Global oscillation mechanism in the stochastic Lotka model

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The microscopic one-parameter kinetic model of the oscillatory $A+B\rightarrow 2\,B$ reaction (Lotka model) is studied using direct Monte Carlo simulations and analytical methods. Percolation is proposed as the mechanism of global oscillations that are not limited to any finite size of a system. An analytical estimate of the oscillation frequency is derived and compared to computer simulations. We also observe the transition from synchronized oscillations to specific f^{-2} noise in two dimensions which was previously reported for self-organized critical models.

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I. INTRODUCTION

The necessity for research on surface reaction systems is dictated by their enormous practical importance for heterogeneous catalysis, as well as by many complex and fascinating nonlinear phenomena they exhibit, such as spatial and temporal oscillations, kinetic phase transitions, pattern formation, chaotic behavior, etc. A comprehensive review of experimental results in this area is given in Ref. [1].

In recent years, significant progress has been achieved for stochastic models of surface reaction systems, which allow one to study in great detail the mechanisms responsible for the above-mentioned phenomena [2–5]. In these models elementary reaction steps, such as adsoption, reaction, and desorbtion, are represented by a set of stochastic rules that describe possible changes in the system configuration. The transition probabilities can be related to reaction rates by considering the master equation of the process [3].

One of the advantages of these models is their simplicity and the possibility of straightforward simulations. However, due to the diversity of reactions taking place in real systems, the number of parameters is usually too large for a complete analysis. Therefore *simplified* model systems are of particular interest. Such models with a minimum number of parameters can represent, however, the essential features of a real system.

The simplest stochastic model possessing temporal oscillations is a Lotka-type lattice model introduced in Ref. [6]. There are two kinds of particles in the Lotka model, say, A and B. Particle A adsorbs on the lattice with rate ζ, B desorbs with rate $1-\zeta$, and the only bimolecular reaction is the infinitely fast autocatalytic conversion $A+B\rightarrow 2\,B$. Mai *et al.* [6] compared the results given by different analytical approaches to the results of Monte Carlo (MC) computer simulations, which appear to be the most reliable. The system shows two different kinetic states: highly regular temporal oscillations of global particle densities and a steady state with nearly constant global coverages. Concentration oscillations appear for small values of the control parameter ζ. Similar simulations in the Lotka model were observed in the reviews by Jansen [7] and Nieminen and Jansen [8], who also reported oscillations. The role of the space dimension was studied by Hovi *et al.* [9]: oscillations were found in two and three, but not in one, dimension. As was shown recently [10], forced periodic perturbation of the control parameter in the Lotka model leads to a series of *resonance phenomena*. On the contrary, the recently studied [11] Lotka-Volterra lattice model has more parameters and shows less unpredictable behavior than the model under consideration.

The Lotka model is of specific interest in several aspects. First, the question how local oscillations in different areas of the surface become coherent is the subject of wide discussion in the literature on surface reaction systems. The existence of globally synchronized oscillations not limited to any finite range is unique for such a simple model as the Lotka model. More complex systems [3,4], on the contrary, exhibit oscillatory behavior only up to a certain size of the system. In these systems the mechanism leading to a synchronization is particle diffusion with a finite rate. Diffusion usually brings synchronization only up to some finite scale which depends on the diffusion constant [4,12]. Only the latest research [13] showed the principal possibility of the amplification of local oscillations in a system with diffusion (by the so-called *stochastic resonance*), thus transforming them into global oscillations in a narrow range of the system parameters. In the Lotka model we have the seemingly more trivial possibility of global synchronization via a chain of infinitely fast reactions $A+B\rightarrow 2\,B$. However, the condition of an infinite reaction rate is not enough for synchronization because both oscillating and steady states are observed in the Lotka model depending on the parameter ζ. A study of the transition between these two regimes has not been done yet and it is one of the tasks in the present study.

The second reason of interest in the Lotka model is its critical behavior which was studied numerically by Hovi *et al.* [9]. They have found a power law dependence in the size distribution of the reaction avalanches with the cutoff size diverging in the limit $\zeta\rightarrow\zeta_c=0$. This fact and some additional features we report on in the present paper (such as $1/f$ noise) are common to forest-fire models of self-organized criticality [14]. Our findings suggest that the criticality of the Lotka model plays a significant role in synchronization.

In this paper, we compare the Lotka model to the well-studied site percolation problem on a discrete lattice. We discuss a special synchronization mechanism—percolation mechanism—and propose an analytical solution based on a simplified analysis of kinetic equations. The analysis is valid

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for the coherently oscillating state near the critical point ζ_c. Analytical expressions of the oscillation frequency are then obtained and tested by MC simulations on different regular two-dimensional lattices. In addition, the transition to the steady state is studied in particular detail. Analysis of the global concentration power spectra reveals a power law f^{-2} in the continuous part of the spectrum. Comparison of different data enables us to draw a qualitative picture of oscillations that connects synchronization on an arbitrary large scale to the criticality of the Lotka model.

The structure of the paper is the following. Section II gives an exact model definition and parameter descriptions. Besides the standard Lotka model, we consider also the possible additional $A \rightarrow B$ reaction. Details of MC simulations and power spectra analysis are discussed in Sec. III. The key section is Sec. IV, in which we introduce a simplified oscillation scheme based on the percolation mechanism and obtain an analytical expression for the oscillation frequency. Simulation results are reported in Sec. V and discussed in connection with analytical results and criticality in Sec. VI. A brief summary in Sec. VII completes the paper.

II. MODEL DESCRIPTION

The Lotka model can be interpreted in terms of heterogeneous catalysis as the following set of reactions between two kinds of particles A and B and a vacant site $*$:

$$A(\mathrm{gas}) + * \xrightarrow{\zeta} A(\mathrm{ads}), \tag{1}$$

$$B(\mathrm{ads}) \xrightarrow{1-\zeta} B(\mathrm{gas}) + *, \tag{2}$$

$$A(\mathrm{ads}) + B(\mathrm{ads}) \xrightarrow{\infty} 2\,B(\mathrm{ads}). \tag{3}$$

Time is rescaled in a way that the only control parameter is the adsorbtion rate ζ for particles A varying from 0 to 1. This condition sets the natural dimensionless rate, frequency, and time units for the model. The autocatalytic reaction in Eq. (3) takes place every time when A and B occur as nearest neighbors.

Interpretation of the autocatalytic reaction [Eq. (3)] in the Lotka model can be different. One possibility is to consider A as B plus an additional ligand that desorbs immediately if A and B come into contact. More evident is the biological interpretation of infection spreading [15]. If healthy species (A) come into contact with infected ones (B), then A also become infected, Eq. (2). Equations (1) and (3) represent the birth of the healthy and death of the infected species, correspondingly.

The subsequent stages of oscillations for small values of ζ can be understood as follows: (i) immortal healthy species A are born with a constant rate ζ [see Eq. (1)]; (ii) occasional contact with already infected organisms immediately infects the whole cluster, thus turning all A's in it into B's [see Eq. (3)]; (iii) infected species B die out according to Eq. (2), providing a free substrate for the birth of new A's.

Nontrivial behavior is possible only if some particles B are initially present in the system, because there is no source of B in the Lotka model based on Eqs. (1)–(3). The absence of such a source brings a principal instability of oscillations on *finite* lattices. If occasionally all particles B desorb from the surface, then the autocatalytic reaction is stopped and the lattice becomes poisoned by particles A.

The nonzero probability of such a ''catastrophe'' poses methodical difficulties for the numerical study of oscillations in the case of large B desorbtion rates (small ζ's). In order to avoid use of extra large lattices or limited simulation times one has to modify the model. One of the possibilities [9] is the so-called constant B-coverage ensemble method. However, this technique does not provide the correct time dependence, which is essential to our study. Instead of modifying simulation algorithm, we consider a weak source of B:

$$A(\mathrm{ads}) \xrightarrow{\gamma} B(\mathrm{ads}). \tag{4}$$

The new reaction should be understood as a weak noise of particles B that could be present in model situations (bond weakening fluctuation on a catalyst surface or spontaneous infection in biological interpretation). In all cases when this additional reaction is switched on, the new *small* parameter γ lies in interval $(10^{-7}$–$10^{-5})$ and condition $\gamma \ll \zeta$ is satisfied.

The main macroscopic observables of the model are global particle concentrations (coverages) $C_X(t)(X=*,A,B)$, which we define as fractions of lattice sites occupied by particles X at time t. The concentrations obey the conservation law

$$C_*(t) + C_A(t) + C_B(t) = 1. \tag{5}$$

The formal exact kinetic equations for the global concentrations read as follows:

$$\frac{d}{dt} C_A(t) = \zeta C_*(t) - \gamma C_A(t) - R, \tag{6}$$

$$\frac{d}{dt} C_B(t) = -(1-\zeta) C_B(t) + \gamma C_A(t) + R, \tag{7}$$

where R is the reaction term. As was shown [6], this term cannot be calculated in the framework of the simplest approximations such as the mean field (MF) or pair approximation. More precisely, such simplified calculations lead to results totally different from those given by the MC simulations and cannot account for oscillations. The difficulties in the analytic description of the system lie not in the infinitely large reaction rate [one can start with some finite rate K in Eq. (3) and then take the limit $K \rightarrow \infty$]. The problem is that the reaction term in Eqs. (6) and (7) is not the function of only concentrations like the traditional the MF expression $R = K C_A C_B$ or analogous, but the functional of the *spatial distribution* of particles. In the Lotka model, this distribution undergoes nontrivial changes during the reaction (we will relate them later to a percolation); therefore, the reaction term is unknown.

Despite these difficulties, we are going to show that Eqs. (6) and (7) are still useful for an analysis of the problem if

applied to timaree intervals on which the reaction term R can be omitted for some reason. The resulting solutions can be then tailored together based on additional heuristic assumptions about oscillation mechanism in the Lotka system.

III. METHODS

A. Simulation procedure

An overview of MC simulation techniques for surface reaction systems can be found in Ref. [16]. Here we give only a brief description of the simulation procedure that was used for the Lotka model in our particular study.

We start at time $t=0$ with the lattice randomly filled by particles B at fraction 0.5. Time evolution according to Eqs. (1)–(3) is based on a constant time step $\Delta t=1/L^2$ and consists of the following steps.

(1) Update the clock $t\rightarrow t+\Delta t$.

(2) Pick a site randomly.

(3) If the site is occupied by B, then make it empty with probability $1-\zeta$ and go to (1).

(4) If the site is occupied by A, then proceed to (7) with probability γ.

(5) If the site is vacant, then fill it with A with probability ζ; otherwise, return to (1).

(6) Check for B's in the nearest-neighbor sites. If no B found, then go to (1).

(7) Identify all A's belonging to the same cluster with the A in the selected site. Turn them all into B's. Go to (1).

A special feature of the Lotka model is the infinite rate of the autocatalytic step. Since the pioneering work of Ziff *et al.* [17] infinitely fast reactions have been successfully treated by the means of MC simulations. In our algorithm *no clock update* is done until all $A\rightarrow B$ conversions dictated by the reaction rule have been processed in step (7).

The system was simulated on two-dimensional square, triangle, and honeycomb lattices with the total number of sites in each lattice $(1-16)\times 10^6$. Periodic boundary conditions have been applied in each case.

Each simulation lasted for $(4-10)\times 10^3$ time units totally. The data for analysis were collected only after the system had relaxed through a transient period of 10^3 time units.

B. Data analysis

In the present study simulation data are mainly analyzed by calculating the power spectrum of global concentrations $C_X(t)$. Therefore the question of correct power spectrum estimation is of special significance.

Direct application of a discrete Fourier transform to $C_X(t)$ sampled at n points $t_k=k\Delta T(k=0,1,\ldots,n-1)$, with $\Delta T=T/n$, gives a periodogram power spectrum estimator (PSE) [18]:

$$\mathcal{P}_X(f)=\frac{8}{n^2}\left|\sum_{k=0}^{n-1} w_k C_X(t_k)e^{-2\pi i f t_k}\right|^2. \tag{8}$$

We use the Hann window function $w_k=(1/2)[1-\cos(2\pi k/n)]$ to minimize distortions in the estimate of a spectrum with sharp features (such as separate harmonics of a periodic signal). The frequency resolution of Eq. (8) is determined by the half-width of the window function in the frequency domain and varies as $\Delta f\propto T^{-1}$ with a proportionality factor of the order of unity.

The normalization of $\mathcal{P}_X(f)$ satisfies the equation

$$\frac{1}{f_N}\int_0^{f_N}\mathcal{P}_X(f)df\approx\frac{1}{T}\int_0^T C_X^2(t)dt, \tag{9}$$

where $f_N=1/(2\Delta T)$ is the Nyquist aliasing frequency.

Spectral analysis of stochastic signals such as MC simulation data is more complicated than in the case of a deterministic model. Statistical variance of $\mathcal{P}_X(f)$ grows as the ratio of the transform window length T to the coherence time of the signal increases [18].

Therefore a more robust amplitude estimate can be achieved by dividing the whole time interval of available data into pieces of possibly smaller length and then performing a separate Fourier transform on each individual subinterval. Results are then averaged, thus giving a statistically more stable power estimate of the cost of the reduced frequency resolution.

Averaging over m successive subintervals each of length T leads to an averaged periodogram PSE

$$P(f)=\frac{8}{mn^2}\sum_{l=0}^{m-1}\left|\sum_{k=0}^{n-1} w_k C_X(t_k+lT)e^{-2\pi i f t_k}\right|^2, \tag{10}$$

which is more suitable for the study of noisy signals than $\mathcal{P}_X(f)$.

The following iterative procedure was used for estimation of the main oscillation frequency f_0 and corresponding power $P(f_0)$.

(1) Sample N values of $C_X(t)$ with $\Delta T=2^{-6}$.

(2) Set the initial approximation of f_0 to $f_0'=\zeta$.

(3) Calculate $n\rightarrow$ the nearest integer to $10/(f_0'\Delta T)$, $m\rightarrow$ the integer part of N/n.

(4) Find the maximum of $P(f)$ in interval $(0-f_N)$. Take its position as the next approximation f_0'.

(5) Repeat steps (3) and (4) until n does not change.

(6) Assume the main frequency f_0 to be equal to the last value of f_0'. The first harmonic power estimate is the value of $P(f_0)$ found in step (4).

The above-described procedure is designed to match the frequency resolution $\Delta f\approx 0.1 f_0$ and gives a stable estimate of the spectral power under the first peak in a discrete spectrum if its width is less than Δf. The values of ΔT and f_0' in steps (1) and (2) are based on a previous analysis of the Lotka model data.

IV. ANALYTICAL RESULTS

The autocatalytic transformation $A+B\rightarrow 2B$ spreads between the nearest neighbors. This feature of the Lotka model relates it to standard problems of percolation theory which we overview very briefly below.

In a standard *site percolation* problem sites of an infinite lattice are marked with some probability p independently

each of the other. The distribution of clusters consisting of these marked sites is of a relevant interest. Percolation theory states [19] that an *infinite* cluster of marked sites can be formed with nonzero probability if and only if the marking probability p is greater than some threshold value p_c, which is called the *percolation threshold*. The value of p_c depends on the dimension and topology of the lattice. The numerical values of p_c for different lattices are cited in many papers (see, for example, Ref. [20]).

Criticality of the standard percolation problem makes all the characteristic linear sizes proportional to a single length scale ξ_p which diverges near the percolation threshold as $\xi_p \propto |p_c - p|^{-\nu}$. The critical exponent ν depends on the spatial dimension of the lattice. Properly defined the average linear cluster size is proportional to ξ_p and also diverges at the percolation threshold.

When performing simulations of a percolation system, only finite lattices may be used. In this case percolation properties depend on the relation between the system's intrinsic length scale ξ_p and the lattice size L. If $\xi_p \ll L$, then the percolation theory for infinite lattices applies well. On the other hand, in the close vicinity of the critical point p_c finite-size effects can be no longer neglected and one has $\xi_p \approx L$. In the latter case, instead of the infinite cluster, one observes a cluster of a linear size L spanning the lattice from top to bottom.

Turning back to the Lotka model, let us consider for a moment only a random deposition of particles A onto an initially empty lattice [pure adsorbtion according to Eq. (1)]. Then the distribution of particles A at some time t is exactly the same as of the marked sites in the percolation problem described above. The average concentration $C_A(t)$ plays the role of the marking probability p. The average cluster size grows as the concentration of particles increases and a spanning cluster of A's appears when $C_A(t) \approx p_c$.

Now we recall that there are also particles B in the system. Thus, we can formulate the following simplified kinetic scheme of the oscillations in the Lotka model applicable for the case of *small* ζ values.

Assume that the largest A cluster is spanning the lattice at the moment when it touches one of the few remaining B particles. At this moment, the autocatalytic conversion becomes dominant and turns most of the A's into B's. The initial moment ($t=0$) immediately after this global turnover can be characterized by conditions $C_A(0)=0$ and $C_B(0)=p_c$. We neglect all spatial correlations between particles afterwards and assume the distribution of both A and B particles to be random and independent. The evolution of the reaction system at times $t>0$ is dominated by two processes occurring at different time scales: (i) the fast decay of particles B [lifetime $1/(1-\zeta) \approx 1$] and (ii) the slow accumulation of new A's (rate $\zeta \ll 1$). We neglect the loss of particles A due to reaction, Eq. (3) [and, if $\gamma \neq 0$, Eq. (4)], until $C_A(t)$ approaches closely to the percolation threshold, and assume that the concentration of B decreases quickly to a very low background value before large A clusters form.

It should be mentioned that the background value of $C_B(t)$ is undoubtedly very sensitive to any B producing reactions even with small rates. It might influence the length scale of synchronization, but appears not to affect the oscillation period that we are going to estimate in a moment.

Based on the assumptions made above we use the kinetic equations (6) and (7) omitting the reaction term R:

$$\frac{d}{dt} C_A(t) = \zeta C_*(t) - \gamma C_A(t), \tag{11}$$

$$\frac{d}{dt} C_B(t) = -(1-\zeta) C_B(t) + \gamma C_A(t). \tag{12}$$

When the concentration $C_A(t)$ reaches the percolation threshold again, a short lasting chain of reactions (3) converts almost all A's into B's and the cycle returns back to the initial state (equivalent to $t=0$). The oscillation period τ is determined from the equation

$$C_A(\tau) = p_c. \tag{13}$$

The simplified oscillation scheme given above contains some significant assumptions, which are not necessarily true in a real simulation. Let us mention some of them: (i) growing A clusters may switch to B before or after the percolation threshold is reached, (ii) the distribution of particles is not exactly random, and (iii) the turnover to the B-dominant state is neither instant nor complete. It is essential, however, that all these details weakly affect the main oscillation characteristic—the period. On the other hand, the detailed picture must be considered when discussing the qualitative aspects of oscillations—such as synchronization between different parts of the lattice.

The exact solution of MF equations (11) and (12) is straightforward but lengthy. Therefore, keeping in mind that we are interested only in the limit of $\gamma \ll \zeta \ll 1$, we omit the terms containing γ and find the solution up to the first order in ζ:

$$C_A(t) = 1 - e^{-\zeta t}, \quad 0 \leqslant t < \tau, \tag{14}$$

$$C_B(t) = p_c e^{-t}, \quad 0 \leqslant t < \tau, \tag{15}$$

$$f_0 \equiv 1/\tau = -\zeta / \ln(1-p_c). \tag{16}$$

We also estimate the minimal value of $\zeta = \zeta_{min}$ at which oscillations on a *finite* lattice are possible (for $\gamma = 0$). As was already discussed in Sec. II, lattice poisoning occurs if all B particles desorb. By setting the minimal concentration of B particles $C_B(\tau)$ equal to one particle per lattice, we find, for the lattice of linear size L and dimension d,

$$\zeta_{min}(L) = -\frac{\ln(1-p_c)}{\ln(L^d p_c)}. \tag{17}$$

The logarithmic dependence of ζ_{min} on L explains why it is very hard to come close to the limit $\zeta \to 0$ in direct simulations. We expect the estimate (17) to be less precise than that of frequency (16) because it depends on the assumption-sensitive background of B particles.

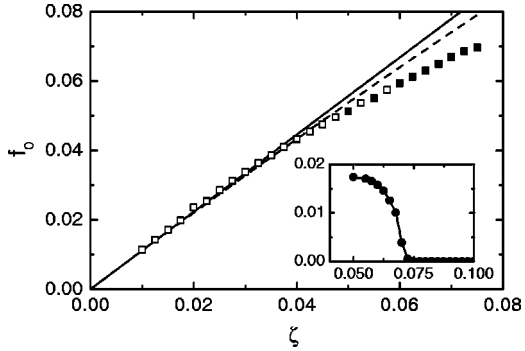

FIG. 1. Oscillation frequency vs adsorbtion rate ζ (both in dimensionless units). Simulation data on a 4096×4096 square lattice with $\gamma=0$ (■) and $\gamma=5\times10^{-6}$ (□) are compared to the frequency estimate, Eq. (16) (the straight solid line). More precise values of the frequency calculated from the exact solution of Eqs. (11) and (12) are indicated by the dashed curve. The inset shows the amplitude of the first harmonic $P_A(f_0)$ in units set by the normalization, Eq. (9), as a function of ζ.

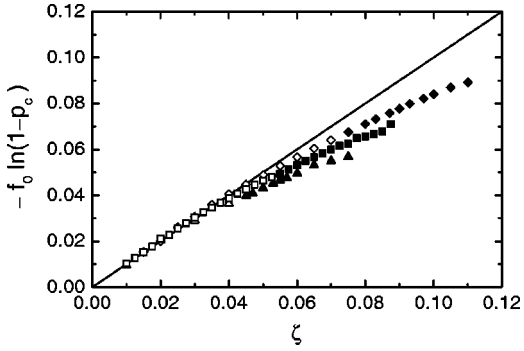

FIG. 2. Oscillation frequencies for lattices of different topology vs parameter ζ (dimensionless units). Simulation data for square (■,□), triangle (▲,△), and honeycomb (◆,◇) lattices lie on a single line if multiplied by $-\ln(1-p_c)$ according to Eq. (16). Solid and open symbols stand for $\gamma=0$ and $\gamma=5\times10^{-6}$ during each simulation, correspondingly.

V. SIMULATION RESULTS

A. Oscillation frequency

First we focus now on the frequency of *synchronized* oscillations which our simplified scheme applies to. As we are interested in the case of small ζ, the problem of lattice poisoning becomes significant. The minimal value of ζ at which oscillations were still observable in simulations on a square 4096×4096 lattice with no modifications of the initial model ($\gamma=0$) was $\zeta_{min}=0.05$. It is in reasonable agreement with the estimate (17) which gives for this case $\zeta_{min}=0.056$. The additional reaction (4) with $\gamma=5\times10^{-6}$ was used for $\zeta<\zeta_{min}$. Results are plotted in Fig. 1. Data points for both cases overlap nicely. This indicates the usefulness of the additional $A\rightarrow B$ reaction in the study of oscillations.

Figure 1 demonstrates the accuracy of the frequency estimate (16) for $\zeta\rightarrow0$. More precise calculations based on the exact solution of Eqs. (11) and (12) reveal a slight deviation from a straight line (dashed curve in Fig. 1). Simulation data for large ζ deviate even stronger due to ignored effects of reaction (3) in the simplified oscillation scheme. Additional B production leads to slower accumulation of A's and, as a result, to smaller frequencies than predicted by the simplified oscillation scheme.

Increasing of ζ above ζ_{min} leads to a rapid decrease of the oscillation amplitude at $\zeta=\zeta_0\approx0.075$ as shown in the inset in Fig. 1. We will return to this desynchronization phenomenon in Sec. V B.

Use of the percolation parameters for different lattices in the frequency estimate (16) allows us to compare lattices with different topologies. Data for square, triangle, and honeycomb lattices ($p_c=0.592\,746$, 1/2, and 0.6962, correspondingly) are given in Fig. 2. We see strong evidence that the oscillation frequency in the Lotka model (in the limit $\zeta\rightarrow0$) is determined only by the kinetic parameter ζ and lattice percolation threshold p_c.

B. Synchronization

In this section we look in more detail at the origin of globally synchronized oscillations. Significant information can be extracted from the power spectra of simulation data. Before we proceed to a further analysis of the model, we first test our spectrum estimation algorithm (described in Sec. III B) on a purely periodic signal.

As the test data we choose the particle A concentration given by the analytical solution, Eq. (14). The power spectrum is expected here to be a set of discrete harmonics located at integer multiples of the main frequency f_0. The amplitudes of the peaks must decrease like $\propto f^{-2}$ as follows from the Fourier transform of Eq. (14). By inspecting the double-logarithmic plot in Fig. 3, one can see that the averaged periodogram PSE (10) gives a correct picture of the spectrum. The minimal value of $P_A(f)$ between the neighboring harmonics ($\approx3\times10^{-9}$) is limited by the frequency resolution of the PSE and by the effects of signal power aliasing at frequencies above f_N into the interval $(0-f_N)$. The aliasing is also responsible for deviations from the straight line at high frequencies observed in Fig. 3.

Let us now examine the power spectra of $C_A(t)$ obtained from simulations of the Lotka model at different values of ζ. Figure 4(a) represents the ultimately synchronized state at $\zeta=0.05$. Totally more than 50 harmonics can be observed in the discrete part of the spectrum with frequencies up to f_c

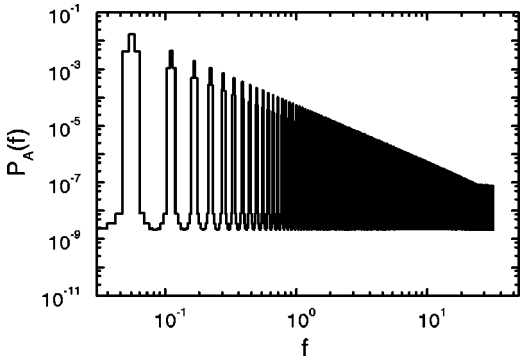

FIG. 3. Power spectrum of the analytical solution, Eq. (14), with $\zeta=0.05$ for a square lattice. The spectrum is estimated by the averaged periodogram PSE, Eq. (10), with maximal frequency $f_N=32$. Absolute values of $P_A(f)$ conform to the normalization, Eq. (9), whereas the frequency is dimensionless.

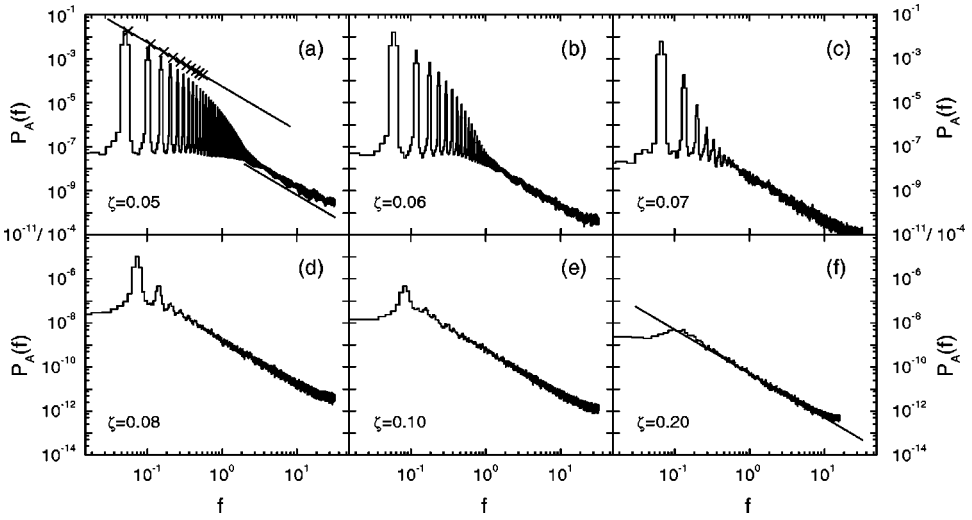

FIG. 4. Power spectra $P_A(f)$ of A particle concentrations at $\zeta = 0.05$, 0.06, 0.07, 0.08, 0.1, and 0.2 [from (a) to (f)]. Simulation data for a 4096×4096 square lattice, aliasing frequency $f_N=32$; units are the same as in Fig. 3. Crosses in plot (a) denote the spectrum of the simplified analytical solution, Eq. (14). All straight lines on the plots have the same slope of -2.

≈ 3 (surprisingly many for a stochastic simulation). Amplitudes of the first few harmonics correspond well to the simplified oscillation scheme [crosses in Fig. 4(a) correspond to the peak values in Fig. 3]. At higher frequencies (but still below f_c) the discrete part of the spectrum goes down faster than f^{-2}. This fact can be easily understood—the major contribution at high frequencies in the analytical solution is due to the instant switch of all A's to B's between the successive cycles. In simulations this turnover is more smooth, thus leading to smaller power in the spectrum at high frequencies.

At frequencies above f_c a qualitative change in the spectrum occurs: the equidistant discrete harmonics transform into noise with continuous spectrum obeying the same power law $\propto f^{-2}$.

Figures 4(b)–4(e) give a clear picture of desynchronization. As the parameter ζ increases, higher harmonics become less distinct and transform into noise at lower frequencies f_c. The amplitude of the first peak decreases dramatically (see the inset in Fig. 1) while its width grows. At $\zeta=0.2$ [Fig. 4(f)] the whole spectrum is dominated by f^{-2} noise with traces of the first harmonic only.

Additional information on the transition from the synchronized to the noise-dominated state can be derived from the analysis of the spatial correlation function which we define here as

$$K_{XY}(\boldsymbol{r};t)=\frac{\langle \sigma_X(\boldsymbol{r}';t)\sigma_Y(\boldsymbol{r}'+\boldsymbol{r};t)\rangle_{\boldsymbol{r}'}}{C_X(t)C_Y(t)}-1, \qquad (18)$$

where $\sigma_X(\boldsymbol{r})$ is equal to 1 if a site with radius vector $\boldsymbol{r}$ is occupied by a particle X and 0 otherwise; $\langle\ \rangle_{\boldsymbol{r}'}$ denotes averaging over the lattice. In the simplified oscillation scheme we neglected all correlations in the spatial distribution of particles by considering, in particular, that all A particles are distributed randomly. This assumption corresponds to $K_{AA}(\boldsymbol{r};t)\equiv 0$. In a real situation, however, the reaction always creates some correlations in the ensemble of particles. Therefore calculation of $K_{AA}(\boldsymbol{r};t)$ from the simulation data can serve for a control of the validity of the simplified oscillation scheme.

We present in Fig. 5 three plots demonstrating the dynamics of the autocorrelation function $K_{AA}(r;t)$ averaged over all spatial directions. In the first graph [Fig. 5(a)], which illustrates the synchronized state, the temporal periodicity of the A-A correlation function is well pronounced. The rapid increase in time of $K_{AA}(\boldsymbol{r};t)$ at small distances corresponds to the beginning of global switching from A to B. A tail of long-range correlations, which can be clearly seen in Fig. 5(a), is a result of a short-lasting chain of large avalanches that spread through the lattice converting most of A's into B's. The following decay of A-A correlations is due to the random accumulation of the new A particles in the place of fast desorbing B. We emphasize that at the end of a cycle, when correlations are minimal, the spatial distribution of A particles is close to that of a random deposition which standard percolation theory applies to. This is a necessary condition for the simplified oscillation scheme to be consistent with simulations.

Last, Fig. 5(c) presents the A-A correlation function in the desynchronized state which possesses almost no time dependence. $K_{AA}(r;t)$ decays on a constant distance which is

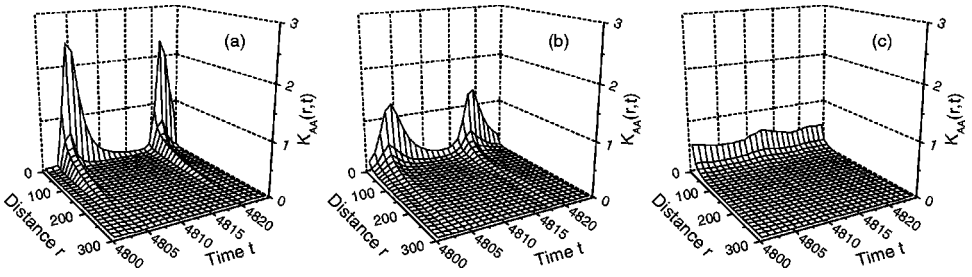

FIG. 5. Time-resolved dimensionless A-A spatial correlation function $K_{AA}(r,t)$ on a square 1024×1024 lattice for (a) $\zeta = 0.06$, (b) 0.07, and (c) 0.08. Distance is given in lattice constants and time in dimensionless units.

much less than the system size. No attempts were made to estimate this distance or its dependence on ζ.

VI. DISCUSSION

The distribution of avalanche sizes $P(s,\zeta)$, where s is the number of B's born in an avalanche, was intensively studied by Hovi *et al.* [9]. Based on a large amount of simulation data, they found that for small ζ the distribution $P(s,\zeta)$ decays as a power law in s up to a certain cutoff size s_{max}. Significant evidence was found that s_{max} diverges at $\zeta=0$ in the thermodynamic limit. Therefore the authors claim that the point $\zeta_c=0$ is critical in the Lotka model.

Depending on how close the system is to its critical point, the cutoff in avalanche size distribution occurs due to either the system's intrinsic dynamics or finite-size effects. In the former case (far from ζ_c), s_{max} depends on ζ but not on L while the opposite in the latter (near to ζ_c). This behavior of s_{max} was observed but not studied in detail [9]. We continue this line of reasoning and propose the following picture of synchronization in the Lotka model.

Let us denote as ζ_0 the value of ζ at which the cutoff distance (linear size of a cluster containing s_{max} particles A) reaches the size of the system.

At sufficiently large $\zeta(>\zeta_0)$ the cutoff size s_{max} is much less than the total number of sites in the system and global avalanches almost never occur. Single avalanche negligibly affects the global concentration of particles and we observe the steady state. In this regime different parts of the lattice are uncorrelated as one can directly see in simulations [Fig. 5(c)]. The power law size distribution of the *independent* avalanches results in a power law spectrum (''$1/f$ noise''). This mechanism of generating $1/f$ noise was discussed in great detail in a classical paper by Bak, Tang, and Wiesenfeld [21], who introduced the concept of self-organized criticality in order to explain the wide abundance of this phenomenon. The forest-fire (FF) model of self-organized criticality by Drossel and Schwabl [22], which we will discuss in a moment, was shown to reveal $1/f$ noise in the fire density power spectrum with the same exponent of -2 in two dimensions as we observe for the Lotka model.

Decreasing of ζ moves the system towards its critical point. For $\zeta<\zeta_0$ finite-size effects start to play a major role. Large avalanches become correlated in time: a single large event influences the whole system which now needs some relaxation time for making another large avalanche possible. Indeed, we saw in the power spectra (Fig. 4) that large size avalanches (which occur at low frequencies) are synchronized while small ones are still uncorrelated, producing power law noise at high frequencies.

Divergence of the cutoff size s_{max} in the limit $\zeta\to\zeta_c$ suggests that global oscillations are possible on *arbitrary large* lattices.

In addition, we return to the plot of the first harmonic $P_A(f_0)$ vs ζ (see the inset in Fig. 1). One can see that globally synchronized oscillations emerge quite sharply after ζ is decreased below some threshold value which provides therefore a good estimate of ζ_0.

Hovi *et al.* have already noted [9] the similarity between the model under consideration and the FF model. If A is understood as a tree and B as a fire, then the FF model can be described by the same equations (1)–(4) as the Lotka model, but with other limiting conditions on the rates. If we denote the rates of the reactions $*\to A$, $B\to *$, $A+B\to 2B$, and $A\to B$ by k_1, k_2, k_3, and k_4 correspondingly, then the FF model is critical provided that

$$k_4(\equiv f)\ll k_1(\equiv p)\ll k_2=k_3(=1) \tag{19}$$

(we give the notation of Ref. [22] in parentheses). In the Lotka model the criticality is observed if

$$k_1(\equiv\zeta)\ll k_2(\approx 1)\ll k_3(=\infty). \tag{20}$$

In both the FF and Lotka models criticality occurs due to a double separation of time scales. As was pointed out [22], such a condition requires no fine-tuning and therefore may be quite general.

VII. CONCLUSIONS

We have studied here the oscillatory irreversible reaction $A+B\to 2B$ by means of analytical methods and MC simulations on different types of two-dimensional lattices. The mean-field approximation fails even in a qualitative description of the system, but the equations of standard chemical kinetics can be still useful for an analytical estimate provided there exist additional heuristic arguments based on the percolation mechanism of auto-oscillations in this reaction.

Oscillations in the Lotka model arise from the separation of time scales: all but a few B particles desorb quickly, providing free space for the slow growing percolating network of A particles. The resulting oscillation frequency is then proportional to the A adsorbtion rate and related to the topology of the lattice through the percolation threshold. Simulations on different lattices confirm this mechanism.

A globally synchronized state is observed for small values of the control parameter ζ. Increasing of ζ brings closer the time scales of the adsorbtion and desorbtion reactions, thus leading to desynchronization. As a result, qualitative changes in the power spectrum of oscillations occur: a set of discrete harmonics transforms into f^{-2} noise previously reported for model systems of self-organized criticality. Based on these observations, we propose a qualitative picture of global synchronization in the Lotka model as a finite-size effect near the systems's critical point $\zeta_c=0$.

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