Lattice-Gas Automata for Coupled Reaction-Diffusion Equations

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We present a lattice-gas automaton approach to coupled reaction-diffusion equations. This approach provides a microscopic basis for exploring systems which exhibit such interesting features as oscillatory behavior and pattern formation. Two-species systems are analyzed in detail. As an applicatin of the formalism, we construct the microscopic dynamics for a system described by the Maginu equations; simulation results show excellent agreement with the phenomenological predictions. Most important is the result showing that we obtain Turing-type structures by a purely microscopic approach.

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Recently, a class of probabilistic cellular automata has been proposed as a microscopic approach to a class of Ginzburg-Landau equations: $\dot{x}=\varphi(x)+\nabla^2 x$, with x real, and where $\varphi(x)$ is a polynomial whose maximum degree is set by the lattice symmetry of the automaton.[1] This class of equations is commonly used for the phenomenological description of dynamical phase transitions with a nonconserved order parameter, and in particular for reaction-diffusion (RD) systems whose dynamics can be reduced to single-species dynamics. The validity of the lattice-gas automata (LGA) approach was shown both theoretically and "experimentally" through numerical simulations.[2] In particular, the application to a specific—but typical—reactive scheme (the Schlögl model) has proved to be fully successful not only in that the automaton model yields the expected phenomenology (bistability, domain formation, etc.) but also in that it provides a microscopic approach which indicates the limits of validity of the phenomenological equation.[2] In this Letter we present a generalization of the LGA approach to a class of coupled RD equations. For the sake of simplicity, here we restrict our analysis to two-species (X,Y) RD systems as described phenomenologically by a set of partial differential equations (PDE's) of the form

$$\partial_t x=\varphi(x,y)+D_x\Delta x,\quad \partial_t y=\phi(x,y)+D_y\Delta y\,. \qquad (1)$$

Such systems are known to exhibit quite interesting features such as oscillatory behavior, phase oscillations, and patterns which are considered to model Turing structures,[3] which have been recently observed experimentally.[4] So far no theoretical microscopic analysis has been constructed which displays all such features at the macroscopic level.[5] The purpose of the present Letter is to show that a class of LGA achieves this goal.

A typical reaction-diffusion system, in general, involves several reactive species. However, most interesting phenomena occur when the system is constrained far from equilibrium,[3] which can be realized by imposing constant concentration values to all species except a few whose dynamics is described by a set of equations of the type (1). As a result an automaton can be constructed by restricting the microdynamics solely to the unconstrained species. The physical space of a LGA is a regular lattice where particles undergo one-dimensional displacements from node to nearest-neighbor node in one time step. So the space and time variables are discrete as well as the velocities, which are unit vectors oriented along the links connecting neighboring nodes. We define a cell as a connecting link, and the number of cells associated to a node is set by the lattice symmetry. In general (and mostly for computational convenience) an exclusion principle is imposed such that each node has the same limited number of possible states; the elementary case is the single-species model where the exclusion principle restricts the occupation of a cell to one particle, i.e., the state of the cell takes the value 0 (unoccupied) or 1 (occupied by a particle). Diffusion of particles is realized by a stochastic procedure that makes the particles undergo random displacements. The automaton realization is performed by random rotations on the nodes of the lattice: Such random deflections can be viewed as elastic collisions of particles with ghost particles of a virtual substrate (a momentum reservoir). Between these elastic collisions, particles propagate from node to node by discrete time steps in the direction of their velocity vector. As a result of the successive sequences of propagation followed by deflection, particles undergo random walks on the lattice as described in the continuous limit by a diffusion equation. Reactive collisions generate creation or annihilation of particles when particles meet at a node; such collisions must be governed by the kinetics of the considered reaction scheme, as described in the continuous limit by a rate equation.[2] Consequently, the updating rule which yields the configuration field at time $t+1$ as a function of the state at time t is given in terms of three basic operators: P (propagation), R (rotation), and C (collisional reactive transformation). So, the evolution operator can be the product $C\circ R\circ P$, but alternative combinations such as $C\circ(R\circ P^m)^n$ are equally possible. Probabilistic rules for the automaton are important in that they offer the desired flexibility for tuning the kinetic rate constants and for adjusting the diffusion

coefficients. How this is realized will be developed below for two-species systems.

In order to allow for the coexistence of several species on a lattice, the single-species exclusion principle must be extended. For two-species models the exclusion principle can be established in two ways.

(i) The single-lattice model.—The exclusion principle applies to all particles regardless of the species. Then each cell on the lattice has three possible states: unoccupied, occupied by an X particle, or occupied by a Y particle.

(ii) The coupled-lattice model.—The exclusion principle applies to the two species independently. Two particles can be simultaneously in the same cell, provided they belong to different species. Then the Universe can be viewed as the superposition of two coupled single-species lattices; each lattice being subject to an exclusion principle is described by a Boolean field (one bit per cell).[6]

How is the updating rule generalized for these models? We first consider the single-lattice model. The generalization of the propagation step is straightforward: Each particle moves in the direction of its velocity vector to the nearest-neighbor node. During this step the velocity and the species of the particle are conserved (only position changes). The generalization of the rotation operation R is also quite logical: The rotation operator maps each node configuration onto a new configuration independently and simultaneously at each lattice node, according to a stochastic rule which conserves the number of particles for each species (only velocities change).[7] The reactive transformation C operates independently at each node of the lattice where particles are randomly created or annihilated through reactive collisions of the type

$$\alpha X+\beta Y\rightarrow \alpha' X+\beta' Y\,, \tag{2}$$

with net reaction probabilities $P_{\alpha\beta;\alpha'\beta'}$. The occurrence of a reaction is independent of the velocity configuration at the node, and the velocity distribution is only modified by particles that are created or destroyed; creation (annihilation) occurs with equal probability on all empty (occupied) cells. The allowed reactions are restricted by the exclusion principle, i.e., the following relation holds for the indices of the transition probability matrix **P**:

$$\alpha,\beta,(\alpha+\beta),\alpha',\beta',(\alpha'+\beta')\in(0,\ldots,b)\,, \tag{3}$$

where b is the total number of cells per node.

The coupled-lattice model offers more flexibility in the generalization of the dynamics: (i) Since propagation and rotation take place on separate lattices for each species, one can tune the respective diffusion coefficients independently by applying the propagation operator with different frequencies to each species. This procedure is not applicable to the single-lattice model. (ii) The coupled-lattice rotation operator produces mixing independently in each lattice. Therefore different rotation operators (one for each species) can be used to obtain different diffusion coefficients for each species. Note that the strict exclusion principle precludes separate mixing in the single-lattice model; however, if the mixing operation is node-configuration dependent, it is possible to obtain different diffusion coefficients with the single-lattice model, but the diffusion coefficients will then become concentration dependent.

In the coupled-lattice model the reactive transformation C applies to pairs of colocated nodes (one on each lattice). Particles are created or destroyed randomly in their respective lattices according to the joint configuration of the two corresponding nodes. The occurrence of a reaction depends on the number of particles (α and β) and is independent of the velocity configuration of the pair of nodes, and the velocity distribution is only modified by particles that are created or destroyed. The reactive transformation operator is determined by a transition probability matrix $P_{\alpha\beta;\alpha'\beta'}$ corresponding to reactions of the type given by Eq. (2), and the allowed reactions are restricted by the coupled-lattice exclusion principle:

$$\alpha,\beta,\alpha',\beta'\in(0,\ldots,b)\,. \tag{4}$$

Having defined the automaton rules, we address the question: What kind of physics can we expect from these rules? We will not give a statistical-mechanical treatment, which will be presented elsewhere as an extension of the theory developed earlier;[2] rather we present a phenomenological analysis to emphasize physical aspects. For reasons that should be clear from the above discussion, here we consider the coupled-lattice model which has more operational flexibility.

If the time between two reactive events is much larger than the time between two mixing operations, it is reasonable to assume that the reactions occur at local diffusive equilibrium. Under this assumption, the average number of particles per cell (i.e., the probability of finding a cell occupied) is cell independent. Consequently the occupation probabilities to be considered are $x(\mathbf{r})$ and $y(\mathbf{r})$, the local densities per cell at node $\mathbf{r}$ of species X and Y, respectively. Under the local-equilibrium assumption, the probability that α X particles and β Y particles be simultaneously at a node factorizes into two binomials; therefore the probability of occurrence of reaction (2) is given by

$$[C_b^\alpha x^\alpha(1-x)^{b-\alpha}][C_b^\beta y^\beta(1-y)^{b-\alpha}][P_{\alpha\beta;\alpha'\beta'}]\,. \tag{5}$$

Taking into account that density variations are due to diffusive transport and reactive processes, we expect that the macroscopic behavior of the automaton should be described by a set of coupled PDE's (1) with reaction rates

$$\begin{aligned}\varphi(x,y)&=(1/b)\sum_{\alpha\beta;\alpha'\beta'}(\alpha'-\alpha)[C_b^\alpha x^\alpha(1-x)^{b-\alpha}]\\&\quad\times[C_b^\beta y^\beta(1-y)^{b-\alpha}]P_{\alpha\beta;\alpha'\beta'}\,,\\ \phi(x,y)&=(1/b)\sum_{\alpha\beta;\alpha'\beta'}(\beta'-\beta)[C_b^\alpha x^\alpha(1-x)^{b-\alpha}]\\&\quad\times[C_b^\beta y^\beta(1-y)^{b-\alpha}]P_{\alpha\beta;\alpha'\beta'}\,.\end{aligned} \tag{6}$$

In Eqs. (6) the factor $1/b$ is introduced so that the rates are expressed in cell units.

Now we address the inverse problem: Given a set of phenomenological rates $\phi(x,y)$ and $\varphi(x,y)$, Eq. (1), do $P_{\alpha\beta;\alpha'\beta'}$'s exist such that these rates identify with the automaton rates (6)? As the polynomials $x^{\alpha}(1-x)^{b-\alpha} \times y^{\beta}(1-y)^{b-\beta}$ $(\alpha,\beta=1...b)$, in (6), form a basis for polynomials in x and y with degree less than or equal to b in x and in y separately, a necessary condition is that the phenomenological rates belong to that ensemble, i.e.,

$$\varphi(x,y)=\sum_{k=0}^{b}\sum_{l=0}^{b}A_{k,l}x^k y^l,$$
$$\phi(x,y)=\sum_{k=0}^{b}\sum_{l=0}^{b}B_{k,l}x^k y^l. \tag{7}$$

If so, then in principle, the RD equation (1) can be constructed with the automaton by establishing the relationship between the coefficients $A_{k,l}$ and $B_{k,l}$ and the transition probability matrix elements $P_{\alpha,\beta;\alpha',\beta'}$. The total number of elements is $(b+1)^4$, with $(b+1)^2$ constraints,

$$\sum_{\alpha',\beta'=0}^{4}P_{\alpha\beta;\alpha',\beta'}=1, \tag{8}$$

while there are $2(b+1)^2$ coefficients $A_{k,l},B_{k,l}$. As a result one has $(b+1)^4-3(b+1)^2$ degrees of freedom which, in principle, provides considerable flexibility to set up a class of automata for a given set of RD equations. However, it follows from the positivity condition

$$0\le P_{\alpha,\beta;\alpha',\beta'}\le 1 \tag{9}$$

that not all reaction rates are realizable with the automaton. For instance, if concentration x takes its upper limit $(=1)$, the reactive rate φ cannot be positive. Similar necessary conditions on the reaction rates are obtained for $x=0$, $y=0$, $y=1$. In fact these necessary conditions are also sufficient in the following sense: If the reaction rates are such that the flow $\dot{x}=\varphi(x,y)$, $\dot{y}=\phi(x,y)$ is confined to the phase-space domain $[0,1]\times[0,1]$, i.e.,

$$\varphi(0,y)\ge 0,\quad \varphi(1,y)\le 0,$$
$$\phi(x,0)\ge 0,\quad \phi(x,1)\le 0, \tag{10}$$

then there exists a constant s such that the rescaled rates φ/s and ϕ/s are realizable with the cellular automaton rule. As the factor s can be absorbed in a scaling of time, all reactive rates that obey (10) can be investigated with the automaton.

We have applied the coupled-lattice formalism to a set of RD equations known as the Maginu model[8] which shows oscillatory behavior and Turing-type structures:

$$\partial_t x=x-x^3/3-y+D_x\Delta x,$$
$$\partial_t y=(x-ky)/c+D_y\Delta y, \tag{11}$$

with $c>0$ and $0<k<1$. As is, this set of equations does not fulfill requirements (10). However, for all values of the parameters c and k there exists at least one linear transformation of x and y which maps the set of equations (11) onto an admissible set,[9] e.g.,

$$x'=\tfrac{1}{2}+x/\sqrt{12(1+1/k)},$$
$$y'=\tfrac{1}{2}+yk/\sqrt{12(1+1/k)}, \tag{12}$$

which is valid for all values of c and for all $k>\frac{1}{2}$.

For the computational realization of the Maginu model, Eqs. (12), we have chosen to restrict the set of transition probabilities by supposing additional (although optional) requirements: (i) Solely reactions with single-particle change for each species are considered, i.e., in Eq. (2), $(\alpha-\alpha'),(\beta-\beta')\in(0,\pm 1)$; (ii) for any number of particle configuration $(\alpha X,\beta Y)$, creation and annihilation transitions are exclusive within each species, i.e., $(\alpha-\alpha')$ is either ≥ 0 or ≤ 0, and similarly for $(\beta-\beta')$; and (iii) $(\alpha-\alpha')$ and $(\beta-\beta')$ are stochastically independent. These constraints combined with (8) determine uniquely the set of $P_{\alpha\beta;\alpha'\beta'}$, and (9) sets the minimum value of the scaling factor s. The rotation operator R_L (the index $L=X,Y$ refers to the species lattice) acts on each node such that its configuration is rigidly rotated by $\pi/2$, π, or $3\pi/2$, with equal probabilities q_L, and is left unchanged with probability $1-3q_L$. The evolution operator can be written symbolically as $C\circ(R_L\circ P_L)^{n_L}$. The exponent n_L governs the diffusion mechanism for

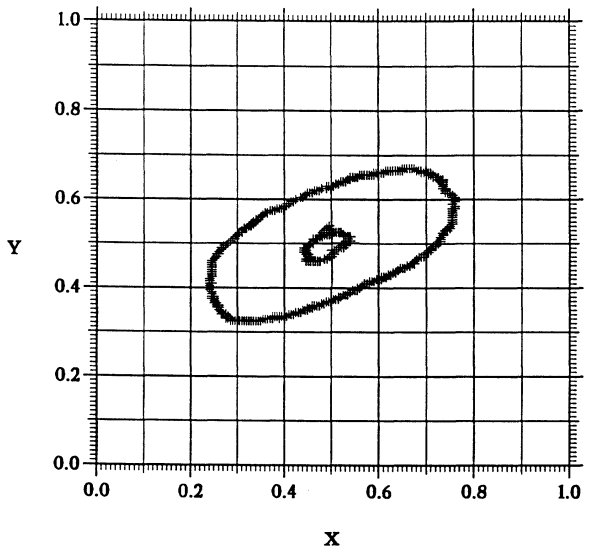

FIG. 1. Limit cycle (outer ring) obtained from the LGA simulation of the Maginu model. Parameter values: $k=0.9$, $c=2$, $s=10$, $q_x=q_y=1$, $n_x=n_y=1$; lattice size: 64×64 nodes. The inner ring shows the shrinking effect when the ratio of reactive collisions to elastic collisions is increased; same parameter values, except $s=2$.

each species: Between two reactive collisions, the X (Y) particles undergo n_X (n_Y) propagation-rotation sequences.

The simulations were performed on a square lattice with periodic boundary conditions; values of relevant parameters are given in the figure captions. Two essential results will be described here. Oscillatory behavior is illustrated in Fig. 1 which shows the limit cycle obtained by averaging the density over the whole system. The phase-space trajectory and the value of the frequency are in quantitative agreement with the predictions from the Maginu equations, Eqs. (12). Now, when the diffusion coefficients are lowered, intrinsic fluctuations play a crucial role: Spatial phase coherence can no longer be maintained by diffusion over the whole system and as a result one observes (after spatial averaging) a dramatic shrinking of the limit cycle, as also shown in Fig. 1. The second most important result is the observation of Turing-type structures. Linear stability analysis of the Maginu equations shows that the homogeneous steady state can become unstable by spatial destabilization when $(D_x/D_y)^{1/2} \leq (1-\sqrt{1-k})\sqrt{c}/k$ for $0<c<k$.[10] Under these conditions, the automaton, when prepared initially in the homogeneous steady state [according to the rate equations (6)] destabilizes and develops worm-like spatial structures (Fig. 2) which we conjecture to be Turing structures on the basis of the following observations: (i) the patterns are quasisteady and are morphologically equivalent to the pattern obtained from the numerical simulations of the phenomenological equations in the same regime; (ii) the characteristic wavelength of the observed pattern is independent of the size of the system and has the correct order of magnitude, i.e, as predicted from Eqs. (11); and (iii) destabilization of the homogeneous steady state occurs only if the values of the diffusion coefficients of the two species differ sufficiently from each other. While some points remain to be explored—in particular, the role of intrinsic fluctuations versus pattern selection mechanism—the present work shows that LGA provide a valuable microscopic approach to the investigation of the spatiotemporal behavior of RD systems.[11]

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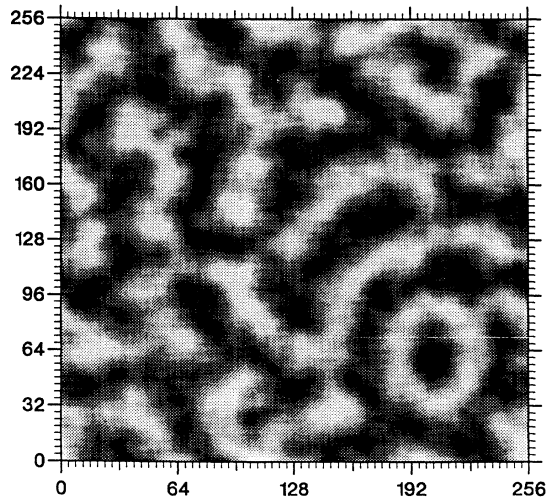

FIG. 2. Turing-type pattern obtained from the destabilization of the homogeneous unstable steady state. The concentration ranges from $x>0.72$ (dark) to $x<0.28$ (light). Parameter values: $k=0.9$, $c=0.45$, $s=10$, $q_x=1$, $q_y=0.6429$, $n_x=1$, $n_y=9$; $D_x=0.25$, $D_y=4.75$; lattice size: 256×256 nodes.

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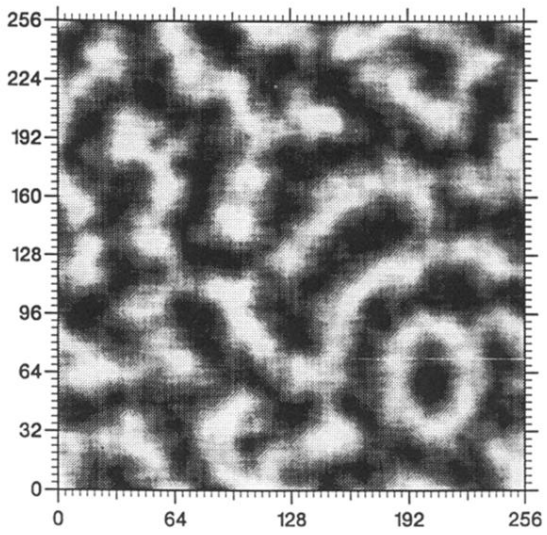

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