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Discrete Model of Chemical Turbulence

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Presumably the simplest model of chemical turbulence is proposed, for which the spatial degree of freedom plays an important role. The concentration, as well as space and time, is discretized. As in the real experiment a turbulent phase is found sandwiched by two ordered phases. In addition, the model predicts a solitonlike phase. Thus the model can exhibit a variety of behaviors encountered in partial-differential-equation systems. It is also closely related to cellular automata.

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In many physical and chemical systems most interesting phenomena such as pattern formation, turbulence, etc., involve both the spatial and temporal degrees of freedom. The recent reviving interest in the collective behavior of coupled simple subsystems stems from the quest for the role of spatial degrees of freedom. Typical examples being studied are coupled nonlinear oscillators,¹ coupled nonlinear maps,² and cellular automata.³ These examples are closely related to nonlinear partial differential equations (PDE) which are often more realistic models for physical systems. In these examples and PDE systems the coexistence of chaos and ordered coherent structure (order in turbulent states) has been given much attention.⁴

For the coupled limit cycles, Kuramoto, Yamada, and other researchers have been conducting extensive studies for more than ten years.^{5,6} One of the most important outcomes is the concept of chemical turbulence. On the basis of the Kuramoto-Yamada-Sivashinsky equation, they predicted the existence of a turbulent phase due to instability induced by diffusion. Notice that this turbulence is conceptually different from the so-called chemical turbulence studied by

Hudson, Roux, and others.⁷ In the latter case, reagents are completely stirred, so that there is no spatial degree of freedom.

Following the Kuramoto-Yamada prediction, Yamazaki, Oono, and Hirakawa⁸ performed an experimental study of chemical turbulence using the Belousov-Zhabotinsky reaction.⁹ In their experiments the regime of the reaction was so chosen that there was purely periodic oscillation when the solution was stirred well. Thus, if a chaotic phenomenon was observed in a vessel without any stirring, it was solely due to the existence of the spatial inhomogeneity. In contrast to experiments in a stirred reactor,⁷ the experiments by Yamazaki, Oono, and Hirakawa had difficulties due to the lack of stirring; carbon dioxide bubbles generated by the reaction had to be kept on the wall of the vessel, possible convective flow induced by the exothermic reaction had to be suppressed by a slight temperature gradient, etc. The rough phase diagram constructed experimentally exhibits a disordered regime characterized by rather abrupt changes of frequencies and phases of the concentration oscillation. The main feature of the diagram was that there

seemed to be a nonturbulent regime on both sides of the turbulent region along the temperature axis. Since the frequency of the reaction is much more sensitive to the temperature than the diffusion rates, the higher the temperature, the less effective is the diffusion.

The main purpose of the present Letter is to propose presumably the simplest possible model of chemical turbulence. The model is in a class of models whose elements are discrete-state oscillators. We show the existence of a large variety of behaviors in this model: ordered phases, turbulence, solitons, etc. It seems that the model proposed here can exhibit the whole array of behaviors exhibited by (dissipative) PDE sys-

tems in general.

The essence of chemical turbulence is the linear coupling of spatially distributed nonlinear oscillators. The simplest way to represent a cyclic chemical oscillation is to use discrete concentration levels. It has turned out that to have nontrivial behaviors three discrete levels, M , 1, and 0, are sufficient, where M is a positive integer. We also use discrete time and space. Hence, in our simplest model at a given discrete time, one of the discrete concentrations M , 1, and 0 is assigned to each spatial cell. The rule of time evolution must contain the effects of both the spatial coupling and oscillation. Specifically, the rule of our cellular model in one spatial dimension is given by

$$A'(n,t) = \alpha[A(n+1,t) + A(n-1,t)]/2 + (1-\alpha)A(n,t), \quad (1)$$

and

$$A(n,t+1) = F(A'(n,t)), \quad (2)$$

where $A(n,t)$ is the concentration at the n th cell at time t , $\alpha \in [0,1]$ is the spatial coupling constant, and the function F is given by

$$F(x) = \begin{cases} 1, & \text{if } 1.5 \leq x, \\ 0, & \text{if } 0.5 \leq x < 1.5, \\ M, & \text{if } x < 0.5. \end{cases} \quad (3)$$

The diffusion-type spatial coupling is represented by (1), where the parameter α may be regarded as the strength of diffusion. On the other hand, (2) with (3) describes cyclic oscillation; when there is no diffusion, i.e., $\alpha=0$, each cell makes an intrinsic cycle of 0- M -1-0: This even mimics the time asymmetry of real concentration oscillation in the Belousov-Zhabotinsky reaction. We designate this rule as 0 M 10. We can make many variants of this rule, but this is the simplest nontrivial one; the two-state 0 M 0 rule gives the simplest oscillating model, but this gives us only ordered phases.

The phase diagram for the 0 M 10 model is shown in Fig. 1 and typical behaviors of spatiotemporal patterns are shown in Fig. 2. Both are with stochastic initial conditions. For large values of M one sees four different phases: three-cycle (3 phase), turbulent (T phase), solitonlike (S phase), and second three-cycle (3' phase) in this order as α is increased. The region X is actually divided into many phases, many of which are phases with several different cycles coexisting. There are also some solitonlike phases. Since the maximum concentration M is comparable to the other concentration levels, 0 and 1, the interplay of the spatial coupling and oscillation becomes complex in X. Furthermore, real experiments were never conducted in the regime where the amplitude of the oscillation is small. Hence from here on we focus on large values of M and the four phases mentioned above (3, T, S,

and 3' phases); we expect that phase diagrams with ordered phases separated by turbulent and solitonlike phases would be general features of chemical turbulence.

Our working definition of the turbulent phase is as follows. The n th cell is said to be unpredictable if and only if the Kolmogorov-Sinai entropy¹⁰ of the sequence $\{A(n,t)\}_{t=0}^{\infty}$ of the discrete concentration levels at the cell n is positive. If the majority of the cells are unpredictable, the phase is said to be nontrivial. If the information in the initial concentration distribution is significantly preserved, we say that the nontrivial phase is turbulent.

Although an attempt to estimate the true Kolmogorov-Sinai entropy (or the Shannon entropy

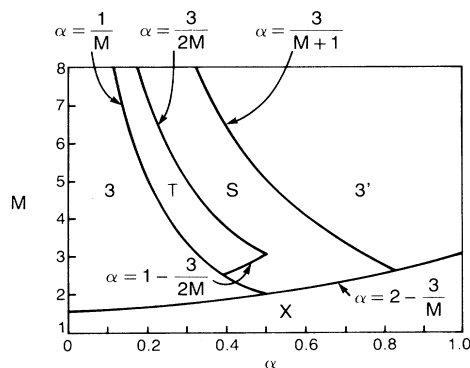


FIG. 1. The phase diagram for the 0 M 10 chemical turbulence model defined by Eqs. (1)–(3). M is the peak value of the autonomous oscillation, which is extended to real values by an obvious modification of the rule, and α is the spatial coupling constant. The symbols 3, T, S, and 3' denote, respectively, a periodic phase with period three, the turbulent phase, the solitonlike phase, and a different periodic phase with period three. The letter X collectively denotes various phases which are not discussed in the text.

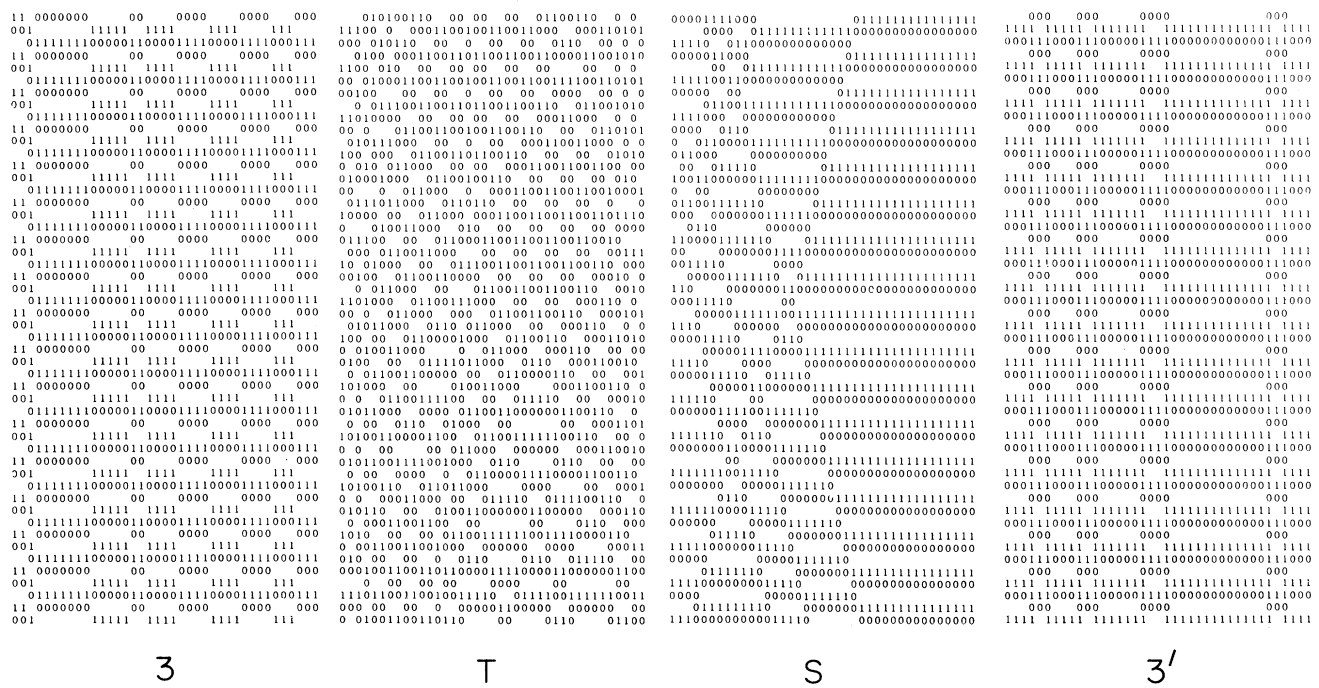


FIG. 2. Typical spatiotemporal patterns of the OM10 model. The time flows from top to bottom of the figures. The horizontal direction is the spatial coordinate. An empty cell actually contains M . The symbols 3, T, S, and $3'$ denote respectively a periodic phase with period three, the turbulent phase, the solitonlike phase, and a different periodic phase with period three.

per letter¹¹ if we regard the state sequence as a sentence) is being made, here, for a preliminary study, the Markov entropy of the sequence,¹² which is the Kolmogorov-Sinai entropy of the empirical simple Markov process obtained from the statistics of the observed sequences, is used to measure the disorder of the state sequence at a given cell. The spatial average of this entropy is denoted by S_t . The spatial disorder is measured by the Markov entropy of the sequence of states along the spatial axis: This entropy is denoted by S_s . Since our system has only three states, the upper bound of S_t and S_s is $\log_2 3 = 1.58496 \dots$ bits. In addition, as a result of the severe restriction on the time sequence imposed by the structure of F , S_t is likely to be less than 0.7 bit. Thus asymptotic values of $S_t \approx 0.5$ bit and $S_s \approx 1.2$ bits specify the turbulent phase; $0 < S_t \leq 0.3$ (bit) and $S_s \approx 0.7$ bit specify the soliton phase; and $S_t = 0$ specifies the periodic ordered phase. Since the number of solitons and their directions of motion strongly depend on the initial condition, so does the value of S_t in the solitonlike phase.

Notice that our system is completely deterministic, so that the temporal disorder is possible only if the (spatial) information in the initial condition is converted to temporal information. This situation is exactly the same as chaos in iterative maps. As an objective of statistical mechanics, such discrete models and cellu-

lar automata can be regarded as measure-theoretic dynamical systems. It is well known that any measure-theoretic dynamical system (with finite entropy) is isomorphic to a shift dynamical system with a finite number of symbols.¹³

The boundaries of the four phases correspond to changes of behaviors of fundamental configurations. Consider a wall which divides two ordered spatial domains. The spatial coupling is not strong enough if $\alpha < 1/M$, so that the two domains do not interact, and the wall cannot move. This parameter region corresponds to the ordered 3 phase. If $1/M \leq \alpha < 3/(M+1)$, then the wall can move. Suppose we have a domain with M 's (M domain) and a domain with 0's (0 domain) at a given time. The 0 domain invades the M domain and the wall between them moves by one spatial unit after three time steps. This parameter region contains the T and the S phases. If $\alpha \geq 3/(M+1)$, the coupling is too strong, so that the wall again cannot move. Notice that although the 0 domain invades the M domain at one time, the 0 domain changes to an M domain and the M domain becomes a 1 domain at the next time step. Now the domain which originally was the 0 domain is invaded by the other domain so that the wall does not move after all. Finally, the boundary of the T and the S phase is related to the existence or nonexistence of self-

organization. Consider an impurity in an M domain. If $\alpha < 3/2M$, then the impurity grows. This region, $1/M \leq \alpha < 3/2M$, corresponds to the T phase. On the other hand, if $\alpha \leq 3/2M$, the impurity disappears and the medium becomes completely ordered. Therefore, in the S phase, $3/2M \leq \alpha < 3/(M+1)$, an initial random spatial configuration is self-organized to ordered domains. The domain walls behave like solitons as seen in Fig. 2, S phase.

In the real experiment,⁹ only the time sequence at one space point was observed. Therefore, the experiment was not designed to discriminate soliton states from turbulent states. We expect solitonlike propagation of the spatial patterns in real experiments of chemical turbulence. Indeed, we see a suggestive numerical result in Kuramoto's work.¹⁴

If one more state, 2, is added to the model with the corresponding modification of F in Eq. (2) (0M210 model), we clearly observe the intermittent structure in a turbulent phase. We also have an additional phase where chaotic behavior seems to be induced by the existence of very many solitons. Thus there can be a turbulent phase which may be understood in terms of nonlinear modes.

The crucial difference between our model and the model for excitable media,¹⁵ which is also related to the Belousov-Zhabotinsky reaction, is the nonexistence of the quiescent state in our model. In the excitable-medium model the state 0 stays indefinitely at 0 unless at least one M state appears in its neighborhood. There is no turbulent phase in this model (at least in one-space). Although there exist solitary propagating waves, they disappear upon collisions. Hence, these waves are distinctly different from the solitons in our model where they go through each other upon collisions. It is easy to make interpolative models of this excitable-medium model and our 0M10 model; symbolically we denote them by the 0M10...0 model. According to our preliminary study, increasing the period diminishes the width of the turbulent region in the phase diagram.

As is mentioned above, our rule does not allow the existence of the quiescent state. Thus as a "cellular automaton," our rule may be said to be illegal.³ However, for any illegal rule, we can make an equivalent legal rule which allows a quiescent state by suitably redefining the time step and the cell neighborhood. Hence, there is a "legal" cellular automaton which is equivalent to the 0M10 model. However, our rule, the combination of the autonomous periodic rule and the diffusionlike linear coupling, is a very special one, so that there is an intriguing question on the algorithmic properties of the model, for example, the universality as an automaton.

In conclusion, we have proposed, presumably, the simplest nontrivial model of chemical turbulence (or

coupled limit cycles) which can exhibit many phenomena that we can observe in nonlinear partial differential equations: turbulence, solitons, intermittency, etc. We believe that it is promising to study such a minimal model in order to understand the complex behavior of real partial-differential-equation systems.

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