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Noise-induced wave nucleations in an excitable chemical reaction

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We study both experimentally and numerically the temporal coherence of noise-induced wave nucleations in excitable media subjected to external fluctuations with finite correlation time. The experiments are performed with the light-sensitive variant of the Belousov-Zhabotinsky (BZ) reaction forced by an exponentially correlated dichotomous fluctuating illumination. We find that there exists an optimal correlation time for which nucleations coherence reaches a maximum. The same behavior is obtained in numerical simulations with a stochastic Oregonator model, modified to describe the light-sensitive BZ reaction.

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Wave initiation in excitable media has been the subject of vast studies in different fields. Nucleations of spiral waves on cardiac tissue, for example, seem to have great relevance in cardiac diseases like arrhythmia [1]; pathologically synchronized firing activity of thalamus neurons has been related to Parkinsonian tremor [2], and random cyclic adenosine monophosphate (cAMP) wave emission permits aggregation in certain amoebae, which fight starvation with this strategy [3,4]. In chemistry, the Belousov-Zhabotinsky (BZ) reaction has been extensively used to investigate wave initiation in subexcitable conditions under the effect of electric or illumination fluctuations [5–7].

Recently, the phenomenon of coherence resonance (CR) has attracted much attention: A sequence of noise-induced excitations shows maximal coherence at a certain optimal noise amplitude [8,9]. Most of the research on CR was focused on the case of white noise [6,7,10,11]. White noise is a good approximation as long as the intrinsic time scales of the deterministic system are much larger than the correlation time of the external fluctuations. This is the case, for example, in neuronal dynamics where a neuron can be externally forced by another randomly bursting neuron. In general, when deterministic and stochastic time scales are not well separated from each other, not only the amplitude but also the temporal correlation is expected to influence noise-induced phenomena, such as CR. Numerical studies revealed the existence of optimal correlation time for the coherence of noise-induced excitations [12,13], but no experimental proof has been given yet. The purpose of this Communication is to show this phenomenon experimentally in a light-sensitive BZ medium subjected to an exponentially correlated dichotomous fluctuating light intensity, and numerically using a stochastic Oregonator model modified to account for the light sensitivity of the reaction. Moreover, we connect the optimal correlation time with the system dynamics.

In an open gel reactor we maintain the BZ system in a stationary nonequilibrium state [14]. The light-sensitive catalyst ruthenium (II)-bipyridyl [Ru(bpy)] is immobilized in a silica gel matrix (0.3-mm thick) of size 18×15 mm^2. A video projector controlled by a computer is used to illuminate the catalyst-loaded gel layer via 8-bit gray level light. Varying the applied light intensity the system can be driven from an oscillatory (low intensity) through an excitable to a nonexcitable regime (high intensity) [15]. The chamber of the reactor (volume 70 ml) is continuously fed with fresh BZ solution ($[BrO_3^-]=2M$, $[H_2SO_4]=0.28M$, $[CH_2(COOH)_2]=0.15M$, $[Br^-]=0.06M$) at a pumping rate of 100 ml/h. The temperature is kept constant at 25 ± 0.5 °C by means of circulating water from a thermostat. Light from the video projector reaches the gel after passing through a bandpass filter ($\lambda=470$ nm, transmission 85%) to enhance the contrast, and a polarizer (10° with respect to the transmission axis) to adjust the intensity (the video projector emits polarized light). Every second transmitted light is captured by a (CCD) camera connected to a second computer and digitized with a frame grabber (512×512 pixel frames) for later processing.

The light pattern projected onto the gel layer is composed of two vertically aligned small squares of size 1.5×1.5 mm^2 superimposed to a uniform background illumination at a light intensity I_B for which the system is excitable (Fig. 1). The upper square is illuminated at intensity I_0 which also always belongs to the excitable regime of the reaction. Although at constant intensity I_0 no wave nucleation occurs the medium supports the propagation of trigger waves. In the lower square the light intensity fluctuates around I_0 according to a random telegraph process, i.e., $I(t)=I_0+\Delta I\eta(t)$ where $\eta(t)$ is a dichotomous signal assuming alternatively the values +1 and −1. Each intensity level lasts an exponentially distributed random time. Therefore $\eta(t)$ has an exponentially decaying correlation function with correlation time τ.

At light intensity $I_0-\Delta I$ the medium is oscillatory. Phase waves can develop which eventually transform into trigger waves at the boundary of the square or when the light intensity becomes equal to $I_0+\Delta I$. Under fluctuating illumination trigger waves are nucleated randomly. Among the three control parameters of the fluctuations I_0, ΔI, and τ, in the following we focus on the effect the correlation time has on the coherence of the stochastically nucleated sequence of waves.

The period of oscillations at intensity $I_0-\Delta I$ (about 40–45 s for the chosen recipe concentrations) has been controlled

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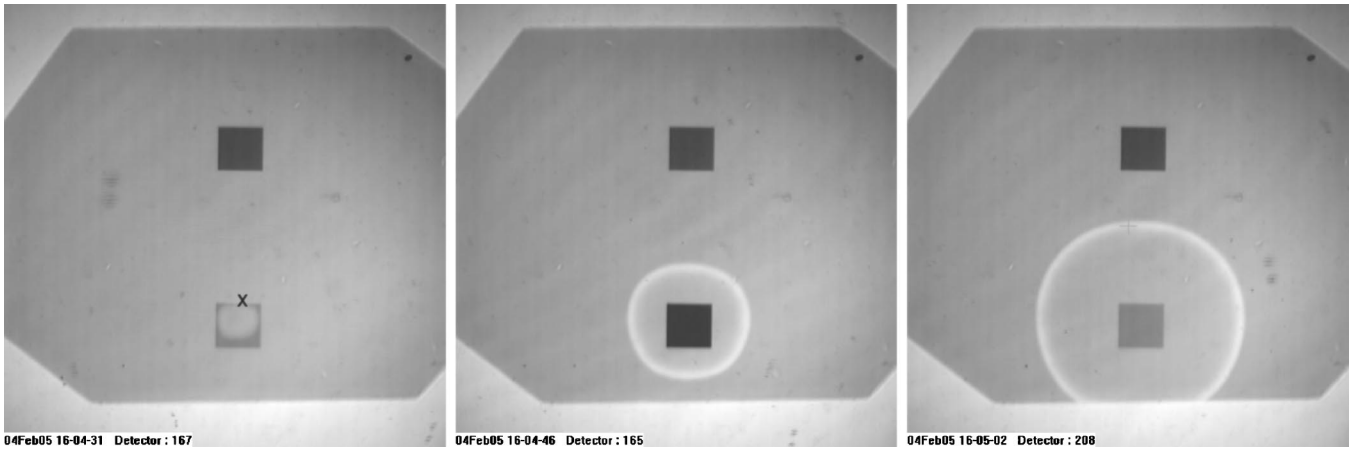

FIG. 1. Three snapshots of a wave nucleation taken at equal time intervals of 15 s. Wave activity is shown through the oxidized form of the catalyst. The cross indicates the detector position where passing wave fronts are counted.

during the experiments. When the shift in the oscillation period due to aging processes became of the order of 10%, at the latest after 1 h, the gel layer was renewed. At least 75 nucleations were detected for each value of the correlation time studied (except for $\tau=2$ s for which nucleation events occurred too seldom to reach this number).

Experimentally measured sequences of noise-induced excitations are shown in Fig. 2. The gray level is a measure of the local concentration of $Ru(bpy)_3^{3+}$. Obviously, both the nucleation rate and the regularity of the sequence depend strongly on the correlation time τ. When fast fluctuations are introduced into the system, nucleation events are randomly induced (top of Fig. 2). As the correlation time is increased, both the nucleation rate and the coherence improves (middle of Fig. 2). For higher correlated fluctuations, bursting behavior emerges and the response becomes again more irregular (bottom of Fig. 2).

In order to get deeper insight into this dependence, from our data we have determined the time between two successive wave nucleations and calculated the mean interspike period $\langle t_p \rangle$. We find that $\langle t_p \rangle$ and hence the nucleation rate can be controlled by the correlation time as shown in Fig. 3 (top). To quantify the coherence of the sequence, in Fig. 3 (bottom) the relative fluctuations of the interspike time

$$R_p = \frac{\sqrt{\langle (t_p - \langle t_p \rangle)^2 \rangle}}{\langle t_p \rangle} \tag{1}$$

are plotted as a function of τ. The results give experimental evidence of CR with respect to the correlation time of the external fluctuations as predicted in Ref. [16]. Best coherence is reached at $\tau \simeq 10$–20 s. Note that this optimal value for the correlation time is much smaller than any oscillation period in the reaction.

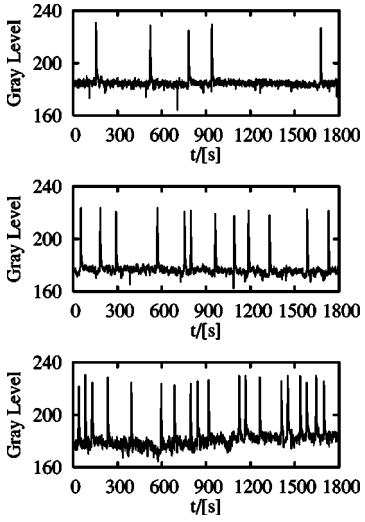

FIG. 2. Noise-induced excitations for three different correlation times of the fluctuating light intensity. From top to bottom: $\tau=5$, 15, and 40 s. The results are obtained by gray level time tracking at the detector point marked by the cross in the left frame of Fig. 1.

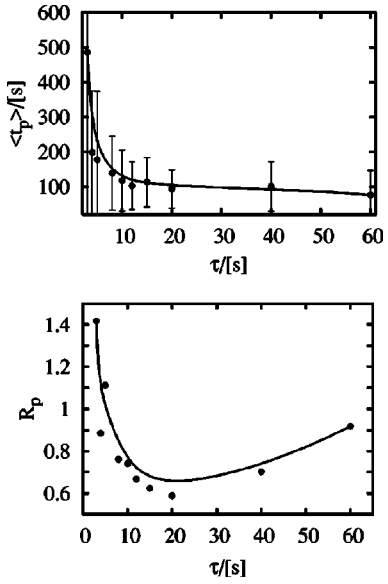

FIG. 3. Experimental results for the mean interspike periods (top) and for the normalized fluctuations of the interspike times (bottom) as a function of the correlation time. Continuous lines are obtained with a spline fitting algorithm, just for illustrative purpose.

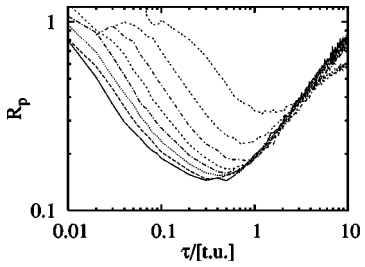

FIG. 4. R_p values obtained from numerical simulations of Eq. (2) with $\phi_0=0.0075$ and $\Delta\phi$ varying from 0.3 ϕ_0 to 1.0 ϕ_0 at steps of 0.1. The smallest value of $\Delta\phi$ corresponds to the upper curve and the largest one to the lowest curve on each graph. Each point is an average over 2×10^4 data.

The experimentally observed behavior has been recovered in numerical simulations of the two-component Oregonator model for the light-sensitive BZ reaction

$$\frac{du}{dt}=\frac{1}{\varepsilon}\left(u-u^2-(fv+\phi_0)\frac{u-q}{u+q}\right),$$

$$\frac{dv}{dt}=u-v. \tag{2}$$

Here u and v stand for the dimensionless concentration of bromous acid and the oxidized form of the light-sensitive catalyst $Ru(bpy)_3^{3+}$, respectively. ε and q are scaling parameters, f is a stoichiometry parameter, and ϕ_0 represents the photochemically induced bromide flow which is assumed to be proportional to the applied light intensity I_0. From the recipe concentrations we obtain $\varepsilon=0.0766$ and $q=0.002$. Together with $f=1.4$ we use these parameter values to numerically integrate Eq. (2) using a second-order Runge-Kutta algorithm with a time step $\Delta t=0.001$ time units [t.u.].

When ε, f, and q are kept fixed, ϕ_0 controls the kinetic regime in the same way as I_0 does in the experimental system: For small ϕ_0 the kinetics is oscillatory and for $\phi_0>\phi_{hb}=4.43\times10^{-3}$ it becomes excitable via a supercritical Hopf bifurcation. The use of a zero-dimensional model is justified in our calculations because in the experiments the extension of the fluctuating light pattern is small enough to neglect diffusion.

To account for the fluctuating light intensity in the experiment we assume the photochemically induced bromide flow ϕ_0 as a stochastic quantity according to

$$\phi_0\rightarrow\phi(t)=\phi_0+\Delta\phi\,\mathrm{sgn}\{\cos[\alpha(t)]\}, \tag{3}$$

where the phase $\alpha(t)$ has the form

$$\alpha(t)=\pi\sum_j\theta(t-t_j),\quad t_j=t_{j-1}+s. \tag{4}$$

Here θ denotes the Heaviside step function and s is an exponentially distributed random variable with probability density $p(s)=(1/\tau)\exp(-s/\tau)$. The fluctuating part in Eq. (3) has zero mean value, variance $(\Delta\phi)^2$, and correlation function $C(t_1,t_2)=\Delta\phi^2\exp(-|t_1-t_2|/\tau)$.

As in the experiments the parameters ϕ_0 and $\Delta\phi$ are chosen such that under deterministic conditions ϕ_0 and $\phi_0-\Delta\phi$ correspond, respectively, to excitable and oscillatory kinetics. Under constant illumination with an intensity corresponding to ϕ_0 the system remains forever in the linearly stable rest state. In the presence of noise it can be driven into the excited state even if the mean value of the fluctuations ϕ_0 is kept within the excitable regime.

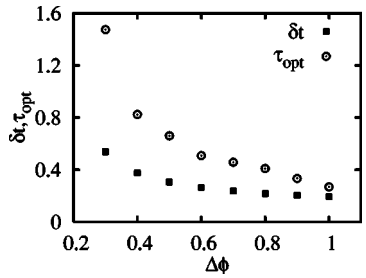

FIG. 5. Comparison between the optimal correlation time (location of the minima in the R_p curves in Fig. 4) and the excitation time for different noise amplitudes.

The results for $R_p(\tau)$ for different noise amplitudes (Fig. 4) show the same trends as those observed in the experiments (Fig. 3). Numerically, we have checked that this phenomenon holds for an extended interval of the mean value ϕ_0, covering almost the whole excitable regime. Moreover, the optimal value of τ decreases with the amplitude of the noise $\Delta\phi$. From the numerical simulations follows that for $\tau<\tau_{opt}$, small fluctuations [with respect to $(\Delta\phi)^2$] with longer correlation time have the same effect on the coherence of the noise-induced sequence as stronger fluctuations with shorter correlation time. In the opposite case $\tau>\tau_{opt}$, excitation bursts are observed and coherence is nearly independent of the noise strength.

Let us consider the system is at the stable fixed point (u_+,v_+) and at excitability ϕ_+. If the system is driven at excitability ϕ_- the u nullcline is shifted, leading the system into the oscillatory regime, i.e., the point (u_+,v_+) becomes unstable and the system leaves it approaching the stable periodic orbit. After a time δt the system reaches the excitation threshold (u_{thr},v_{thr}) which is beyond the unstable branch of the u nullcline for ϕ_+: from this point and at excitability ϕ_+ the system is able to perform an excitation loop. τ_{opt} turns out to be of the order of magnitude of minimal δt to induce an excitation (compare Fig. 5).

Summarizing, we have demonstrated experimental evidence for CR with respect to the correlation time adding nonwhite illumination noise to the light-sensitive BZ reaction. The optimal value to achieve CR scales with the fast deterministic dynamics of the system. At this value, the system's response is almost periodical.

The experimental results are in good agreement with numerical simulations of the underlying Oregonator model. Numerical results obtained with fluctuating spatiotemporal illumination have shown that in the spatially extended BZ medium there is also CR with respect to the correlation length of the noise [16]. The experimental verification of this prediction remains a challenge for future research.

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