition $\eta\tilde{B}_e^+ < 2$ holds for all $\eta > 2$. The consequence is that Eq. (28) has always one negative root and one positive root independent of the value of ε^2 . Since $|M(q^2)| < 0$ for $q = 0$ [see Fig. 6(a)], this opens already an instability band for pattern formation as regards the state B_e^+ . On the other hand, for the state B_e^- , two distinct positive real roots are necessary for an instability band of patterns. Thus the following condition has to be satisfied:

$$\eta\tilde{B}_e^- < \varepsilon^2 < -8 + 11.65\eta\tilde{B}_e^- \text{ (Turing space)}. \quad (42)$$

The first condition, $\varepsilon^2 > \eta\tilde{B}_e^-$ as discussed above, is necessary for the formation of Turing patterns while the second one reflects the requirement of a positive discriminant in

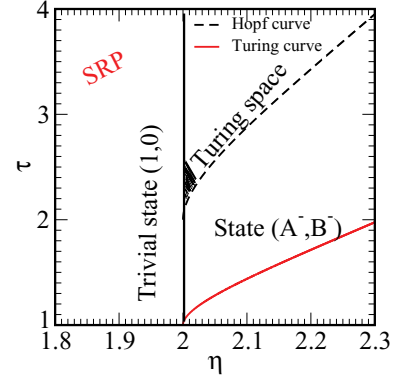


FIG. 7. (Color online) The phase diagram of the model at $D_A/D_B = 2$ showing the Turing curve and the Hopf curve for parameter space spanned by τ and η . For $\eta < 2$ only the trivial state exists and the self-replicating spots (SRP) are observed in this regime only. For $\eta > 2$ two additional states (A_e^\pm, B_e^\pm) emerge. In this figure we have only shown the states (A_e^-, B_e^-). The majority of the patterns formed from the B^- state in our lattice Boltzmann simulations are observed in the shaded region of the Turing space.

Eq. (41). Using the definition of ε^2 the first condition can be re-written as

$$\frac{l_A^2}{l_B^2} > \eta\tilde{B}_e^-. \quad (43)$$

This means that the diffusive length scale for the species A ($l_A = \sqrt{D_A\tau_A}$) must be at least $\eta\tilde{B}_e^-$ times larger than that of B ($l_B = \sqrt{D_B\tau_B}$). In other words, for a given value of parameter τ , the diffusion coefficient of species A has to be $(\eta\tilde{B}_e^-/\tau)$ times larger than that of B . In η and τ parameter space, the curves $\tau = (D_B/D_A)\eta\tilde{B}_e^-$ and $\tau = (D_B/D_A)(-8 + 11.65\eta\tilde{B}_e^-)$ define the limits of stability with respect to Turing patterns. The first curve $\tau = (D_B/D_A)\eta\tilde{B}_e^-$ is plotted as the Turing curve in Fig. 7 for $D_A/D_B = 2$. The second curve $\tau = (D_B/D_A)(-8 + 11.65\eta\tilde{B}_e^-)$ lies above the first curve and falls outside the plotted range. It is therefore not shown in the figure. At zero mode ($q = 0$), another important instability known as the Hopf instability occurs when the real part of a pair of complex eigenvalues passes through zero. In other words, a Hopf instability characterizes the transition from a decaying oscillating mode [$\text{tr}M(0) < 0$] to an oscillation with growing amplitude [$\text{tr}M(0) > 0$]. Thus the limit of Hopf instability is given by the condition $\text{tr}M(0) = 0$. The dashed black line in Fig. 7 indicates the limit of the Hopf instability. The small dashed area in the Turing space is the region where most patterns are expected to be observed.

D. Lattice Boltzmann simulation of the spatially inhomogeneous dynamics

In this section we perform lattice Boltzmann simulations of the Gray-Scott model for different values of parameters τ and η . Here our simulation and parametrization is based on the nontrivial states (A_e^\pm, B_e^\pm). Starting from the homogeneous steady state (A_e^-, B_e^-), we apply a small amplitude density fluctuation of the form $\delta\rho = \phi \cos(q_x x) \cos(q_y y)$ where ϕ is the amplitude given and q is the wave number of the perturbations. We have chosen $\phi = 0.01$ and $q_x = q_y = 1$ in

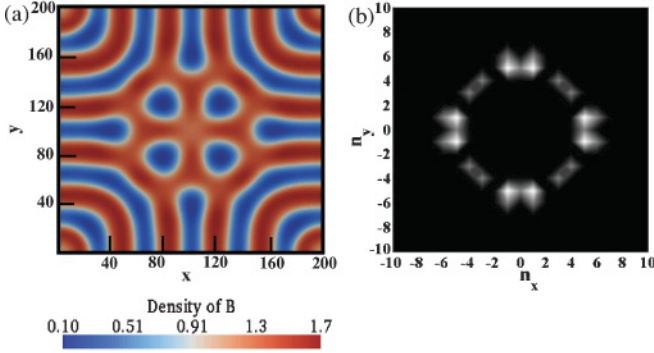


FIG. 8. (Color online) (a) Spatial distribution of the density \tilde{B} at time $t = 400\,000$, $\eta = 2.0139$, and $\tau = 2.7330$. The formation of stripes can be observed at these parameters. (b) Amplitude of the Fourier components of density fluctuation ($\tilde{B} - \tilde{B}_e$) at time $t = 400\,000$, as a function of the wave vector $q = 2\pi[(n_x/L_x)^2 + (n_y/L_y)^2]^{1/2}$. The dimensionless density \tilde{B}_e corresponds to the unstable homogeneous state for the selected set of parameters η and ϵ . The white region in the Fourier spectrum corresponds to the excited wave numbers (n_x, n_y) .

the simulations. Figure 8(a) shows a developed stable structure from the small amplitude initial perturbation to the B^- state with parameters $\eta = 2.014$ and $\tau = 2.733$. For the purpose of comparison with the prediction of linear stability analysis, we perform the Fourier transform of the pattern in Fig. 8(a) and calculate the excited wave numbers in the Fourier spectrum using the relation

$$q = 2\pi[(n_x/L_x)^2 + (n_y/L_y)^2]^{1/2}, \quad (44)$$

where n_x and n_y satisfy $-L_x/2 < n_x < L_x/2$ and $-L_y/2 < n_y < L_y/2$, respectively.

The Fourier spectrum is shown in gray scale in Fig. 8(b) and the excited wave numbers are $(n_x, n_y) \in \{(\pm 1, \pm 5); (\pm 3, \pm 4); (\pm 4, \pm 3); (\pm 5, \pm 1)\}$. Using these values of n_x and n_y in Eq. (44) to calculate q and the parameters $\eta = 2.0139$ and $\epsilon^2 = 5.466$ in Eq. (41), we found that all the excited wave numbers from the simulation fall within the instability band predicted by linear stability analysis in Eq. (41). This provides a further validation of our lattice Boltzmann simulation with regard to this model.

By performing a number of similar simulations with different values of τ and η , we have found that Turing patterns develop over some part of the region where the B^- state is Hopf or Turing unstable (indicated as the Turing space in Fig. 7). The panels in Fig. 9 show developed stationary structures for typical values of the parameters η and τ . These parameter values fall between the saddle node bifurcation curve ($\eta = 2$) and the Turing curve (see Fig. 7). One observation from Fig. 9 is that increasing the value of η within the Turing regime leads to the development of a lacelike structure in the patterns.

IV. BEYOND LINEAR STABILITY: SELF-REPLICATING SPOTS

In this section, a further example is provided for the maturity of the lattice Boltzmann method in studying pattern formation within the Gray-Scott model. The patterns discussed so far

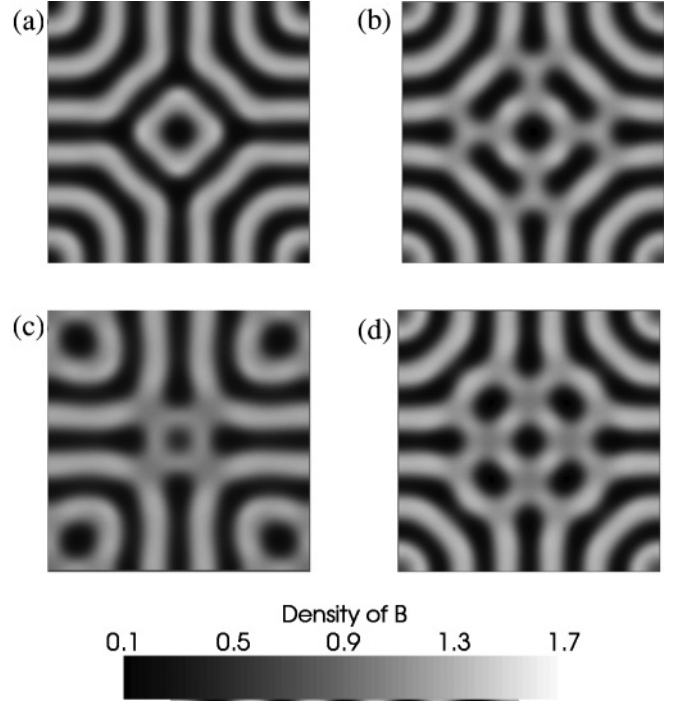


FIG. 9. Stable time independent Turing structures developed from infinitesimal perturbations to state (A_e^-, B_e^-) at parameters (a) $\eta = 2.007\,281\,9$, $\tau = 2.742\,424$, (b) $\eta = 2.013\,958$, $\tau = 2.733\,33$, (c) $\eta = 2.017\,971$, $\tau = 2.707\,462$, and (d) $\eta = 2.018\,433\,6$, $\tau = 2.7272$. The system size in all cases considered above is 200×200 lattice units.

are in the part of the phase diagram (Fig. 7), where linear stability analysis predicts that homogeneous solutions are unstable with respect to small perturbations. There are also other types of structures, which occur in a regime, where the trivial homogeneous state is linearly stable. These patterns emerge only if the homogeneous state is perturbed strongly enough. A prominent example of this type of structure are the so-called self-replicating spots.

Figure 10 illustrates the patterns emerging from a finite amplitude perturbation of the trivial state. The starting configuration corresponds to a rectangular box of species A and B with densities $\tilde{A} = 0.5$, $\tilde{B} = 0.25$ placed at the center of a domain filled with species A and B at densities $\tilde{A} = 1.0$, $\tilde{B} = 0$, respectively. Obviously, such an initial state represents a strong perturbation of the trivial state ($\tilde{A} = 1$, $\tilde{B} = 0$, i.e., $A = A_0$, $B = 0$). The sequence of images in Fig. 10 demonstrates how spots form, elongate, and then replicate as time proceeds. This self-replication process continues until the whole simulation cell is filled with the spots. Interestingly, the number of spots increases with time, while the size of an individual spot seems to remain roughly constant. We have repeated this simulation for a larger box size but otherwise exactly the same parameters. The result of this study is also shown in Fig. 10. As seen from the last image in Fig. 10, the size of a spot does not change, but only the number of spots increases to fill the entire simulation cell.

In the light of the above presented results, one may raise the question of whether it is possible to keep the number of spots constant but tune their size. An answer to this question

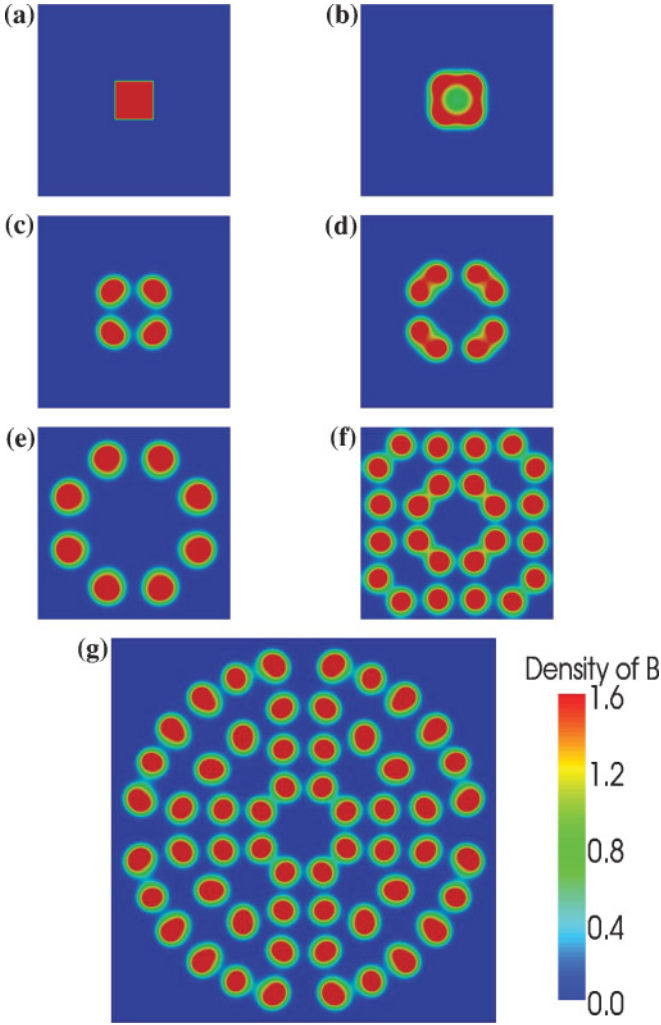


FIG. 10. (Color online) Snapshots of the density distribution of species B showing different stages in the self-replication process at times (a) $t = 0$, (b) $t = 10\,000$, (c) $t = 20\,000$, (d) $t = 50\,000$, (e) $t = 100\,000$, (f) $t = 300\,000$ with size 200×200 lattice units, and (g) $t = 300\,000$ with size 400×400 lattice units.

is obtained by noting that the dynamics of Eqs. (25) and (26) depends only on the dimensionless quantities ε , τ , and η . In other words, we must check whether it is possible to tune the length scale of the problem without altering the values of these dimensionless parameters. This would ensure that the thus-obtained new solution will have exactly the same shape (and thus the same number of spots) but a different length scale (different size spots). Indeed, a look at parameter ε reveals that it is equal to the ratio of two characteristic lengths l_A and l_B , $\varepsilon = l_A/l_B = \sqrt{\tau_A D_A / \tau_B D_B}$. Thus if we multiply both l_A and l_B by a constant factor λ , the parameter ε remains unchanged. Furthermore, in order to keep also the other two parameters τ and η constant, the simplest choice to achieve such a change of length scale is via diffusion coefficient, i.e., via $D_A \rightarrow \lambda^2 D_A$ and $D_B \rightarrow \lambda^2 D_B$.

In order to test the above idea, we design two systems such that system 1 has a linear dimension of $L_1 = 200$ lattice units with diffusion coefficients $D_{A,1} = 0.016 \Delta x^2 / \Delta t$ and $D_{B,1} = 0.008 \Delta x^2 / \Delta t$. For system 2, we choose $L_2 = 400$ lattice units, which means that $\lambda = 2$. Following the above

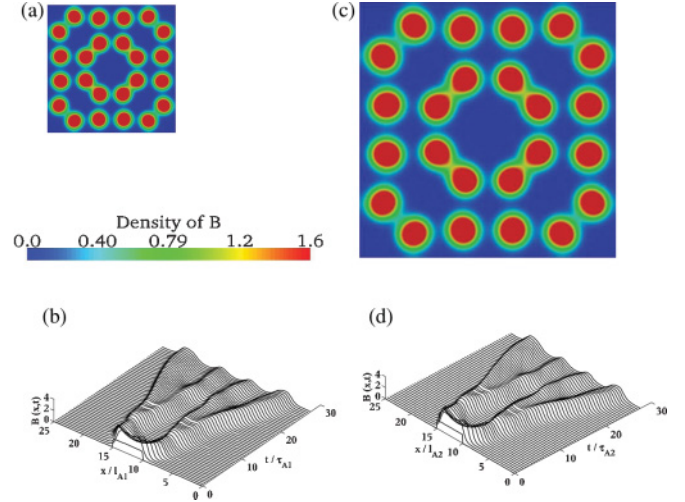


FIG. 11. (Color online) Snapshot of the spatial distribution of the density \tilde{B} showing self-replicating spots at time 300 000, $\eta = 1.86$, $\tau = 3.40$, and $\varepsilon = 2.61$ for the two systems with lattice size (a) 200×200 lattice units, (c) 400×400 lattice units. (b) and (d) show space-time plots of the density profile of the self-replicating spots along a line in the y direction for the two system sizes in (a) and (c), respectively.

arguments, we set the diffusion coefficients of species A and B in system 2 to $D_{A,2} = \lambda^2 D_{A,1} = 0.064 \Delta x^2 / \Delta t$, and $D_{B,2} = \lambda^2 D_{B,1} = 0.032 \Delta x^2 / \Delta t$, respectively. As the initial state, we perturb the trivial state exactly in the same way as described in the context of Fig. 10 and impose periodic boundary conditions in both the x and y directions. Note that the size of the square perturbation must also be multiplied by λ in conformity with the change of length scale. Results of these simulations are shown in Figs. 11(a) and 11(c). The structure of the patterns is identical for the two systems within numerical discretization errors. The inner core diameter of the spots is found to scale as the diffusion length of species A , l_A . A more quantitative comparison of the data is provided in Figs. 11(b) and 11(d), where time evolution of the density profiles is shown for both studied system sizes in a space-time plot along the x direction.

The above arguments on how to tune the length scale while keeping the shape of the patterns unchanged is quite general and applies to any other solution of the Gray-Scott model as well. Here, we provide an example from the Turing regime. This is an interesting test, as linear stability analysis predicts that when the system size is increased, the number of stripes or segments is increased accordingly. However, this applies only if all other parameters are kept constant. Interestingly, by proper regulation of the diffusion coefficient, our numerical simulations in the Turing regime confirm that it is possible to make the wavelength proportional to the system size and keep the number of stripes or segments invariant. Results of these simulations are shown in Fig. 12. For parameters η and τ in the Turing regime, we choose $\eta = 2.014$ and $\tau = 2.733$ and consider two systems with a scaling factor $\lambda = 2$. The spatial density distribution obtained from the simulations is shown in Figs. 12(a) and 12(b) for the two systems, respectively. Not unexpectedly, the patterns exhibit the same structure with equal number of stripes and segments. To further support

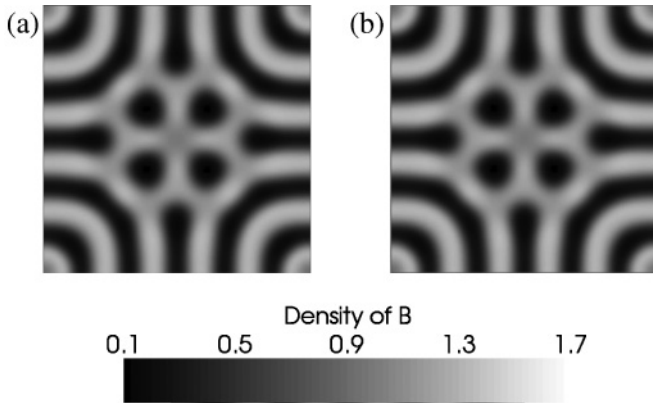


FIG. 12. Turing pattern showing spatial distribution of the density \bar{B}^- at time $t = 400\,000$, $\eta = 2.016\,933$, and $\tau = 2.730\,30$ for two systems of size (a) 200×200 lattice units, (b) 400×400 lattice units. The patterns are clearly identical in the two cases.

this observation, we carried out numerical simulations over a range of system sizes from 50 to 500 lattice units. The thus-obtained density profiles along the line $y/L = 0.85$ are plotted in Fig. 13(a) for all studied system sizes. For the sake of visibility, each individual curve is shifted by a multiple of 2 along the vertical axis. It is clear from the figure that the number of stripes does not change with the system size. To further emphasize the similarity of the patterns, we directly compare in the same figure all the data using the same shift for all the curves. Clearly, the data collapse into a single curve.

As an additional demonstration of wavelength regulation and proportion preservation of the patterns, we perform Fourier transform of the patterns obtained from each system size in the range of 50 to 500 lattice units. We calculate the maximum excited wave number q_{\max} in the Fourier spectra of the density field using Eq. (44). Figure 13(b) shows the plot of the wave number excited with the system size for different diffusion coefficients. It is clear from the plot that the maximum excited wave number q_{\max} decreases in proportion to L and thus the generated pattern is expected to preserve the proportion as observed in our numerical simulation.

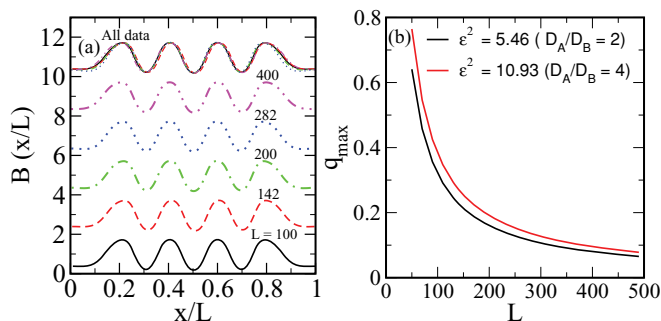


FIG. 13. (Color online) (a) Density profile of \bar{B}^- along the line $y/L = 0.85$, obtained from the Turing pattern in Fig. 12 at time $t = 400\,000$, with parameters $\eta = 2.014$ and $\tau = 2.733$. The number of stripes are invariant over an appreciable range of system size. (b) Characteristic wave number of the Turing pattern in Fig. 12 plotted against the system size for different values of ϵ^2 (realized via a variation of D_A/D_B). The curve shows preservation of proportionality between the wave number and the system size.

V. SUMMARY

In this work, we study reaction-diffusion systems via lattice Boltzmann computer simulations. Starting from the analytical solution of a simple prototypical model (a single species undergoing transformation reaction and diffusion), we perform a systematic study of the lattice Boltzmann truncation error of the model. We uncover interesting behavior of the truncation error with the system parameters. The error is found to have a minimum at a given value of diffusion coefficient. The position of minimum is shifted for increasing values of the reaction rate constant. These observations are in agreement with the analytical findings from a third-order Chapman-Enskog multiscale expansion.

A study of the Gray-Scott reaction-diffusion model is also provided. Here, we perform a linear stability analysis of the model and determine the relevant parameter range for pattern formation. Lattice Boltzmann simulations of this interesting reaction-diffusion system are found to be in good agreement with the predictions of the linear stability analysis. In addition to a test of the linear stability phase diagram, lattice Boltzmann simulations provide valuable information on the details of the patterns formed in different regions of the parameter space. An example is the formation of striped patterns in most parts of the Turing regime above the Hopf bifurcation curve. Another very interesting example is provided by the so-called self-replicating spots, which lie beyond the linear stability regime. Self-replicating spots occur via large amplitude perturbations of the trivial homogeneous solution.

Furthermore, a survey of the parameters entering the scale invariant form of the Gray-Scott model suggests that the simplest choice to tune the length scale of the obtained patterns (while keeping its shape unchanged) is to multiply all the relevant diffusion coefficients with the same constant factor without any modification of the reaction rates. Interestingly, this leaves the time scale of the process unaffected. In other words, in systems with different diffusion coefficients but the same reaction rates, patterns exhibit the same shape (but different sizes) exactly for the same physical time. Results obtained via lattice Boltzmann simulations confirm this behavior. It is noteworthy that this act of regulating diffusion coefficient of species or morphogens, as the case may be, is also observed in some biological systems. This observation is by no means limited to this model—the analysis can also be extended to other reaction-diffusion models.

ACKNOWLEDGMENTS

This work was supported by the Max-Planck Initiative for Multiscale Materials Modeling of Condensed Matter (MMM) and the Interdisciplinary Center for Advanced Material Simulation (ICAMS). We are most grateful to Timm Krüger for useful discussions.

APPENDIX: CHAPMAN-ENSKOG PROCEDURE FOR REACTION-DIFFUSION EQUATION

In this appendix, we derive the macroscopic reaction-diffusion equation from the lattice Boltzmann model. The LB

equation for the reaction-diffusion equation is written as

$$f_{i,s}(\mathbf{x} + \mathbf{e}_i \Delta t, \Delta t + t) - f_{i,s}(\mathbf{x}, t) = \frac{f_{i,s}^{\text{eq}}(\mathbf{x}, t) - f_{i,s}(\mathbf{x}, t)}{\tau_s} + \Delta t w_i R_s. \quad (\text{A1})$$

To obtain a corresponding macroscopic partial differential equation from the finite difference Eq. (A1), we perform a Taylor series expansion of the left hand side of Eq. (A1) and obtain

$$\sum_{n=1}^{\infty} \frac{\Delta t^n}{n!} (\partial_t + e_{i\alpha} \partial_{x_\alpha})^n f_i(\mathbf{x}, t) = \frac{f_{i,s}^{\text{eq}}(\mathbf{x}, t) - f_{i,s}(\mathbf{x}, t)}{\tau_s} + \Delta t w_i R_s. \quad (\text{A2})$$

The Chapman Enskog procedure introduces two time scales, a fast time scale t_1 , associated with convective transport, and a slow time scale t_2 , associated with diffusion. The time derivative is then expanded as

$$\partial_t = \epsilon \partial_t^{(1)} + \epsilon^2 \partial_t^{(2)}. \quad (\text{A3})$$

The spatial derivative is written as

$$\partial_{x_\alpha} = \epsilon \partial_{x_\alpha}^{(1)}. \quad (\text{A4})$$

The equilibrium distribution as a reaction term is expanded as

$$f_{i,s} = f_{i,s}^{(0)} + \epsilon f_{i,s}^{(1)} + \epsilon^2 f_{i,s}^{(2)} + \epsilon^3 f_{i,s}^{(3)} + \mathcal{O}(\epsilon^4), \quad (\text{A5})$$

$$R_s = R_s^{(0)} + \epsilon R_s^{(1)} + \epsilon^2 R_s^{(2)} + \epsilon^3 R_s^{(3)} + \mathcal{O}(\epsilon^4). \quad (\text{A6})$$

Inserting Eqs. (A6), (A5), (A4), and (A3) in Eq. (A2), one obtains

$$\left[\Delta t (\epsilon \partial_t^{(1)} + \epsilon^2 \partial_t^{(2)} + \epsilon e_{i\alpha} \partial_{x_\alpha}^{(1)}) + \frac{\Delta t^2}{2} (\epsilon^2 \partial_t^{(1)} \partial_t^{(1)} + 2\epsilon^2 e_{i\alpha} \partial_t^{(1)} \partial_{x_\alpha}^{(1)} + \epsilon^2 e_{i\alpha} e_{i\beta} \partial_{x_\alpha}^{(1)} \partial_{x_\beta}^{(1)} + 2\epsilon^3 e_{i\alpha} \partial_t^{(2)} \partial_{x_\alpha}^{(1)} + 2\epsilon^3 \partial_t^{(2)} \partial_t^{(1)} + 2\epsilon^4 \partial_t^{(2)} \partial_t^{(2)}) \right] \times [f_{i,s}^{(0)} + \epsilon f_{i,s}^{(1)} + \epsilon^2 f_{i,s}^{(2)} + \mathcal{O}(\epsilon^3)] = \frac{1}{\tau_s} (f_{i,s}^{\text{eq}}(\mathbf{x}, t) - [f_{i,s}^{(0)} + \epsilon f_{i,s}^{(1)} + \epsilon^2 f_{i,s}^{(2)} + \epsilon^3 f_{i,s}^{(3)} + \mathcal{O}(\epsilon^4)]) + \Delta t w_i [\epsilon R_s^{(1)} + \epsilon^2 R_s^{(2)} + \epsilon^3 R_s^{(3)} + \mathcal{O}(\epsilon^4)]. \quad (\text{A7})$$

Grouping terms of the same order in ϵ yields the following successive approximations:

$$\mathcal{O}(\epsilon^0): f_{i,s}^{(0)} = f_{i,s}^{\text{eq}}, \text{ implying that } R_s^{(0)} = 0. \quad (\text{A8})$$

Note that this condition follows directly from the conservation of mass.

$$\mathcal{O}(\epsilon^1): \Delta t (\partial_t^{(1)} + e_{i\alpha} \partial_{x_\alpha}^{(1)}) f_{i,s}^{(0)} = -\frac{1}{\tau_s} f_{i,s}^{(1)} + \Delta t w_i R_s^{(1)}. \quad (\text{A9})$$

On the level of ϵ , there is no mass diffusion; the diffusion process takes place on the scale of ϵ^2 . Furthermore, for diffusion driven reactions, the diffusive flux must bring the species together before the reaction and the reaction becomes a second-order effect. Since on the scale of ϵ , there is no mass diffusion, that implies $R_s^{(1)} = 0$. We would consider this case in this derivation and Eq. (A9) becomes

$$\mathcal{O}(\epsilon^1): \Delta t (\partial_t^{(1)} + e_{i\alpha} \partial_{x_\alpha}^{(1)}) f_{i,s}^{(0)} = -\frac{1}{\tau_s} f_{i,s}^{(1)}. \quad (\text{A10})$$

$$\begin{aligned} \mathcal{O}(\epsilon^2): & \Delta t [\partial_t^{(2)} f_{i,s}^{(0)} + (\partial_t^{(1)} + e_{i\alpha} \partial_{x_\alpha}^{(1)}) f_{i,s}^{(1)}] \\ & + \frac{\Delta t^2}{2} (\partial_t^{(1)2} + 2e_{i\alpha} \partial_t^{(1)} \partial_{x_\alpha}^{(1)} + e_{i\alpha} e_{i\beta} \partial_{x_\alpha}^{(1)} \partial_{x_\beta}^{(1)}) \\ & \times f_{i,s}^{(0)} = -\frac{1}{\tau_s} f_{i,s}^{(2)} + \Delta t w_i R_s^{(2)}. \end{aligned} \quad (\text{A11})$$

$$\begin{aligned} \mathcal{O}(\epsilon^3): & \Delta t [\partial_t^{(3)} f_{i,s}^{(0)} + \partial_t^{(2)} f_{i,s}^{(1)} + (\partial_t^{(1)} + e_{i\alpha} \partial_{x_\alpha}^{(1)}) f_{i,s}^{(2)}] \\ & + \frac{\Delta t^2}{2} (\partial_t^{(1)3} + 2e_{i\alpha} \partial_t^{(1)} \partial_{x_\alpha}^{(1)} + e_{i\alpha} e_{i\beta} \partial_{x_\alpha}^{(1)} \partial_{x_\beta}^{(1)}) f_{i,s}^{(1)} \end{aligned}$$

$$\begin{aligned} & + \Delta t^2 \partial_t^{(2)} (\partial_t^{(1)} + e_{i\alpha} \partial_{x_\alpha}^{(1)}) f_{i,s}^{(0)} + \frac{\Delta t^3}{6} (\partial_t^{(1)} + e_{i\alpha} \partial_{x_\alpha}^{(1)})^3 \\ & \times f_{i,s}^{(0)} = -\frac{1}{\tau_s} f_{i,s}^{(3)} + \Delta t w_i R_s^{(3)}. \end{aligned} \quad (\text{A12})$$

Putting the expression for $f_{i,s}^{(1)}$ from Eq. (A10) into Eq. (A11) yields

$$\begin{aligned} \frac{1}{\tau_s} f_{i,s}^{(2)} = & -\Delta t \partial_t^{(2)} f_{i,s}^{(0)} + \Delta t^2 \left(\tau_s - \frac{1}{2} \right) (\partial_t^{(1)} + e_{i\alpha} \partial_{x_\alpha}^{(1)})^2 \\ & \times f_{i,s}^{(0)} + \Delta t w_i R_s^{(2)}. \end{aligned} \quad (\text{A13})$$

In Eq. (A12), we insert the expression for $f_{i,s}^{(1)}$ and $f_{i,s}^{(2)}$ from Eqs. (A10) and (A11) and obtain

$$\begin{aligned} \frac{1}{\tau_s} f_{i,s}^{(3)} = & -\Delta t \partial_t^{(3)} f_{i,s}^{(0)} + \Delta t^2 (2\tau_s - 1) (\partial_t^{(1)} + e_{i\alpha} \partial_{x_\alpha}^{(1)}) \partial_t^{(2)} f_{i,s}^{(0)} \\ & - \Delta t^3 \left(\tau_s^2 - \tau_s + \frac{1}{6} \right) (\partial_t^{(1)} + e_{i\alpha} \partial_{x_\alpha}^{(1)})^3 \\ & \times f_{i,s}^{(0)} - \tau_s \Delta t^2 (\partial_t^{(1)} + e_{i\alpha} \partial_{x_\alpha}^{(1)}) w_i R_s^{(2)} + \Delta t w_i R_s^{(3)}. \end{aligned} \quad (\text{A14})$$

Next we take the moments of the distribution functions in Eqs. (A9), (A13), and (A14). Note that in order to preserve the isotropy of the lattice tensors, the chosen lattice speeds and weights in the equilibrium distribution function must obey the

following moments or symmetry conditions:

$$\begin{aligned} (a) \sum_i w_i &= 1, & (b) \sum_i w_i e_{i\alpha} &= 0, \\ (c) \sum_i w_i e_{i\alpha} e_{i\beta} &= c_s^2 \delta_{\alpha\beta}, & (d) \sum_i w_i e_{i\alpha} e_{i\beta} e_{i\gamma} &= 0, \\ (e) \sum_i w_i e_{i\alpha} e_{i\beta} e_{i\gamma} e_{i\delta} &= c_s^4 (\delta_{\alpha\beta} \delta_{\gamma\delta} + \delta_{\alpha\gamma} \delta_{\beta\delta} + \delta_{\alpha\delta} \delta_{\beta\gamma}). \end{aligned} \quad (\text{A15})$$

Using Eq. (A15) and given that the local equilibrium takes the form $f_i^{\text{eq}} = f_i^{(0)} = w_i \rho_s$, we impose the following conditions of conservation of mass on the equilibrium distribution function,

$$\sum_i f_{i,s}^{(0)} = \rho_s, \quad \sum_i e_{i\alpha} f_{i,s}^{(0)} = 0, \quad \sum_i e_{i\alpha} e_{i\beta} f_{i,s}^{(0)} = \rho_s c_s^2 \delta_{\alpha\beta}. \quad (\text{A16})$$

We further assume that higher order corrections of the equilibrium distribution do not contribute to the local values of the mass, whereby obtaining

$$\sum_i f_i^{(n)} = 0 \quad \text{for } n \geq 1. \quad (\text{A17})$$

Taking \sum_i of Eq. (A9) and using Eqs. (A15), (A16), and (A17) yields

$$\partial_t^{(1)} \rho_s = 0. \quad (\text{A18})$$

Taking \sum_i Eq. (A13) and using Eqs. (A15), (A16), (A17), and (A18) leads to

$$\partial_t^{(2)} \rho_s = \Delta t c_s^2 \left(\tau_s - \frac{1}{2} \right) \partial_{x_\alpha}^{(1)} \partial_{x_\beta}^{(1)} \rho_s \delta_{\alpha\beta} + R_s^{(2)}. \quad (\text{A19})$$

Taking \sum_i of Eq. (A14) and using Eqs. (A15), (A16), (A17), and (A18) leads to

$$\begin{aligned} \partial_t^{(3)} \rho_s &= -3c_s^2 \Delta t^2 \left(\tau_s^2 - \tau_s + \frac{1}{6} \right) \partial_t^{(1)} \partial_{x_\alpha}^{(1)} \partial_{x_\beta}^{(1)} \rho_s \delta_{\alpha\beta} \\ &\quad - \tau_s \Delta t^2 \partial_t^{(1)} R_s^{(2)} + R_s^{(3)}. \end{aligned} \quad (\text{A20})$$

We multiply Eq. (A18) by ϵ , Eq. (A19) by ϵ^2 , and Eq. (A20) by ϵ^3 and add all this together, thus arriving at

$$\begin{aligned} \partial_t \rho_s &= c_s^2 \Delta t \left(\tau_s - \frac{1}{2} \right) \partial_{x_\alpha}^2 \rho_s + R_s - 3 \Delta t^2 c_s^2 \\ &\quad \times \left(\tau_s^2 - \tau_s + \frac{1}{6} \right) \partial_t \partial_{x_\alpha}^2 \rho_s - \tau_s \Delta t^2 \partial_t R_s. \end{aligned} \quad (\text{A21})$$

We can further re-write Eq. (A21) as the macroscopic reaction-diffusion equation and a third-order truncation error term E ,

$$\partial_t \rho_s = D_s \partial_{x_\alpha}^2 \rho_s + R_s - E, \quad (\text{A22})$$

where the diffusion coefficient is given by $D_s = c_s^2 \Delta t (\tau_s - 0.5)$ and the error term takes the form

$$E = 3 \Delta t^2 c_s^2 \left(\tau_s^2 - \tau_s + \frac{1}{6} \right) \partial_t \partial_{x_\alpha}^2 \rho_s + \tau_s \Delta t^2 \partial_t R_s + \mathcal{O}(\epsilon^4). \quad (\text{A23})$$

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