Noise-induced spatiotemporal dynamics in a linear array of excitable chemical oscillators

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The effects of additive noise on spatiotemporal dynamics are investigated in a one-dimensional array of excitable elements in which the Belousov-Zhabotinsky reaction is localized. At the appropriate separation between adjacent elements, we find that the resonance effect becomes larger for the element being more apart from the first element when only the first element is subjected to the external noise. This phenomenon is a sort of array-enhanced resonance. Furthermore, we find that phase locking between the first element and the other elements is induced via coherence resonance of the first element. Experimental observations are approximately reproduced in a numerical simulation with a forced Oregonator reaction-diffusion model.

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I. INTRODUCTION

Nontrivial effects of noise in nonlinear systems have recently attracted much attention. Especially, stochastic resonance (SR) is a well-known phenomenon, which is characterized as the enhancement of the response to a weak periodic input signal [1–5]. Even in the system without external periodic forcing, noise can also be helpful in inducing the resonance-like phenomenon of the coherent motion, called coherence resonance (CR) [6], autonomous SR [7,8], or internal signal SR [9]. This has been experimentally confirmed in various systems [10–12]. In the spatially extended system, such a noise-induced effect manifests itself as phenomena of noise-supported traveling waves [13] and noise-sustained pulsating patterns [14]. In ecological systems, furthermore, a nonmonotonic behavior of the pattern formation was induced by a multiplicative noise [15]. A more recent interest has been shifted to spatiotemporal dynamics in an array of similar stochastic elements. Coupling of SR or CR oscillators further enhances the ordering properties of noise compared with that of a single stochastic resonator. These behaviors are called array-enhanced stochastic resonance (AESR) [16–19] and array-enhanced coherence resonance [20]. Furthermore, the signal-to-noise ratio of the output signal can be significantly improved when stochastic bistable elements are coupled and a periodic signal is injected into the first element. This behavior, like AESR, is called noise-enhanced propagation (NEP) [21–24].

In a coupled array of stochastic elements, phase synchronization phenomena can also occur [25–27]. Phase synchronization and resonances significantly influence each other. A recent study has demonstrated that the interplay between coupling and noise plays an important role in enhancing the resonant response of a coupled array, using a coupled excitable FitzHugh-Nagumo model [20,28]. On account of such an interplay, conversely, it is not clear to what extent the coupling effects authentically contribute to array-enhanced resonances.

In this paper, we experimentally and numerically address these problems in a one-dimensional array of excitable elements in which the Belousov-Zhabotinsky (BZ) reaction is localized. On the basis of the idea of NEP, we construct a coupled array in such a way that external noise is injected into only the first element in the absence of an external periodic signal, in order to clarify the authentic effect of coupling. The coupling in this system is accomplished via a mass diffusion, so that a coupling strength can be controlled by varying the separation between excitable elements. We find various modes of phase locking depending on a noise level and the separation. Furthermore, we find that, at the appropriate separation, the more the elements are apart from the first element, the larger their resonance effects become. A reaction-diffusion model, which takes into account the effect of noise, is able to reproduce qualitatively the observed behaviors.

II. EXPERIMENT

Experiments were carried out with a localized reaction system in which the tris-(2,2′-bipyridine) ruthenium (II) complex $[Ru(bpy)_3^{2+}]$ was immobilized in cation exchange beads of a submillimeter size [29,30]. The initial composition of the BZ reaction solution was $[NaBrO_3]=0.45M$, $[NaBr]=0.05M$, $[CH_2(COOH)_2]=0.3M$, and $[H_2SO_4]=0.4M$. Reagent grade chemicals were used without further purification. The cation exchange beads (DOWEX, 50W-4X) of 500 μm in diameter were loaded with a solution of $[Ru(bpy)_3^{2+}]=1.2\times10^{-5}$ mol/g beads. At this composition, the system was initially in an oscillatory regime. The excitable steady state was realized by taking advantage of a high photosensitivity of $Ru(bpy)_3^{2+}$ [31]. The period of oscillation increased monotonically with increasing illuminated light intensity I, and finally the excitable steady state appeared for the light intensity of more than about $I_c=9$ mW. Then the period under the dark was estimated by extrapolation to be approximately $T_c=50$ s. In the experiment, we fixed I at 10 mW. The temperature of the BZ solution was maintained at 24 ± 0.5 °C. 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. Electrodes of 0.6 mm in diameter were placed 2 mm apart and parallel to the surface of the reaction medium. In order not to exert the direct effect of the electric

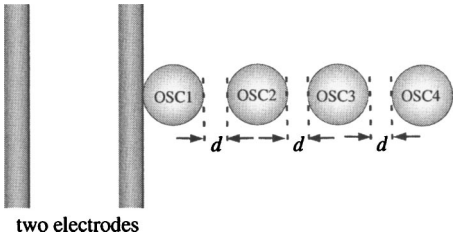

FIG. 1. Schematic representaion of the experimental setup. OSC1 contacts one of two electrodes. The linear array of beads immobilizing $Ru(bpy)_3^{2+}$ is placed perpendicular to the two electrodes.

field on other beads except one bead, we placed one excitable bead in contact with one of the two electrodes and three other beads were placed each d apart, as shown in Fig. 1. Here the bead placed in contact with the electrodes was tentatively termed the oscillator 1 (OSC1), and three other beads were termed the oscillator 2 (OSC2), the oscillator 3 (OSC3), and the oscillator 4 (OSC4) in that order. In this experimental setup, an external electric noise is injected into only OSC1, while the ith oscillator except OSC1 is mainly stimulated by the firing of the $(i-1)$th oscillator. In fact, waves were always initiated at OSC1, and no waves were excited on three other oscillators in the absence of OSC1. In this sense, OSC1 plays the role of a master oscillator in the behavior of entrainment. 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 of a noise pulse. In the measurement, we chose the duration time of $\delta=0.225$ s.

Coupling behaviors can be controlled by varying d, so that d acts as a control parameter governing the onset of oscillations and the period in excitable oscillators, in addition to β. When $d>140$ μm, four oscillators were always in an uncoupled state for any value of β. At a stronger coupling regime of $d\leqslant 50$ μm, on the contrary, four oscillators behave like a single oscillator for any β. At an intermediate value of d, the coherence of oscillations induced on four oscillators differed from one another, depending on d. To characterize the degree of coherence of these oscillations, we used the coherence measure R_c defined by

$$R_c = \frac{\langle \tau \rangle}{\sqrt{\langle \tau^2 \rangle - \langle \tau \rangle^2}}, \tag{1}$$

where $\langle \tau \rangle = (1/N)\Sigma_{i=1}^{N} \tau_i$, N is the number of firings, and τ_i is the time interval between the ith and $(i+1)$th firing events.

At the region of $d \geqq 140$ μm, four oscillators were completely uncoupled for any value of the noise amplitude β. Figure 2 shows the coherence measure R_c depending on d. At $d=140$ μm, only OSC1 becomes oscillatory above a certain value of the noise amplitude β, while three other oscillators are still in the excitable state. Thus four oscillators behave like independent oscillators. It should be noted that R_c of OSC1 passes through a maximum with increasing β. This suggests that the present oscillators have an ability to cause a coherence resonance. The oscillation having maximum R_c is almost periodic, and its value approximately coincides with

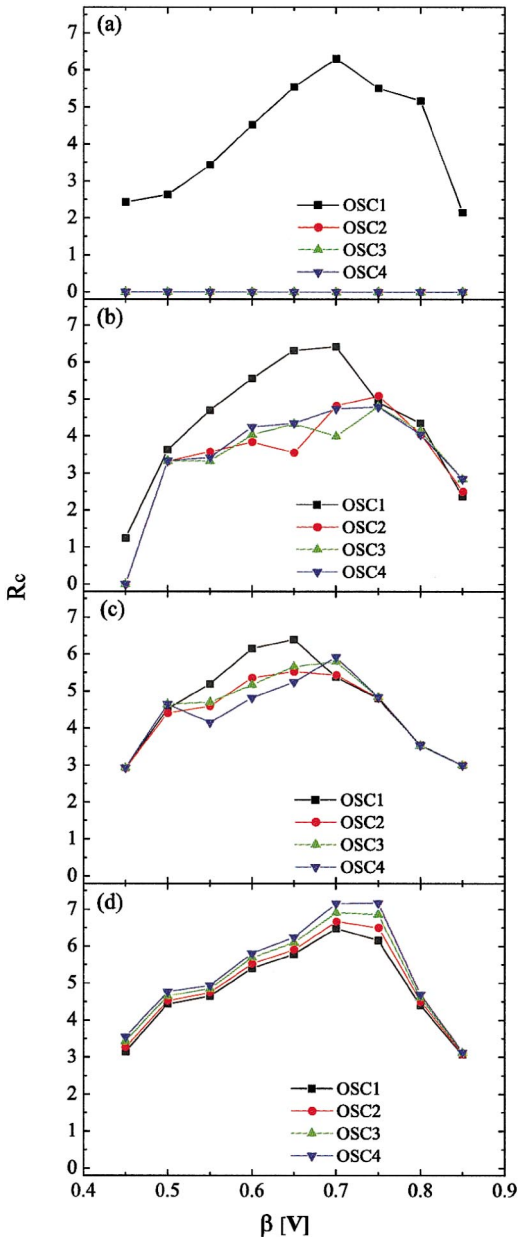

FIG. 2. (Color) Coherence measure R_c of four oscillators as a function of the noise amplitude β with a variation of the separation d: (a) $d=140$ μm, (b) $d=120$ μm, (c) $d=100$ μm, and (d) $d=80$ μm.

the period under the dark T_c, which is considered to be that of the intrinsic oscillation. This suggests that the coherent oscillation induced by noise is determined by a time scale of the intrinsic dynamics of the system. At the spacing of $d=120$ μm, all the oscillators exhibited firings. The maximum in R_c is clearly observable for all oscillators near $\beta=0.70$ V, as shown in Fig. 2(b), thus evidencing that there always exists the optimal noise amplitude β_{opt}, inducing the most regular oscillation. The R_c value of OSC1 is still larger than those of three other oscillators for all values of β. When $d=100$ μm, four oscillators showed almost the same β dependence of R_c. At the spacing of $60\leqslant d\leqslant 80$ μm, the degree of coherence for OSC4, OSC3, OSC2, and OSC1 be-

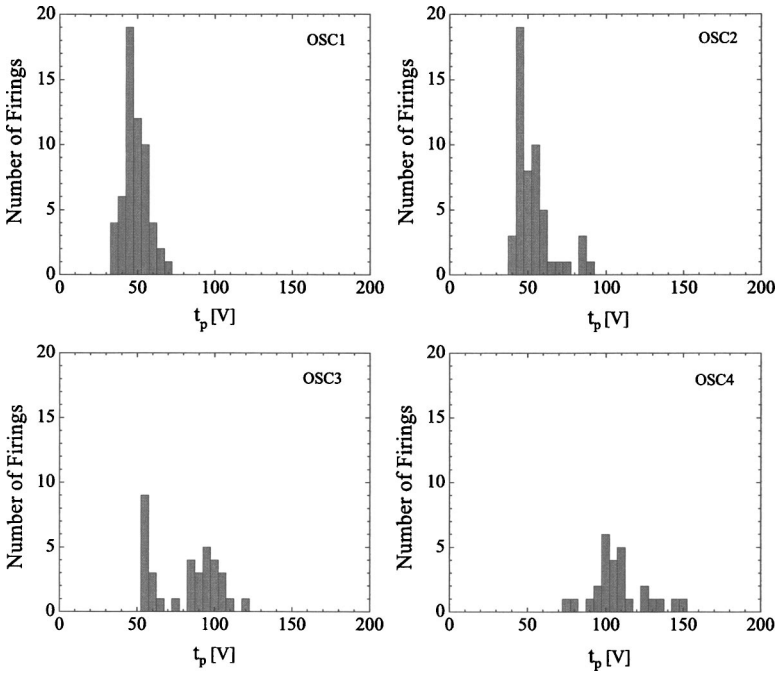

FIG. 3. Interspike interval histograms for four oscillators at the spacing $d=120$ μm and the noise amplitude $\beta=0.70$ V, where t_p denotes the time interval between successive firing events.

came larger in that order, as shown in Fig. 2(d). This indicates that, as the firing event on OSC1 is transferred to OSC2, OSC3, and OSC4, the coherence of the firing process is enhanced. The oscillation of OSC1 induced by an external noise is more or less irregular, so that OSC2 is stimulateded more or less irregularly. However, OSC2 shows a more periodic response to it through the assistance of nonlinearity of the chemical reaction. In the same way, the regularization of the period sequentially occurs with the propagation of stimulation from OSC2 to OSC3, and from OSC3 to OSC4. It should be emphasized that, although firings on the first element are purely induced by noise, the coherence of firing is enhanced by the cooperation between coupling and nonlinearity of the chemical reaction. We think that the observed array-enhanced phenomenon is due to an authentic effect of coupling. Although this phenomenon is apparently similar to noise-enhanced propagation, both greatly differ from each other in application of external forcing. Hence the observed phenomenon may be termed *array-enhanced propagation*. With further decreasing d, four oscillators behaved like a single oscillator for any β.

We examined the behaviors of noise-induced oscillations in terms of phase locking. For this purpose, it is necessary for the corresponding oscillations to be regular. As seen from Fig. 2, values of β_{opt} for four oscillators are almost the same. Hence we investigated the phase locking between OSC1 and one of three other oscillators at $\beta=\beta_{\mathrm{opt}}$, with varying d. In the strong-coupling regime, a 1:1 phase locking always occurred over the whole oscillator. In the weak-coupling regime, in contrast, various phase-locking modes appeared depending on d. Figure 3 shows the interspike interval histograms (ISIH) at $d=120$ μm. Going away from OSC1, the averaged period gradually shifts from $T_1\simeq 49$ s of OSC1 to $T_4\simeq 102$ s of OCS4. In this connection, the total number of firings of OSC4 reduces to almost one-half of that of OSC1. From the period ratio T_1/T_4 being almost $1/2$, we see that the phase locking of 1:2 occurs between OSC1 and OSC4. Furthermore, a two-mode behavior appears on OSC3 interposed between these oscillators, as seen from two peaks in ISIH in which incommensurate periods are clearly distinguishable [Fig. 3(c)]. From the viewpoint of phase locking, such a two-mode behavior corresponds to a nonresonant state which is regarded as a precursor of the transition between 1:1 and 1:2 modes. Thus a phase locking between largely separated oscillators not interacting directly with each other is achieved via irregular oscillations of oscillators interposed between them, in contrast to a phase locking due to a direct interaction as in the usual coupled system.

Let us now focus on the phase locking due to an indirect interaction, such as that between OSC1 and OSC4, for a fixed noise intensity β. In Fig. 4, the evolution of the ISIHs for both oscillators is shown for a variation of d at $\beta=\beta_{\mathrm{opt}}$. When d is decreased from 120 to 80 μm, the peak position of OSC1 remains unchanged, while the peak of OSC4 shifts to the position of approximately one-half of the initial value. With a shift of the peak, the profile of ISIH becomes sharper than that of OSC1, which is predictable from the corresponding coherence measure [Fig. 2(d)]. Thus changing d can cause a transition between 1:2 and 1:1 modes even if an interaction is indirect. This feature is similar to that of phase locking due to a direct interaction [27]. At an intermediate value of d, a two-mode behavior appears on OSC4. Such a mode also appears on OSC3 when $d=120$ μm, as shown in Fig. 3(c). This may indicate that, in a linear array of oscillators, a sequence of mode changes as shown in Fig. 4 is realized on the constituent oscillators when d is an appropriate value. This may be one of the features of phase locking in a linear array of oscillators. It should be noted that phase locking due to an indirect interaction is governed by CR on the master oscillator initiating oscillations, i.e., OSC1, although the features of phase-locking modes are apparently similar to those in directly coupled deterministic oscillators.

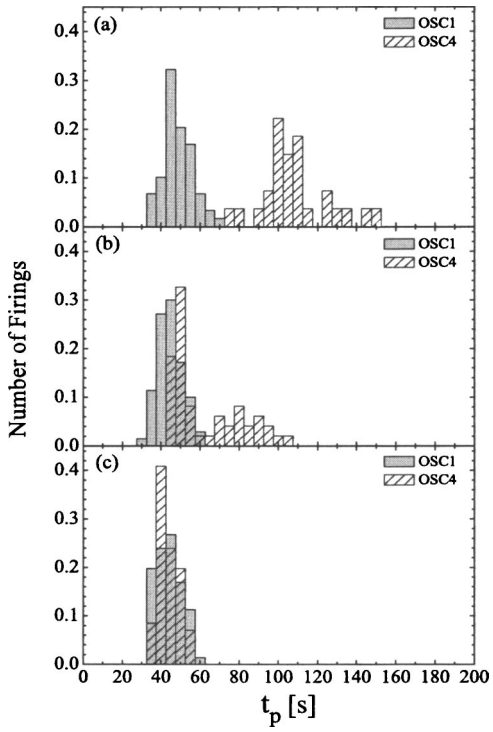

FIG. 4. Interspike interval histograms as a function of the separation d at the noise amplitude $\beta=0.70$ V: (a) $d=120$ μm, (b) $d=100$ μm, and (c) $d=80$ μm, where t_p denotes the time interval between successive firing events and histograms are normalized by the total number of firings.

III. REACTION-DIFFUSION MODEL

We approximate the present system as a one-dimensional system consisting of interacting four excitable elements separated by an inactive area, i.e., a diffusion area. We employ the three-variable Oregonator model modified to take into account the effects of an external electric noise. An electric field induces additional drifts of reacting ionic species. In reaction-diffusion models, the terms describing those additional drifts appear coupled to a gradient term [32]. However, this is not our case, because the electric field is not applied across the excitable areas under consideration, as seen from the experimental setup in Fig. 1. Such a field-induced drift of ions seems not to arise on those areas, although it can occur in the diffusion area between two electrodes. Of three fundamental species in the Oregonator model, only the inhibitor Br^- is influenced by the electric field, because the activator $HBrO_2$ is not charged and the catalyst $Ru(bpy)_3^{2+}$ is immobilized in the beads. Now we assume the following reaction-diffusion process, taking into account that the waves are initiated at the point of contact with the electrode. The waves are first excited on OSC1 by a field-induced change in the concentration of Br^- around the electrode, and stimulate OSC2, traveling across the inactive area. Subsequently, the waves travel to OSC3 and OSC4 in a similar manner. Under this assumption, we express the noise term in the form of not a gradient term but an additive term. Then the model equations are given by

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

$$\frac{\partial v}{\partial t} = \gamma(u - v), \tag{3}$$

$$\frac{\partial w}{\partial t} = \frac{\gamma}{\epsilon'}[fv - w(u+q) + \phi] + \beta\xi_\delta + D_w \frac{\partial^2 w}{\partial x^2}, \tag{4}$$

where the variables u, v, and w describe the concentrations of $HBrO_2$, the $Ru(bpy)_3^{2+}$ catalyst, and Br^-, respectively. D_u and D_w denote the diffusion coefficients of $HBrO_2$ and Br^-, respectively. ϵ, ϵ', f, q, and γ are control parameters. ϕ is the light flux, and $\beta\xi_\delta$ is the electric noise with the noise amplitude β and Gaussian white noise ξ_δ with the duration time δ. Here the noise term is considered only at the end of OSC1, namely, the point of contact with the electrode. Then the external control parameters are ϕ and β.

We assume that, in four excitable areas, both reaction and diffusion are allowed, i.e., $\gamma=1$, and in the diffusion areas separating excitable elements only the diffusion term remains, i.e., $\gamma=0$. 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 $D_w=1.12$. With increasing ϕ, the period of oscillation increased monotonously, 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.

In the computations, the whole interval was divided into equally spaced grids. When the diffusion area is located between the grid points n_i and n_{i+1} with an odd number i ($\leqslant 5$), the separation l between neighboring elements can be estimated by $l=n_{i+1}-n_i-1$. 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 zero flux, while that between excitable and diffusion areas was taken to be free. The noise is subjected to only OSC1 in the same manner as in the experimental setup. It is assumed that OSC2, OSC3, and OSC4 are stimulated only through firings of the preceding oscillators. The computation was performed by the fourth-order Runge-Kutta method with a grid spacing $\Delta x=0.05$, time steps $\Delta t=0.0001$, and the duration time $\delta=50\Delta t$. When $l\geqslant 51$, only OSC1 became oscillatory above a certain value of the noise amplitude β, while three other oscillators were still in the excitable state. That is, four oscillators were always uncoupled for any value of β. When the system was brought into the weak-coupling regime by decreasing l, all oscillators became oscillatory. We used the coherence measure R_c to characterize the degree of coherence of the induced oscillations. For every oscillator, R_c passes through a maximum at the some optimal noise amplitude β_{opt} with an increase in β. Thus the present model has an ability to cause a coherence resonance. When the system was brought in the strong-coupling regime, R_c became larger on the order of OSC4, OSC3, OSC2, and OSC1, as shown in Fig. 5. The values of β_{opt} for four oscillators are almost coincident. These results are consistent with the experimental observations. We checked the occurrence of phase locking between OCS1 and one of three other oscillators at fixed β. We now focus on phase locking between the most largely

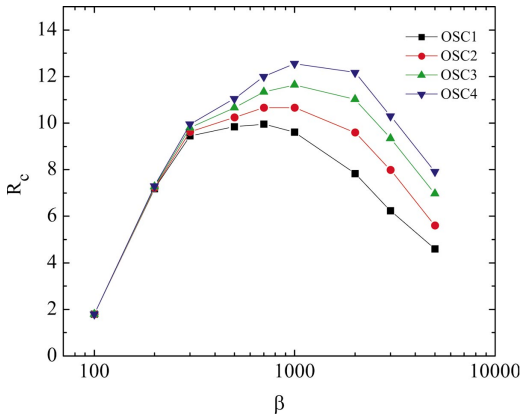

FIG. 5. (Color) Coherence measure for four oscillators as a function of the noise amplitude β at $d=4$.

separated oscillators. Figure 6 shows the evolution of the ISIHs of both OSC1 and OSC4 at $\beta=\beta_{\mathrm{opt}}$. With a decrease in l, it is clearly seen how the peak periods of two oscillators approach each other and become coincident at some value of l. Thus the transition from 1:2 to 1:1 phase locking is controllable with l. In the transition process, a two-mode behavior, as seen from two peaks with incommensurate periods, appears on OSC4 as a precursor to stable phase locking. Such a sequence of transitions is consistent with the experimental observations.

IV. CONCLUSION

We have experimentally and numerically investigated the spatiotemporal dynamics induced in a linear array of four excitable elements in which the Belousov-Zhabotinsky (BZ) reaction is localized, where only the first element is subjected to external noise. The most striking result is that the resonance effect becomes larger for the element being more apart from the first element at the appropriate separation between adjacent elements. We believe that the cooperation between coupling and nonlinearity of the chemical reaction plays the essential role for the occurrence of this phenomenon, termed array-enhanced propagation. Thus we can say that a linear array of excitable elements has the ability to clean up a noisy oscillation. It is of interest to clarify to what extent such resonance effects are enhanced with a further increase in the number of elements. Further inspection in this connection is in progress. Furthermore, we have examined

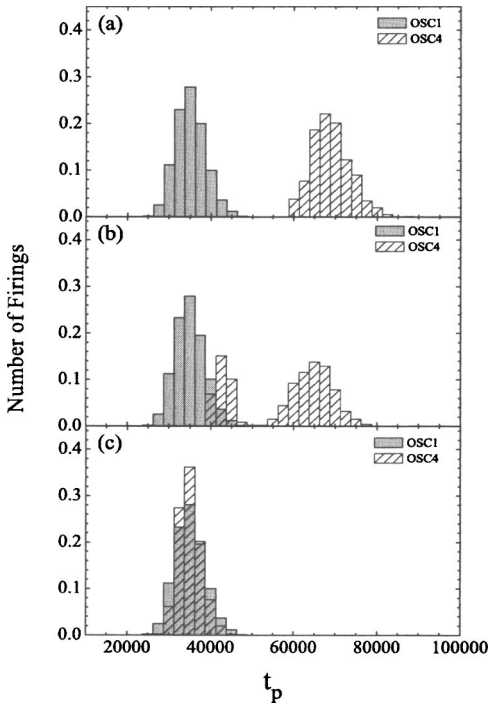

FIG. 6. Interspike interval histograms as a function of the separation l at the noise amplitude $\beta=1000$: (a) $l=40$, (b) $l=26$, and (c) $l=4$, where histograms are normalized by the total number of firings.

phase locking between the first element and one of three other elements by controlling both the noise amplitude and the separation between elements. The coupling fashion characteristic of the present system is that phase locking between largely separated elements occurs via irregular oscillations of elements interposed between them, which is governed by coherence resonance of the first element initiating firings. This fashion is entirely different from that of coupled deterministic oscillators. The experimental results have been approximately reproduced, using a modified Oregonator model with a noise term.

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