Internal spatiotemporal stochastic resonance in the presence of weak noise

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We show the existence of internal stochastic resonance in a microscopic stochastic model for the oscillating $A + \frac{1}{2}B_2$ reaction on a square lattice. This stochastic resonance arises directly from the elementary reaction steps of the system without any external input. The lattice gas model is investigated by means of Monte Carlo simulations. It shows oscillation phenomena and mesoscopic pattern formation. Stochastic resonance arises when homogeneous nucleation of the individual lattice site states is considered. This nucleation is modeled as a weak noise process. As a result, synchronization of the kinetic oscillations is obtained. We show that all characteristics known from the research on stochastic resonance are obtained in our model. We also show that the model explains easily several phenomena observed in the experiment. Internal stochastic resonance may thus be an internal regulation mechanism of extreme adaptability.

The term stochastic resonance (SR) is given to the somewhat counterintuitive phenomenon that in a nonlinear system a weak signal can be amplified by the assistance of noise. It has been introduced in 1981 by Benzi *et al.* [1,2] in the context of a study about the periodically recurrent ice ages. Over the last two decades it has continuously attracted increasing attention and was shown to occur in many systems [3–5] in biology [6–8], chemistry [9–11], and physics [12–14]. Generally, systems showing SR are described in a formal mathematical way using phenomenological macroscopic equations of the mean field type including (i) a bistable system with an activation barrier or some sort of threshold, (ii) a weak coherent input, and (iii) a strong external noise that helps to overcome the activation barrier. These macroscopic equations in a sense are able to describe many different systems (because stochastic resonance is a general phenomenon that occurs in many natural systems) and have been used in the description of stochastic resonance phenomena in Nd:YAG (Yttrium aluminum garnet) lasers [15], homogeneous [16] as well as heterogeneous [17,18] chemical reactions, bistable quantum systems [19], or the Lotka-Volterra model [20]. More complex systems (e.g., a summing network of excitable units [21], sheep populations [22], two-dimensional excitable media showing spatiotemporal pattern formation [23], sensory systems in crayfish [6] or in the visual cortex [24], or neuronlike systems [25]) are generally modeled via Langevin equations or the Fitzhugh-Nagumo model.

In addition, a few special systems have been investigated in a more general manner via a macroscopic mathematical description, e.g., an autonomous oscillating system [26], a system in the limit of weak noise [27], a system showing stochastic multiresonance [28], and nondynamical systems with both internal and external noise [29]. Note that in the latter case the internal noise is modeled in the same way as an external noise and it is only regarded as a general internal noise without specifying the physical background. Computer simulations performed to date consider coupled neurons or general threshold devices, which are mesoscopic models, i.e., the microscopic physical picture is again neglected.
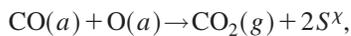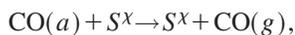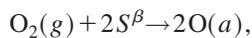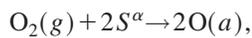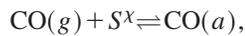
Our present model system is very unusual in the research on SR and maybe adds to the field a new feature. It gives a microscopic description of the phenomenon of stochastic resonance without any external input (neither oscillatory signals nor noise input). All processes are internal to the system itself. This is the reason we will call internal stochastic resonance as a short hand description. Our model also demonstrates the physical reasons for stochastic resonance on the microscopic (atomic) length scale. It is based on stochastic transitions, each with a clear physical meaning. Without noise, the system exhibits a spatially extended heterogeneous stable state with inherent local oscillations. More important, the noise is not an external input but corresponds to a physically realistic internal nucleation process. Because of the clear physical picture on the microscopic (atomic) level our model is of course specialized and cannot describe a large variety of different systems. But on the other hand, the results of this model and the conclusions that can be drawn are very general ones and they suggest that internal SR may be the reason for inherent synchronization and cooperative phenomena in many physical, chemical, and biological systems.

We consider a slightly modified version of a previously presented model for the catalytic $CO + 1/2\,O_2$ reaction on Pt single crystal surfaces [30–33], which shows different types of kinetic oscillations in agreement with experimental results. The model involves A (CO) adsorption, desorption and diffusion, dissociative B_2 (O_2) adsorption, and two surface phases (reconstructed and nonreconstructed), which form and propagate governed by the coverage with A. The details are given below. An extended version of the model for the $CO + NO$ reaction on Pt(100) is able to describe the experimentally observed transition into chaotical behavior via the Feigenbaum route [34]. Our model follows the well-known model by Ziff, Gulari, and Barshad [35] (ZGB model) and is investigated by means of Monte Carlo (MC) simulations.

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The Pt(110) surface of the catalyst is represented by a square lattice of side length L and lattice constant $a=1$. From experiment [36], it is well known that kinetic oscillations are closely connected with the $\alpha \rightleftharpoons \beta$ reconstruction of the Pt(110) surface, where α and β denote the 1×2 and the 1×1 surface phase, respectively. Each site can belong to the α or the β phase, respectively. Therefore only the different physical but not geometrical properties of the phases are considered because it is almost impossible to give a *local* geometric specification of the phase. This is justified because simulations on the square and triangular ($z=6$) lattice show only quantitative but not qualitative differences. In our model, A is able to adsorb onto a free-surface site with rate $y_A = y$ and to desorb from the surface with rate k, independent of the surface phase the site belongs to. B_2 adsorbs dissociatively onto two nearest-neighbor (NN) sites with different sticking coefficients s onto the two phases ($s_\alpha = 0.5$, $s_\beta = 1$). Therefore we get the rate constants $y_B^\alpha = 1 - y$ and $y_B^\beta = 2(1-y)$ for B adsorption on the α and β phase, respectively. For B_2 adsorption directly at the phase border where one site belongs to the α and the other one to the β phase the geometric mean of these adsorption rates is used. In addition, A is able to diffuse with rate D via hopping onto a vacant NN site. The $A+B$ reaction occurs, if A hops to a site that is covered by B and the reaction product AB desorbs immediately from the surface. All these processes are associated with the above kinetic transition rates of the stochastic model which therefore determine the relative speed of the individual reaction processes.
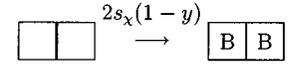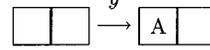
Summarizing the above transition definitions, we simulate the following model written in the more usual form of reaction equations. For the $CO + O_2$ reaction, the basic equations are

$$CO(g) + S^\chi \rightleftharpoons CO(a),$$

$$O_2(g) + 2S^\alpha \rightarrow 2O(a),$$

$$O_2(g) + 2S^\beta \rightarrow 2O(a),$$

$$CO(a) + S^\chi \rightarrow S^\chi + CO(g),$$
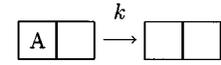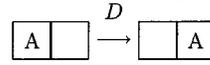
$$CO(a) + O(a) \rightarrow CO_2(g) + 2S^\chi,$$

where S stands for a free adsorption site, χ stands for either α or β, and (a) or (g) for a particle adsorbed on the surface or in the gas phase, respectively. Note that in the model simulated here O_2 adsorbs onto sites belonging to the α or β phase with different sticking coefficients.

The kinetic MC computer simulations are based on the pair algorithm that is explained in detail in Refs. [37,38]. Because we only consider pairs of NN sites a graphical representation of the MC rules corresponding to the reaction equations above can be shown.
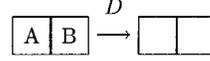

The $\alpha \rightleftharpoons \beta$ phase transition is modeled as a linear phase border propagation. Consider two NN surface sites in the state $\alpha\beta$. The transition $\alpha\beta \rightarrow \alpha\alpha$ ($\alpha\beta \rightarrow \beta\beta$) occurs if none (at least one) of these two sites is occupied by A. This phase border propagation mechanism mimics the growth of the β phase because of the larger adsorption energy of A on the 1×1 phase than on the 1×2 phase [36]. The individual phases are stable or metastable. The direct transition from a globally homogeneous α phase into a homogeneous β phase (or vice versa) is impossible; the activation barrier is infinite. The stability of the individual phases depends on the chemical coverage Θ_i of species i on the surface of the catalyst. For $\Theta_A < 0.3$ the α phase is stable and for larger values the β phase is stable [31]. The coverages of A and B vary in the course of the reaction because of the different sticking coefficients of B_2 on the two surface phases. If we start with an almost homogeneous α phase the adsorption of A is preferred. Therefore the A coverage increases whereas almost all adsorbed B atoms are removed from the surface via $A + B$ reaction. If the A coverage is large enough small β phase nuclei start to grow up to homogeneous β phase islands of mesoscopic size. On the β phase the adsorption of B is preferred and the A atoms are actively removed from the surface. The large B coverage on the other hand now initiates the growth of the α phase and the whole cycle starts again. Starting with a heterogeneous distribution of the α and β phase the activation barrier for the surface phase transition is finite, but the transition into a globally homogeneous phase does not occur because of the finite surface phase propagation velocity, i.e., the α or β phases cannot grow to macroscopic islands. Therefore the oscillations remain local, interfere, and cancel each other on sufficiently large surfaces [30–32]. The system exists in a heterogeneous, dynamically stable state with oscillations, which are locally synchronized by A diffusion but disappear on the macroscopic length scale for large lattices. The α and β phase, however, build almost homogeneous islands on a mesoscopic length scale. The power spectrum for the case of the Pt(110) surface clearly shows the existence of one basic frequency ω_0, which depends on the individual parameters $\omega_0 = \omega_0(k, V, D, y)$. At constant y the basic frequency increases with increasing k ($k > k_c = 0.07$ [30]) and V, and decreases with D as $\omega_0 \sim k\sqrt{V/D}$. This is proven for $D \in (10, 10^3)$ and $V \in (10^{-1}, 10)$ [32].

The nucleation is modeled as a spontaneous $\alpha \rightarrow \beta$ or $\beta \rightarrow \alpha$ transition of a single site, completely independent of its neighbors or the particle adsorbed onto this site. Therefore this nucleation is a homogeneous process that corresponds to

a weak noise that generates dynamic defects on the surface. The term weak noise is used because the nucleation rate $\gamma \in [10^{-5}, 10^{-2}]$ is for the relevant values several orders of magnitude smaller than the other transition rates that are of the order of 10^{-1}–10^{3}. The defects grow or vanish via the $\alpha \rightleftharpoons \beta$ reconstruction depending on their chemical neighborhood, i.e., the presence or absence of A. It has to be emphasized that all phenomena such as local oscillations, growth and decline of the heterogeneously distributed surface phase islands, and quasiperiodical and chaotical behavior exist even without the consideration of the nucleation process [30–34]. Please note that in addition to our previous theoretical work there exists a recent experimental study [39] where the authors conclude that "the inclusion of defect creation and annealing as an important kinetic step in the oscillatory cycle can explain the experimental observations [···]." Because of the very small nucleation rate, which introduces only very few defects into the existing heterogeneous surface state, one might suppose that the defects could have no influence on the oscillating system, but we shall demonstrate that the nucleation process may have a profound influence on the system behavior leading to cooperative phenomena and the synchronization of the local oscillations.

In comparison with the standard problem of SR there exist several major differences.

(i) We have a system with two surface phases and additional chemical coverage instead of a bistable system. In addition, none of the two surface phases becomes homogeneous. Both coexist in a dynamically stable heterogeneous state.

(ii) No external coherent input is considered in our model. The local oscillations originate from the kinetic definition of the model itself and are very small on a macroscopic, global length scale. These internal oscillations can be seen as a substitute of the commonly used coherent input. The macroscopic synchronization of the local oscillations then corresponds to a large output signal.

(iii) The source of noise is an internal physical process of the system, the homogeneous nucleation, which breaks the mesoscopic homogeneity of the surface phase islands. The noise process is therefore not an addition to a periodic input but is independent of the oscillations. In most of the previous studies a weak coherent input is coupled with a strong external noise.

(iv) In those cases where thermal internal noise has been considered in previous studies it has been used as an addition to an existing process. As far as we know our model is the first one in which a thermal noise process is a separate process in the system itself. It can be considered as a thermal noise as the phase nucleation requires a temperature to get started. Furthermore, it is clearly defined and explicitly modeled and simulated on the atomic length scale, whereas in previous studies the thermal noise process has been dealt in a formal mathematical way. Please note that many of the above processes have been investigated in previous papers on stochastic resonance, but our model combines all of these aspects.

(v) The model is defined on the atomic length scale and investigated via MC simulations that correspond most
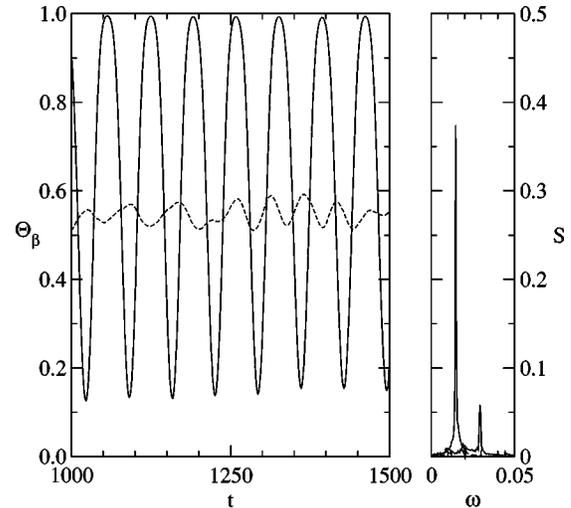

FIG. 1. Oscillations (left) and power spectra (right) of the lattice coverage Θ_β of the β phase in simulations with (solid line, $\gamma = 10^{-3}$) and without (dashed line) nucleation. The other parameters are kept constant at $y = 0.51$ (CO adsorption), $k = 0.1$ (CO desorption), $D = 100$ (CO diffusion), $V = 1$ (surface phase propagation), and $L = 1024$.

closely to a hierarchy of master equations with all correlations included and then mapped onto a finite lattice. The simulation procedure contains additional noise by its very nature but such a noise does not lead to SR.

In the present study, we use $y = 0.51$, $D = 100$, and $k = 0.1$ as standard values if not stated otherwise, because A diffusion is by far the fastest process. Each simulation result is obtained as the average of 20 independent runs of a length of $t = 5000$. The simulations start with an adsorbate-free lattice and randomly distributed surface phases with the coverage $\Theta_\alpha = \Theta_\beta = 0.5$. For details see Refs. [30–34,38]. The value $D = 100$ of the diffusion parameter in our article is a compromise value. It leads to a good statistics of the results. On the other hand, it is possible to use large system sizes (up to $L = 8192$) in the simulations. The existence of the stochastic resonance is independent of the value of D; to each value of D corresponds a particular nucleation rate $\gamma = \gamma(D)$, which gives rise to stochastic resonance.

As can be seen in Fig. 1 the nucleation (noise) has a strong influence on the system behavior. Without nucleation the system shows only very weak oscillations on the global length scale because the existing local oscillations interfere and cancel each other (dashed line). Please note that the lattice side length of $L = 1024$ yet cannot be seen as a macroscopic system in comparison to real physical systems. The oscillations become even weaker and vanish with increasing lattice size. This has been confirmed by simulations with L up to $L = 8192$ and in our previous studies [30,31]. Therefore the oscillations cannot be synchronized only by adsorbate diffusion for larger lattices. If nucleation as a very weak noise process is considered the whole picture changes completely. Now the oscillations are globally synchronized over the whole lattice (solid line). This also holds for lattices up to a lattice side length of $L = 8192$. Because of the long simulation times we did not study the system on larger lattices.
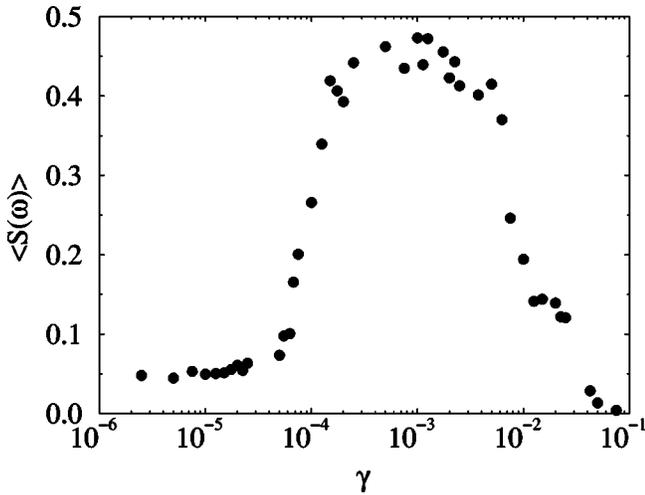
FIG. 2. Global amplitude $S(\omega)$ of the β phase coverage as a function of the nucleation rate γ that gives the noise strength. A stochastic resonance maximum can be seen. The scatter of the points around the maximum is a result of the finite frequency interval due to fast Fourier transform analysis. The values of the amplitude are averaged over 20 simulation results. All other parameters are kept constant at $y=0.51$ (CO adsorption), $k=0.1$ (CO desorption), $D=100$ (CO diffusion), $V=1$ (surface phase propagation), and $L=256$.

In Fig. 2, we show how the amplitude of the synchronized oscillations depends on the nucleation rates. For very small nucleation rates $\gamma < 5 \times 10^{-5}$ only local oscillations exist. The amplitude of the global oscillations vanishes for simulations on large lattices [30,31]. With increasing nucleation rate (strength of the noise) the local oscillations are synchronized on a macroscopic length scale and almost reach the theoretical maximum of $S(\omega) = 0.5$ (Θ_β varies between 0 and 1). This holds also for larger lattices up to $L=8192$, which is the current limit for our simulations. Further increase of the noise decreases this synchronization until at a nucleation rate of $\gamma = 10^{-1}$ the system is completely governed by strong noise. This behavior is the fingerprint of SR [4]. Simulations on lattices with $L=1024$ give almost the same picture but cannot be investigated in such a detail that statistical data can be obtained because of the longer simulation times.

In addition, the noise has an influence on the frequency of the oscillations (see Fig. 3). The normal frequency of the system without nucleation is ω_0, which can only be observed in simulations on small lattices. For nucleation rates $\gamma > 5 \times 10^{-5}$ the phenomenon of SR occurs. The nucleation forces the system to oscillate with a different frequency ω starting at $\omega \approx 2/3\, \omega_0$ for $\gamma = 5 \times 10^{-5}$. With increasing nucleation rate the frequency increases as well up to a value of about $\omega \approx 2\, \omega_0$ at $\gamma = 5 \times 10^{-2}$. This increase in the frequency is based on the increasing number of dynamic phase defects that grow very fast and accelerate the corresponding phase transition. If the number or density of nucleation defects becomes too large the oscillating behavior breaks down and the system is completely governed by noise. Please note that the maximum amplitude coincides with $\omega/\omega_0 = 1$, i.e., with the case where the strength of the nucleation corresponds to the
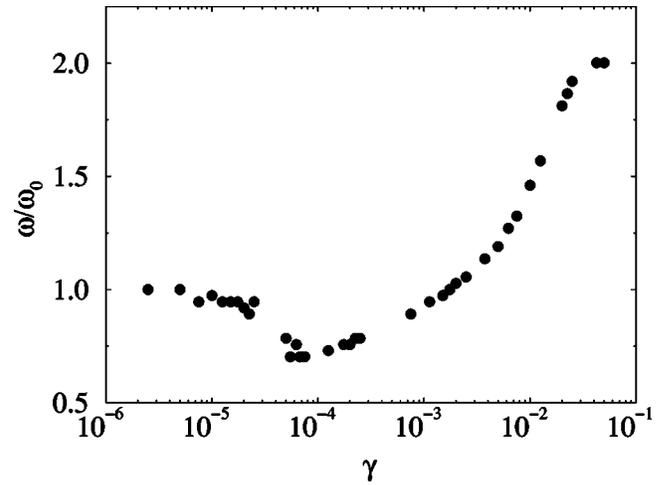

FIG. 3. Ratio of the frequencies ω/ω_0 as a function of the nucleation rate γ that gives the noise strength. Each value is averaged over 20 independent MC simulations. The errors are smaller than the symbol size. All other parameters are kept constant at $y=0.51$ (CO adsorption), $k=0.1$ (CO desorption), $D=100$ (CO diffusion), $V=1$ (surface phase propagation), and $L=256$.

normal frequency of the system without nucleation (see below). The shape of the curve in Fig. 3 in the interval $\gamma \in [10^{-5}, 10^{-1}]$ is in very good agreement with the one obtained in the study of a general autonomously oscillating system by Haken and co-workers [26].

The underlying mechanism of this internal stochastic resonance effect is unexpectedly simple. During the growth and decline of the individual surface phases only a few very small residual phase islands remain in domains where the local synchronization leads to large amplitudes in the phase oscillations. These residual islands are spatially separated at a mean distance R_r (see Fig. 4). Adsorbate diffusion is well known to synchronize individual surface domains within the so-called synchronization length [30,31]
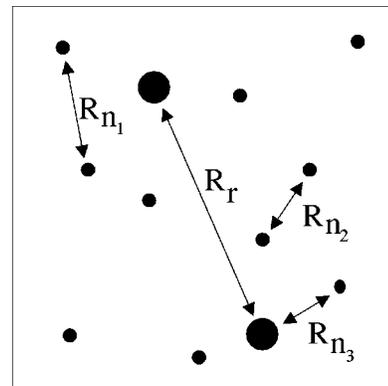
$$\xi \propto \sqrt{DT}, \qquad (1)$$

FIG. 4. This figure shows a simplified picture of the distance R_r between remaining phase islands (large circles) and the distances R_{n_i} between new nucleated phase defects (small circles) or between a new phase defect and an old phase island, respectively. The mean of all R_{n_i} gives the mean distance R_n.
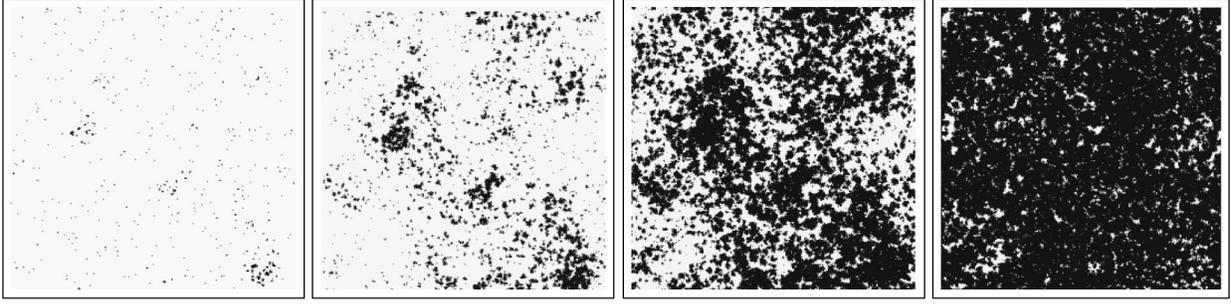
FIG. 5. Snapshots of the system with spontaneous surface phase nucleation ($\gamma = 10^{-3}$). Starting with an almost homogeneous β phase (white), the nucleation of the α phase (black) creates small phase nuclei. These lead to a homogeneous growth of the α phase resulting in an almost complete α phase over the whole lattice. The parameters are $y = 0.51$ (CO adsorption), $k = 0.1$ (CO desorption), $D = 100$ (CO diffusion), $V = 1$ (surface phase propagation), and $L = 1024$.

where D is the diffusion rate and T is the time for one oscillation period. Without nucleation $\xi < R_r$ holds and only locally synchronized oscillations exist, i.e., the process of adsorbate diffusion cannot synchronize the above mentioned remaining phase islands. In addition, the residual islands can only combine into a homogeneous phase if the second condition $R_r \sim VT$ for the phase border propagation is fulfilled. But this condition is also violated because $R_r > VT$ holds. Therefore synchronization can only occur if both conditions are fulfilled. The distance between remaining surface phase islands has to be smaller than the synchronization length of the surface adsorbate diffusion and also smaller than the distance the surface phase borders can propagate over the surface during one oscillation period. On large lattices these conditions are not fulfilled in the system without nucleation.

Nucleation of surface phase defects and subsequent growth of these nuclei now leads to new small phase islands with density $\varrho = \gamma/V \ll 1$ [32], which follows from

$$d\varrho/dt = \gamma - V\varrho \qquad (2)$$

for $t \rightarrow \infty$. The mean distance between these defects is given by $R_n = \varrho^{-1/2} = \sqrt{V/\gamma}$. Because nucleation is a noise process, the individual new phase nuclei and the growing surface phase islands are randomly and homogeneously distributed over the lattice. The mean distance between these nucleated and the old phase islands decreases to R_n, for which $R_n < \xi < R_r$ and $R_n \sim VT$ holds for proper nucleation rates, i.e., in a certain interval of the value of the nucleation rate both condition from above can be fulfilled.

It has been shown in Ref. [31] that the oscillation period is given by $T \sim k^{-1}\sqrt{D/V}$. Because oscillations only occur for values of $k \sim 10^{-1}$ [30], we can use this value of k as a constant. With $R_n = \sqrt{V/\gamma}$ and $VT \sim R_n$, we get

$$\gamma \sim k^2 D^{-1}, \qquad (3)$$

i.e., in a system with physically reasonable diffusion rates a weak nucleation is sufficient to create defects at distances $R_n < \xi$. In our model we use $D = 100$ that leads to a value of $\gamma \sim 10^{-4}$. The present system (Fig. 2) has shown a SR maximum at $\gamma \in [10^{-4}, 10^{-2}]$. In real reaction systems a much

weaker nucleation should be sufficient because the diffusion is underestimated in our present study due to computer limitations.

But there is some additional aspect to the second condition $R_n \sim VT$. Because one oscillation cycle ends when almost the whole surface is in the α or β state, the nucleation rate should have an influence on the frequency of the oscillations via the relationship $R_n \propto T$ with constant V. This can also be seen in Fig. 3. Please remember that the normal frequency of the system without nucleation can only be observed on small lattices and vanishes on macroscopic lattices. Starting from small values the nucleation rate γ starts to synchronize the oscillations at about $\gamma \approx 5 \times 10^{-5}$. For this small nucleation rate the mean distance R_n between the generated α (β) phase defects inside an otherwise homogeneous β (α) phase is relatively large. Therefore the phase propagation, i.e., the growth of these defects needs a longer time T to connect the individual phase islands to an almost homogeneous α (β) phase. Because this time T needed for the connection of individual islands determines the period and therefore the frequency of the oscillations, the nucleation rate determining the mean distance between the nucleated phase defects is able to force the system into oscillations with a frequency different from its normal frequency. Small nucleation rates force the system into oscillations with lower frequencies compared to its normal frequency, whereas larger nucleation rates result in higher frequencies. The latter case is based on the relatively small mean distance between the phase defects that now can be connected in a short time. In the case where the forced frequency due to nucleation is almost equal to the normal frequency of the system the amplitude of the oscillations has its maximum because both synchronizing processes, nucleation and diffusion, lead to the same frequency.

Therefore the separated phase islands can now be connected via island growth and synchronized via A diffusion. This results in macroscopic synchronized oscillations completely independent of the lattice size because only the distance between the phase islands but not the lattice side length is responsible for the appearance of synchronized oscillations. This can clearly be seen in Figs. 5 and 6. In Fig. 5 snapshots of the surface phase distribution are shown for the system with nucleation. Starting with an almost homoge-
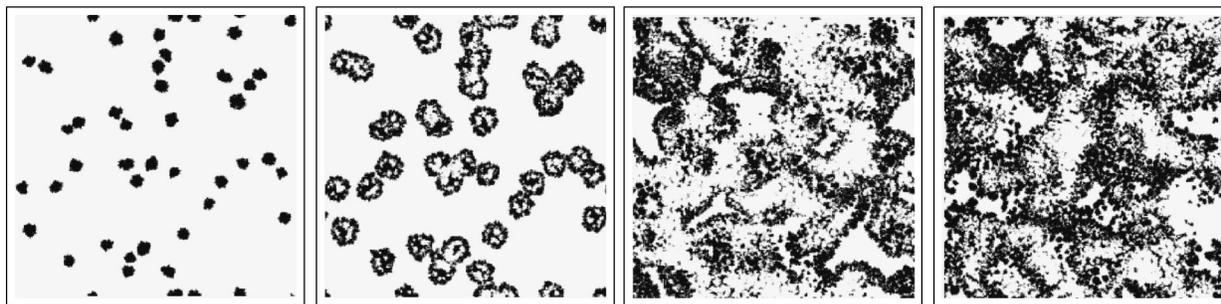
FIG. 6. Snapshots of the system without spontaneous surface phase nucleation. In this system mesosocopic phase domains of both the α (black) and the β phase (white) coexist in a dynamically stable heterogeneous state. The size of the traveling phase domains is determined by the value of the diffusion parameter of adsorbate diffusion which gives the size of the synchronization length. For simulation parameters see Fig. 5.

neous β phase (compare to the time-dependent coverages in Fig. 1) the remaining α phase islands (black) start to grow. In those surface domains where a homogeneous β phase exists on a mesoscopic length scale, α phase islands cannot be seen at the very beginning of one oscillation period, but after a short time new small α phase islands are nucleated over the whole lattice and their mean distances fulfill the above conditions for synchronized oscillations independent of the lattice size. If we start the simulation of the system without nucleation on a lattice covered mainly by the β phase, the behavior is completely different. The initially existing α islands grow but the surrounding β phase remains homogeneous because spontaneous nucleation is lacking and the phase of the surface can only change directly at the border between the α and β phase depending of the local A coverage, i.e., the nucleation and therefore the growth of the α phase inside a homogeneous β phase island is impossible. The α phase islands grow to some extend until the adsorbate coverage changes from B to A and the growth of the β phase starts again. This leads to coexisting, almost homogeneous α and β phase islands that travel over the lattice. This can clearly be seen in Fig. 6.

In real systems the existence of the surface phase nucleation is well known but has not been investigated experimentally in detail yet, in contrast to the initial growth of small surface phase domains [40]. The nucleation rate should depend on the temperature $\gamma = \gamma(T)$, but it is almost impossible to achieve an isolated variation of the nucleation rate under experimental conditions because all other parameter such as CO desorption and CO diffusion also strongly depend on the temperature. It might thus be very difficult to experimentally verify the mechanism behind the SR phenomenon in our model but there should be other systems where it is feasible.

The nucleation of dynamic surface defects as an internal process generates globally synchronized oscillations in our model for $CO + O_2/Pt(110)$ reaction via SR. As has been shown in Ref. [32] this mechanism is a simple and realistic alternative to the mechanism of self-synchronization via the gas phase as it is proposed to date. Furthermore, our model can explain [33] in an elegant way the experimentally observed growth rate [40] as well as the experimentally determined critical coverages [36] that are observed in systems showing adsorbate driven surface reconstruction.

Because noise is always present in real systems, this type of internal SR should be a very general phenomenon and it may be the reason for cooperative phenomena and internal synchronization via noise processes in many physical, chemical, and biological systems where is has not been investigated experimentally or even suspected to be present to date. This especially holds for systems that exhibit cooperative phenomena or inherent oscillations that are synchronized on macroscopic length scales. In this case often noise effects are supposed to be negligible, but as shown above they can also be the microscopic origin of those cooperative phenomena. We believe that SR may thus be an internal regulation mechanism of extreme adaptability. This conclusion is drawn to search for internal SR in a variety of experiments, because to date experiments designed to study the role of internal noise have been inconclusive [41].

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