

Noise-induced phase locking in coupled coherence-resonance oscillatorsMasako Ohtaki,¹ Takayuki Tanaka,² and Kenji Miyakawa^{1,2,*}¹*Advanced Materials Institute, Fukuoka University, Fukuoka 814-0180, Japan*²*Department of Applied Physics, Fukuoka University, Fukuoka 814-0180, Japan*

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We investigate the effects of additive noise on coupled excitable chemical oscillators, particularly focusing on how oscillatory coupled modes can be induced by noise. We find that phase locking in the weak coupling regime occurs through coherence resonance, although the resulting phase locking modes are apparently similar to those in coupled deterministic oscillators. Experimental observations are approximately reproduced in a numerical simulation with a forced Oregonator reaction-diffusion model.

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I. INTRODUCTION

Phase locking in coupled or periodically forced nonlinear self-sustained oscillators is a typical phenomenon of self-organization in physical, chemical, and biological systems [1–4]. External periodic forcing above a threshold induces $n:m$ phase locking pattern with m firings for n forcing periods, depending on the forcing amplitude. For a periodic signal below a threshold, the addition of a certain amount noise can also induce phase locking. This phenomenon, called noise-enhanced phase locking [5–7], is a kind of stochastic resonance. Self-sustained oscillators have primarily their natural frequencies and amplitudes that can be controlled by random forcing in the process of phase locking. In contrast, excitable oscillators do not possess any eigenfrequencies. In excitable systems driven by noise alone, however, the oscillation is induced above a certain noise intensity and becomes quite regular for an optimal noise intensity. This phenomenon is referred to as coherence resonance (CR) [8–12]. In a stochastic oscillator, i.e., a “CR oscillator,” the noise-activated time scale is reconciled with the time scale characteristic of the system at an optimal noise intensity. This has been experimentally confirmed in excitable optical [13] and chemical reaction systems [14]. In coupled systems consisting of stochastic oscillators, therefore, more fruitful noise effects can be expected, compared with those of deterministic oscillators. Recently, Postnov *et al.* have found that a coupled excitable system can possess several noise-induced oscillatory modes, using an array of excitable units using a monovibrator circuit [15].

In this paper, we experimentally and numerically investigate behaviors of noise-induced oscillatory modes in an excitable coupled reaction-diffusion system. In the experiment we use a discrete type of the Belousov-Zabotinsky (BZ) reaction as the excitable units. The coupling in this system is due to mass diffusion, so that coupling is inherently accompanied by a time delay. A time-delayed coupling is commonly seen in the information transmission between biological oscillators. We find that the some phase locking occurs in

an apparently similar way to that of coupled deterministic oscillators. Some experimental results are numerically simulated using a forced Oregonator reaction-diffusion model.

II. EXPERIMENT

Experiments were carried out with a localized reaction system in which the tris-(2,2'-bipyridine) ruthenium (II) complex $[\text{Ru}(\text{bpy})_3]^{2+}$ was immobilized in cation exchange beads of submillimeter size [16,17]. The experimental setup is almost the same as those described in an earlier paper [14]. The initial composition of the BZ reaction solution was $[\text{NaBrO}_3]=0.45\text{M}$, $[\text{NaBr}]=0.05\text{M}$, $[\text{CH}_2(\text{COOH})_2]=0.3\text{M}$, and $[\text{H}_2\text{SO}_4]=0.4\text{M}$. Reagent grade chemicals were used without further purification. The cation exchange beads (DOWEX, 50W-4X) of 0.5 mm in diameter were loaded with a solution of $[\text{Ru}(\text{bpy})_3]^{2+}=1.2 \times 10^{-5}$ mol/g beads. At this composition, the system was initially in the oscillatory regime. The excitable steady state was realized by taking advantage of the high photosensitivity of $\text{Ru}(\text{bpy})_3^{2+}$ [18]. The period of oscillation increased monotonically with increasing illuminated light intensity I , and finally the excitable steady state appeared for a light intensity of more than about $I_c=9$ mW. Then the period under the dark was estimated by extrapolation to be approximately 40 s. In the experiment we fixed I at 10 mW. We employed an electric field as an external forcing to stimulate the excitable medium, since the electric field has pronounced effects on dynamic behaviors of chemical waves.

Sensitive responses to electric fields are related to the fact that most chemical species relevant to wave propagation are ionic. The electric field induces additional drifts of ions, which result in a change in the ion concentration around the electrodes. In order to exert such a field effect on only one excitable bead, we placed one bead in contact with one electrode, and placed another excitable bead d apart, as shown in Fig. 1. Here the former bead was tentatively termed the oscillator 1 (OSC1), and the latter bead was termed the oscillator 2 (OSC2). In this experimental setup, only OSC1 is stimulated by an external electric noise. In fact, noise-induced waves were always initiated at the contact point of the bead with the electrode. In contrast, OSC2 is stimulated only through the firing of OCS1. When OSC1 was removed

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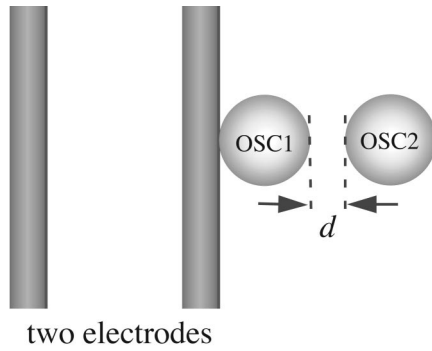


FIG. 1. Schematic representation of the experimental setup. OSC1 contacts one of two electrodes. Electrodes are placed 2 mm apart and parallel to the surface of the reaction medium. The array of beads is placed perpendicular to the two electrodes.

from the reaction medium, in fact, no waves were excited on OSC2 in the range of the noise amplitude investigated in this study. The added electric noise is represented by $\beta\xi_\delta(t)$, where β is the amplitude and $\xi_\delta(t)$ are random numbers equally distributed between -1 and 1 with the duration time δ . In the measurement, we chose the duration time of $\delta = 0.225$ s.

The coupling behavior can be controlled by varying d , so that d acts as a coupling strength. When $d > 100 \mu\text{m}$, the two oscillators were always decoupled for any value of the noise amplitude β . When $d \leq 70 \mu\text{m}$, to the contrary, 1:1 entrainment always took place for any β . This region is in the tight coupling regime. At an intermediate value of d , corresponding to the weak coupling regime, various modes of entrainment appeared depending on β . The time evolutions of the two oscillators and the corresponding interspike interval histograms (ISIHS) at $d = 100 \mu\text{m}$ are shown in Fig. 2. For small β , irregular firings were induced on OSC1, but OSC2 did not exhibit any response to them. We evaluated the degree of

coherence of the noise-induced oscillation by using R , defined by $R = \sqrt{\langle T^2 \rangle - \langle T \rangle^2} / \langle T \rangle$, where $\langle T \rangle = (1/N) \sum_{i=1}^N T_i$, N is the number of firings, and T_i is the time interval between the i th and $(i+1)$ th firing events. With increasing β , the oscillation of OSC1 became more regular, and R passed through a maximum at $\beta_{\text{opt}} = 0.65$ V. This suggests that CR occurred on OSC1. The value of β_{opt} is consistent with the earlier result [14]. With the occurrence of CR, phase locking appears with the period ratio of $T_1:T_2 = 1:2$, where T_1 and T_2 denote the periods of OSC1 and OSC2, respectively [Fig. 2(d)]. It should be noted that the phase locking between two oscillators is governed by CR, although such a mode is apparently similar to that in coupled deterministic oscillators. With a further increase in β , the oscillation of OSC1 became irregular, accompanied by a decrease of the firing rate [Fig. 2(b)]. A nonresonant state appeared, as seen from the ISIHS in which incommensurate periods are clearly distinguishable [Fig. 2(e)]. When $\beta = 0.85$ V, phase locking of $T_1/T_2 = 1$ occurred [Fig. 2(f)], although the firing of OSC1 became more irregular and its averaged period was approximately doubled [Fig. 2(c)].

Next we investigated the d dependence of entrainment for fixed $\beta (= 0.65$ V). For $d \leq 80 \mu\text{m}$, the interaction between two oscillators is strong, so that the mode of 1:1 always appears as shown in Figs. 3(a) and 3(b). With increasing d , the interaction between them no longer becomes strong enough to keep the 1:1 mode, so that coexistence of the 1:1 and 1:2 modes, namely, two-mode behavior, occurs as a precursor of stable entrainment of 1:2 [Fig. 3(c)]. Finally the 1:2 resonance behavior appears in the weak coupling regime [Fig. 3(d)].

To further characterize the coherent behavior, we introduce the ratio n_2/n_1 , where n_1 and n_2 are the number of firings induced on OSC1 and OSC2, respectively. This quantity provides a measure of the stability of phase locking: in stable phase locking, the value of n_2/n_1 does not change so sensitively with variations of the external control parameters.

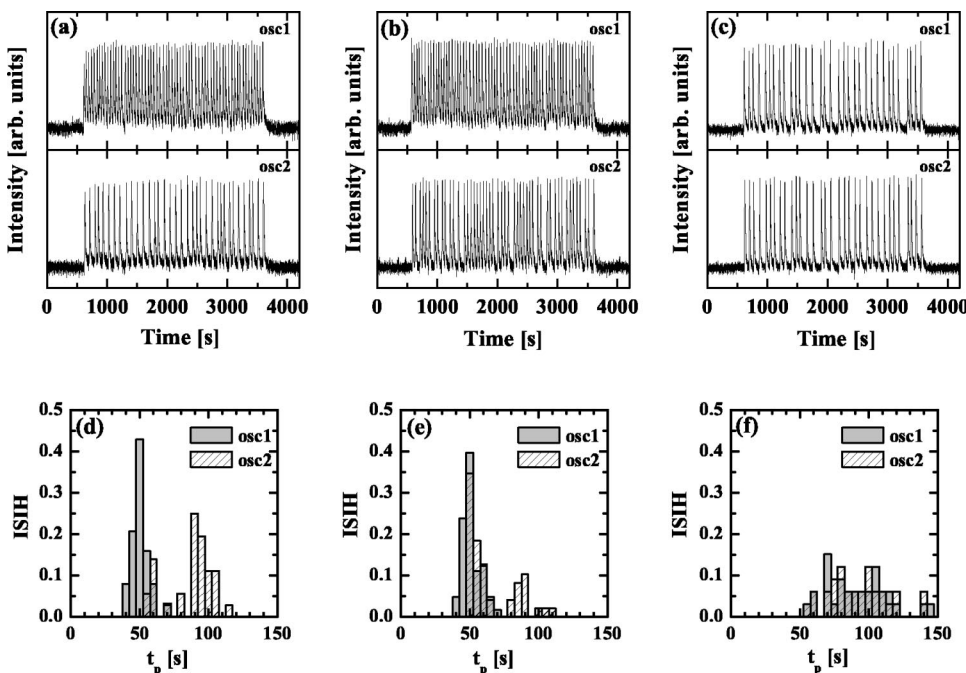


FIG. 2. Time series of the light intensity from OSC1 and OSC2, and interspike interval histograms as a function of the noise amplitude β for the spacing $d = 100 \mu\text{m}$, where t_p denotes the time interval between successive firing events: (a) and (d) $\beta = 0.65$ V; (b) and (e) $\beta = 0.75$ V; (c) and (f) $\beta = 0.85$ V.

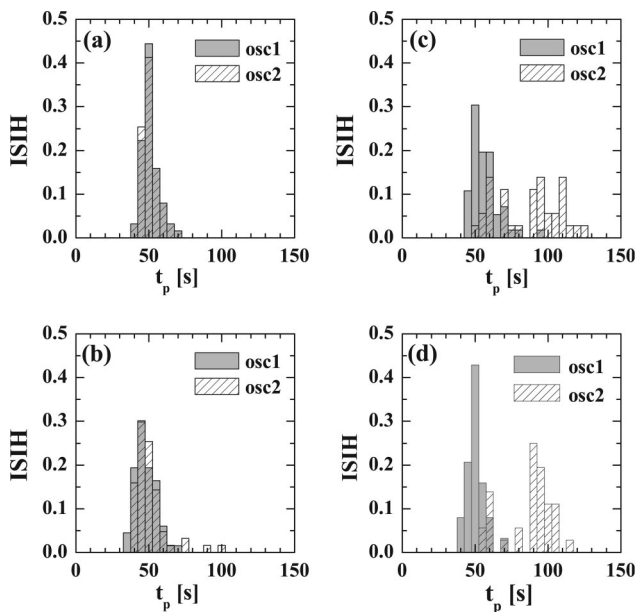


FIG. 3. Interspike interval histograms as a function of the spacing d between two beads for the noise amplitude $\beta=0.65$ V, where t_p denotes the time interval between successive firing events: (a) $d=70$ μm , (b) $d=80$ μm , (c) $d=90$ μm , and (d) $d=100$ μm .

The dependence of n_2/n_1 on the noise amplitude at $d=100$ μm is shown in Fig. 4(a). The remarkable feature is that, with the occurrence of CR at $\beta_{\text{opt}}=0.65$ V, n_2/n_1 takes a nonzero value abruptly. This means that the information transmission between excitable units is enhanced by CR. In

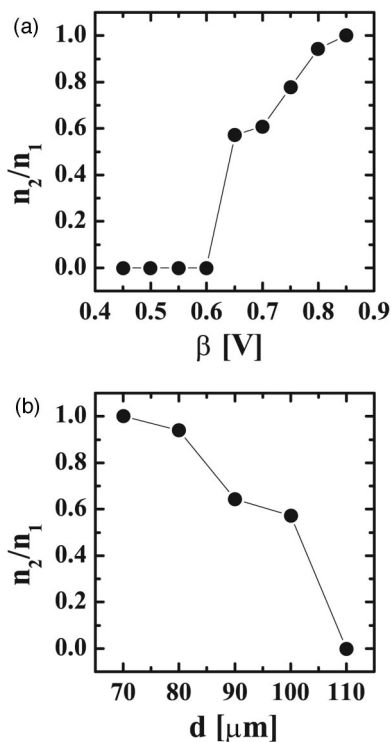


FIG. 4. Ratio of the firing number of OSC2 to OSC1 as a function of the noise amplitude β or the separation d between two oscillators: (a) $d=100$ μm and (b) $\beta=0.65$ V.

the case that β is fixed at 0.65 V, the d dependence of n_2/n_1 is observed only for $70 < d < 110$ μm , as shown in Fig. 4(b). This region is just in the weak coupling regime. For a variation of d or β , n_2/n_1 does not change smoothly but has a plateau near 0.5. This means that entrainment of 1:2 is stable.

III. MODEL

We consider a simple one-dimensional BZ reaction system consisting of interacting excitable units separated by an inactive area, i.e., a diffusion area. Recently, frequency transformations of chemical signals in a similar system were studied using FitzHugh-Nagumo model [19]. We employ the three-variable Oregonator model modified to take into account the effects of an external electric noise. An electric field influences transport processes of reacting ionic species. In reaction-diffusion models the terms describing those additional drifts appear coupled to a gradient term [20,21]. In our experimental setup, however, the electric field is not applied across excitable beads (see Fig. 1). Therefore, such a field-induced drift of ions seems not to occur on the excitable areas under consideration, although in the diffusion area between two electrodes it can occur. Of three fundamental species in the Oregonator model only the inhibitor Br^- is influenced by the electric field, and both the autocatalytic species HBrO_2 and the catalyst $\text{Ru}(\text{bpy})_3^{2+}$ are not directly affected because HBrO_2 is not charged and $\text{Ru}(\text{bpy})_3^{2+}$ is immobilized in the beads. Now we assume that OSC1 is excited by a field-induced change in the concentration of Br^- around the electrode, which initiates waves at the point of contact with the electrode. Then the model equations in excitable areas are given by

$$\frac{\partial u}{\partial t} = \frac{1}{\epsilon} [u - u^2 - w(u - q)] + D_u \frac{\partial^2 u}{\partial x^2}, \quad (1)$$

$$\frac{\partial v}{\partial t} = u - v, \quad (2)$$

$$\frac{\partial w}{\partial t} = \frac{1}{\epsilon'} [fv - w(u + q) + \phi] + \beta \xi + D_w \frac{\partial^2 w}{\partial x^2}, \quad (3)$$

where the variables u , v , and w describe the concentrations of HBrO_2 , the $\text{Ru}(\text{bpy})_3^{2+}$ catalyst, and Br^- , respectively. D_u and D_w denote the diffusion coefficients of HBrO_2 and Br^- , respectively. ϵ , ϵ' , and q are scaling parameters, f is the stoichiometry parameter, ϕ is the light flux, and $\beta \xi$ is the electric noise with the noise amplitude β and Gaussian white noise ξ . Here the noise term is considered only at the end of OSC1, namely, the point of contact with the electrode. Thus the external control parameters are ϕ and β . In the absence of the light flux, these parameters were chosen such that the system was in the oscillatory regime: $q=0.002$, $f=1.4$, $\epsilon=0.01$, $\epsilon'=0.0001$, $D_u=1.0$, and $D_w=1.12$. Then the period was 4.2. With increasing ϕ , the period of oscillation increased monotonically, and finally the excitable steady state appeared. We fixed the illumination intensity at $\phi=0.008$

such that the system was initially in an excitable regime close to a Hopf bifurcation point.

We assume that in the diffusion area separating two excitable beads the reaction terms are absent in the corresponding equations. Then the model equations in this area are given by

$$\frac{\partial u}{\partial t} = D_u \frac{\partial^2 u}{\partial x^2}, \quad (4)$$

$$\frac{\partial v}{\partial t} = 0, \quad (5)$$

$$\frac{\partial w}{\partial t} = D_w \frac{\partial^2 w}{\partial x^2}. \quad (6)$$

In the computations the whole interval was divided into equally spaced grids. When the diffusion area is located between the grid points x_1 and x_2 , the separation d between the two beads is estimated by $d = x_2 - x_1 - 1$. This quantity plays the same role as the separation d in the experiment. The boundary condition at both ends of the interval was taken to be no flux, while that between excitable and diffusion areas was taken to be free.

First, we investigated noise-induced effects on one oscillator. To characterize the degree of coherence, we used R , the standard deviation of the time interval between successive firing events. We confirmed that R passes through a minimum at the optimal noise amplitude β_{opt} , which clearly shows the occurrence of coherence resonance at β_{opt} . The period of coherence oscillation at β_{opt} almost coincided with the period under the dark, in agreement with the experimental observation.

Let now consider the case of interacting CR oscillators placed d apart. We assume that the noise is applied only to the end of OSC1, and OSC2 is stimulated only by firings of OSC1, in the same manner as in the experimental setup. The computation was performed by the improved Euler method with a grid spacing $\Delta x = 0.25$ and time steps $\Delta t = 0.001$. Figures 5(a)–5(c) show the evolution of the power spectra with β at $d = 6$. We can see that, in addition to simple entrainments such as $T_1 : T_2 = 1 : 1$ and $1 : 2$, a two-mode behavior appears at the intermediate value of β , as observed in the experiment. The dependence of the ratio n_2/n_1 on the external control parameters is shown in Figs. 5(d) and 5(e), where n_1 and n_2 are the numbers of firings induced on OSC1 and OSC2, respectively. We can see that there exists a plateau near $n_2/n_1 = 0.5$, except for $n_2/n_1 = 1$. This denotes that in addition to the entrainment of $1 : 1$, the $1 : 2$ mode is also stable against a variation of the external control parameters. When $d \geq 13$, the two oscillators were always decoupled for any value of the noise amplitude β . When $d \leq 2$, to the contrary, $1 : 1$ entrainment always took place for any β . In the range of d from 6 to 8, both $1 : 1$ and $1 : 2$ modes appeared depending on β , which means that this region corresponds to the weak coupling regime observed in the experiment. Thus the numerical simulation approximately reproduces the experimental observations. Comparing Fig. 5(d) with Fig. 4(a), however, there still exists a discrepancy between them; the profiles of n_2/n_1 vs β in the weak coupling regime are opposite to each other.

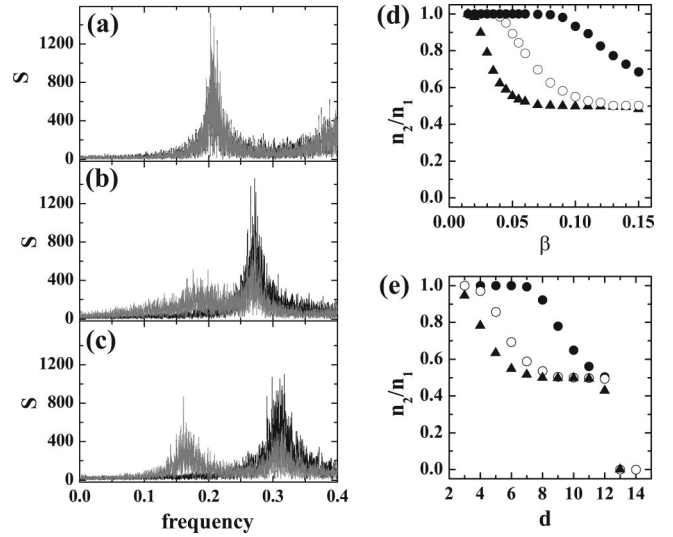


FIG. 5. Power spectral density of noise-induced oscillation on OSC1 (black) and OSC2 (gray) at $d = 6$: (a) $\beta = 0.03$, (b) $\beta = 0.07$, (c) $\beta = 0.1$; and the ratio of the firing numbers of OSC2 to OSC1 as a function of the noise amplitude β or the separation d : (d) $d = 3$ (black circle), $d = 6$ (open circle), and $d = 9$ (black triangle); (e) $\beta = 0.03$ (black circle), $\beta = 0.07$ (open circle), and $\beta = 0.1$ (black triangle).

This affects the order of the occurrence of entrainment with β . In the model, $1 : 1$ entrainment, two-mode behavior, and $1 : 2$ entrainment occur in that order, while in the experiment they occur in the opposite order. This probably comes from the difference in the β dependence of firing. In the model the firing number increases monotonically with β , while in the experiment it increases until β_{opt} is reached and then decreases to zero with increasing β . In addition, as seen from Fig. 5(d), the present simulation cannot reproduce the region where n_2/n_1 does not take a nonzero value until β increases beyond a certain value of β . The existence of such a region may be related to the observation that the coupling between two excitable oscillators is accomplished through a manifestation of coherence resonance. Unfortunately, such a coupling fashion is not considered in the present model.

In the sense that a monotonic increase of the firing number with β as observed in the present simulation has also been observed in other experiments and numerical simulations [9,12,13], the present experimental system may be rather peculiar. Sensitive responses of BZ reaction media to an electric field are mainly related to a field-induced transport of the inhibitor Br^- [20,21]. A strong noise electric field with $\beta > \beta_{\text{opt}}$ forces a rapid change in the transport direction of Br^- . However, Br^- cannot diffuse in response to such a rapid change because of the finite diffusion coefficient of Br^- . As a result, a field-induced redistribution of Br^- would likely be incomplete. This would lead to a net accumulation of the inhibitor around OSC1, and consequently firing of OSC1 would be inhibited. This may account for the decrease in the firing number with increasing β above β_{opt} .

IV. CONCLUSION

We have experimentally and numerically investigated the response of two coupled excitable BZ oscillators to an exter-

nal electric noise. Various modes of phase locking were induced by controlling both the noise amplitude and the separation between the two oscillators. The most striking characteristic of the present coupled system is that phase locking in the weak coupling regime occurs through coherence resonance. This fashion is entirely different from that in coupled self-sustained oscillators. Furthermore, two-mode dynamics with incommensurate periods was observed, in addition to oscillatory modes with simple period ratios. The experimental results were approximately reproduced, using a modified Oregonator model with a noise term. Noise effects, such as inhibition of firing for large β and entrainment governed by CR, may be characteristic of the present coupled

systems with diffusion coupling. As diffusion coupling is not so peculiar in excitable biological systems, however, an understanding of those noise effects appears to be necessary for a better understanding of noise-enhanced intercellular communication.

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