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Noise-Induced Entrainment between Two Coupled Chemical Oscillators in Belouzov-Zhabotinsky Reactions

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Abstract. The entrainment dynamics of two chemical-oscillators in an excitable medium separated by some distance *d* was experimentally studied in the light-sensitive Belousov-Zhabotinsky reaction under application of external noise. Two chemical-oscillators were always spontaneously synchronized at sufficiently small *d* or frequency difference in the absence of noise. For insufficient distance to entrain each other, however, the frequency and phase were self-synchronized by application of noise. Increasing the noise intensity the entrainment frequency area between two oscillators increased and showed a maximum at a certain noise intensity, that is, an optimum noise intensity existed. We call this the noise-synchronaization phenomenon.

1. Introduction

The Belousov-Zhabotinsky (BZ) chemical oscillator using ionic absorbent beads has been paid much attention to understand the coupling behavior among spatially discrete oscillators (MASELKO and SHOWALTER, 1989). As expexted, then, the ordinary resonance phenomenon was observed and additional interesting aspects, such as diffraction of chemical waves due to non-uniformity of the size of beads and popping dynamics of synchronization from in-phase to anti-phase, were found (OOSAWA and KOMETANI, 1996; NISHIYAMA and MATSUYAMA, 1997).

On the other hand, recently, for optical, biological and chemical systems, different types of resonance phenomena related to the noise-induced one (KAI *et al*., 1979) have been studied extensively (MOSS *et al*., 1994; JUNG and MAYER-KRESS, 1995; GANG *et al*., 1996; GAMMAITONI *et al*., 1998). It is well known that noise can enhance a hidden periodicity in certain nonlinear systems, that is, external noise improves the periodicity of a signal. This phenomenon is called the stochastic resonance (SR). In a single chemical oscillator system (the continuously stirred tank reactor: CTSR), for example, the evidence of SR has been reported recently (GUDERIAN *et al.*, 1996), where the signal-to-noise ratio (SNR) had a maximum at a certain noise intensity, when a periodic flow rate was imposed on the system externally superimposed with noise, under conditions near the Hopf bifurcation point from excitable focal steady state to periodic spontaneous oscillation. For spatio-temporal chemical systems on the other hand, the optimal noise intensity provided a longer life of traveling chemical waves without deforming the wave profile in sub-excitable media (KADAR *et al.*, 1998). However, there has been no experimental report about noise effect on spatial entrainment in discrete chemical oscillators. In this letter we will report a strange noise effect to discrete chemical ones, such as noise-induced entrainment, which is different from SR.

2. Experimental

Figure 1 shows the experimental set-up. The experiment was carried out by use of the photo-sensitive Belousov-Zhabotinsky reaction system which contains the ruthenium(II)-bipyridyl-complex ($Ru(bpy)_3^{+2}$) as a catalyst (YONEYAMA *et al.*, 1993; STEINBOCK and MUELLER, 1993). The usual chemicals and their mixture rates were used such as 0.833 M H_2SO_4, 0.277 M $KBrO_3$, 0.083 M KBr and 0.111 M $CH_2(COOH)_2$ at room temperature 20 ± 1 C. The catalyst was absorbed into ionic absorbent beads with a diameter of 0.8 mm with concentrations in the range from 4.17 to 20.83×10^{-6} M/g. One period of oscillation using these concentration ranges of a catalyst was from 220 to 320 s. Two beads were chosen and placed in the BZ solution at a distance *d* so that the oscillatory reaction was started only on the beads. Each bead oscillated with a particular period, i.e. each frequency (*f*) depending on small differences of their size and concentration of absorbent catalyst.

A photo-illumination usually suppresses oxidation in the BZ solution and even can stop the oscillation at sufficiently strong intensity. Therefore we could control a stop and start of the reaction by illumination intensity. The illumination was supplied by use of a slide projector with a filter to cut infrared-lights off in order to avoid heating-up of our solutions. The light intensity at the surface of the BZ solution, the depth of which was about 1.5 mm, was changed from 0 to 0.7 mW/cm^2. However as we did not calibrate the absolute intensity of the light on beads, the relative value is meaningful. (The absolute intensity on beads could be very slightly weaker than the above value because of absorption by the solution.)

Noise was produced by a computer using a software for random digits, power spectrum of which was completely flat between 3×10^{-5} to 10^{-1} Hz. The externally imposed noise intensity I_N was defined as $I_N^2 = \langle I_N(0)^2 \rangle$ after calculating its autocorrelation function. The oscillation was detected as a video image through a CCD camera which was recorded on a magnetic tape and a hard disk of the computer for further analysis. The analysis of video images was carried out by use of NIH-image software.

3. Result and Discussion

The entrainment between two separated beads with a distance *d* and with various different frequencies in the excitable medium is shown in Fig. 2. Obviously at sufficiently close *d* (~typically half of a bead diameter) for all the cases the two different beads were

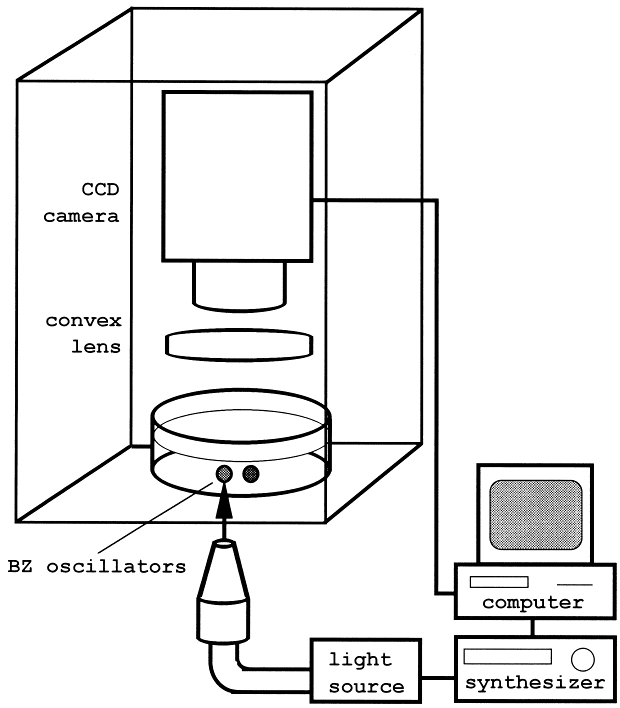

Fig. 1. Experimental set-up.

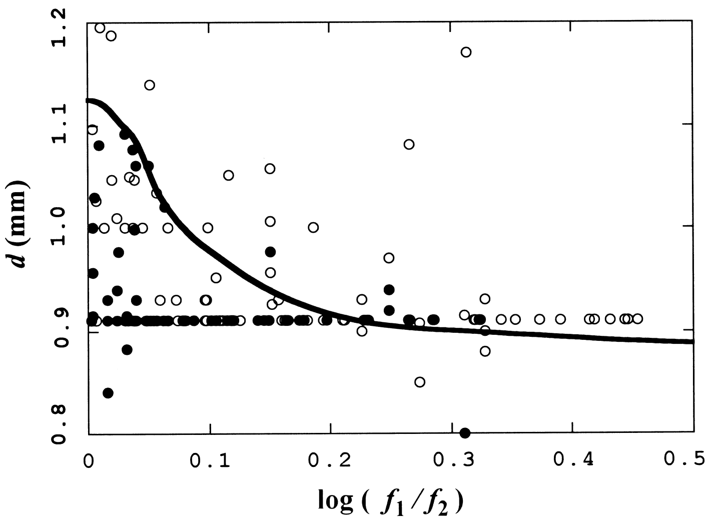

Fig. 2. Entraiment between two beads separated with distance d. Open circle: non-entrainment. Full circle: entrainment.

always synchronized with each other. The smaller the frequency difference between two beads, the easier the entraiment of each other as expected, that is, the synchronization took place with larger d for smaller frequency difference. Based on these results we have properly chosen d in order to study the noise effect to the entrainment phenomenon. First, we took one bead to determine the influence of light intensity on its oscillation. Figure 3 displays the oscillation of a bead under illumination of light. When a bead was illuminated during its oscillation by light, as seen in Fig. 3, there was no light intensity dependence of the oscillatory period. Instead the oscillatory amplitude gradually decreased with increase of the light intensity. Increasing light intensity its oscillation suddenly stopped at a certain light intensity. The threshold intensity I_{Nc} was in the present case 0.34 mW/cm^2. Beyond the value no chemical oscillation was observed. By application of the light stronger than 0.34 mW/cm^2 the oscillation was even stopped completely.

Typical examples of resonance curves of two beads are shown in Fig. 4 in a logarithmic scale, where f_i and f'_i are, respectively, the spontaneous and entrained frequencies of the two beads (i = 1 and 2). Here one oscillator was chosen for noise illumination (as shown in Fig. 1) and the other was not illuminated. The illuminated oscillator was then always entrained by the other oscillator which oscillated with own spontaneous period. The flat area indicates that the entraiment occurs, which we call hereafter the *entrainment width* (EW). In the absence of noise, the center of the entrainment shifted to rather larger value of ratio of f_1/f_2. Increasing the noise intensity I_N , in the small f_1/f_2 range resonance was induced, i.e. the flat area extended into the area of small values of the ratio f_1/f_2. Upon further increasing I_N desynchronization started at the larger f_1/f_2 ratio indicating that the flat area (EW) was shrunk there. Thus EW gradually increased with I_N , had its maximum value around I_N^* = 0.25 mW/cm^2 and then beyond I_N^* decreased, i.e. it has a optimal value of noise. For I_N = 0.66 mW/cm^2, EW becomes a half of that at I_N^*. In Fig. 5 such a noise

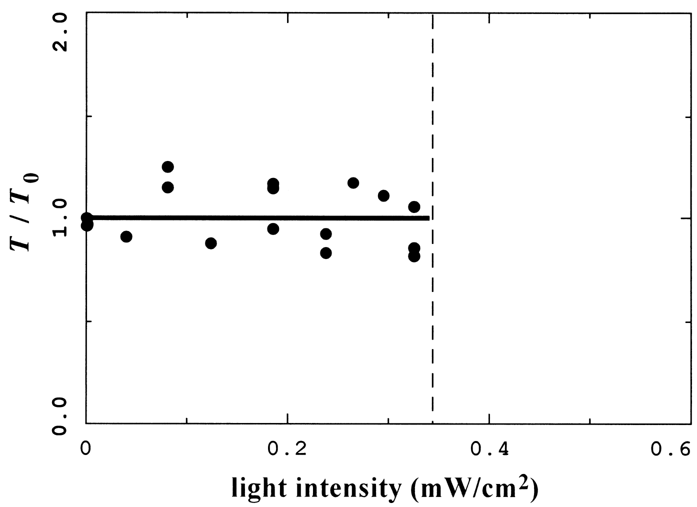

Fig. 3. Oscillation under illumination. T_0:spontaneous period of oscillation without light illumination.

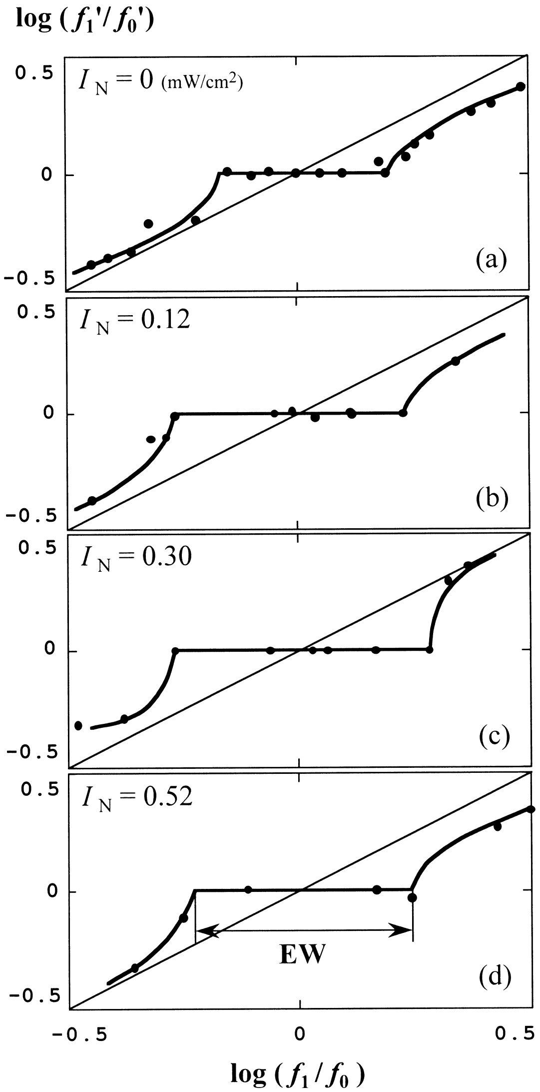

Fig. 4. Resonance curve of two BZ oscillators. The flat area in the figure indicates that the synchronization takes place. A straight line indicates a non-entrainment line. I_N :noise intensity.

intensity dependence of EW is summarized. Clearly it displays the existence of an optimal noise intensity for synchronization.

In the above we focused only the one-to-one synchronization. However the one-to-two synchronization is also observed (however it is much smaller width than that in the one-to-one case). Due to application of noise it disappeared and instead EW of the one-to-one synchronization was extended. The mechanism of this dynamics is not understood yet at this moment.

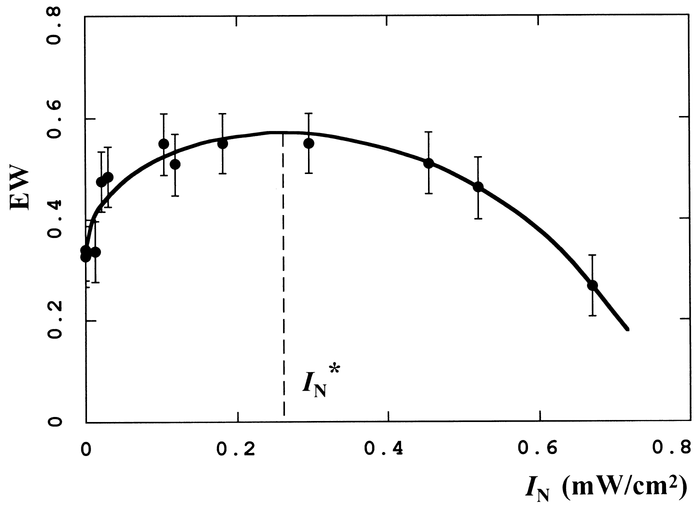

Fig. 5. Noise dependence of entrainment width (EW). At a certain noise intensity I_N^* the maximum EW can be observed. The bars in the figure indicate error scatters. I_N : noise intensity I_N^*: noise intensity at maximum EW.

4. Summary

Application of noise leads to an extended synchronization area in the frequency domain, that is, noise induces easier synchronization of two BZ oscillators. It happens strongly at a certain noise intensity I_N^*, that is, better resonance takes place at the most appropriate noise intensity. This is an aspect somewhat similar to SR, if SNR in SR is replaced by EW in this study (FUJII *et al.*, 1999). Therefore we have named it noise-synchronization (KAI *et al.*, 1979; FUJII *et al.*, 1999). In the present experiment, not only one-to-one entrainment but also one-to-two entraiment were observed. However by application of noise the higher degree of entrainment disappeared. Instead the width of entrainment was increased. The corresponding simulation has been done also using Amemiya's model (AMEMIYA *et al.*, 1998), but the extended width of resonance due to noise application could not be explained (FUJII, 1999). We believe, this is a new nonlinear phenomenon induced by noise. Further studies are neccessary for clarification of its mechanism.

Finally we would like to mention an application of the idea of the noise synchronization found in the present study to information transmission processes, for example in nerves cells and related sensors. The noise synchronization phenomenon shows us that all oscillators, which are not entrained in the absence of noise and oscillate with independent frequencies and phases each other, can be entrained to a leading one if the optimum noise is applied to oscillators except the leading one. That means that the information (e.g. its frequency and phase) of the leading oscillator is transmitted to other locations through many oscillators entrained in the presence of certain noise intensity. It may happen more likely in information transmission of a higher organism.

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