Feedback-controlled dynamics in a two-dimensional array of active elements

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We investigate collective behaviors in a two-dimensional array of active elements controlled by time-delayed feedback, where elements are prepared by localizing the Belousov-Zhabotinsky reaction in a gel matrix. We demonstrate that both the spatial and temporal coherence can be effectively controlled by varying feedback parameters, such as the time delay and the gain. For a sufficiently high feedback gain, the fully synchronized state with low temporal coherence appears, which might be the state induced only by the delay feedback. Experimental results are approximately reproduced in a numerical simulation with a forced Oregonator reaction-diffusion model.

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I. INTRODUCTION

When many elements capable of autonomous oscillation are coupled, cooperative interactions among them reveal complex but ordered collective dynamic behaviors as basic manifestations of self-organization. The most significant of these patterns are phase synchronization and clustering that are frequently observed in physical [1], chemical [2–4], and biological systems [5,6]. The coherence of spatiotemporal dynamics can be controlled by external forcing, especially, efficiently by feedback utilizing the inherent sensitivity to external stimulations. Feedback techniques allow each active element to interact with any other element in the array. This offers possible approaches for designing desired interactions among elements. In addition, variation of the feedback gain and of the delay time can result in a qualitative change in spatiotemporal patterns [7–12].

Photosensitive Belousov-Zhabotinsky (BZ) reaction has offered one of the best experimental systems to investigate effects of feedback, in which the feedback can be achieved by monitoring the local excitability of reaction media, processing its image by computer, and illuminating the signal on reaction media. In spatially extended excitable media, a spiral wave and generic patterns have been effectively manipulated using a real-time global feedback [13–16]. Similarly, collective dynamics in discrete BZ systems, such as synchronization and clustering, have also been effectively controlled [17–19]. However, much less is known about the effect of a time-delayed feedback on discrete oscillatory systems.

In this paper we experimentally investigate the effect of time-delayed feedback on spatiotemporal dynamics in a two-dimensional array of autonomous active elements in which the BZ reaction is localized. We find that, when the delay time is optimal, phase synchronization is induced at the feedback gain more than a threshold value. We reveal that the spatiotemporal coherence of oscillations can be effectively controlled by varying feedback parameters, such as the time delay and the gain. The observed behaviors are numerically reproduced, using an Oregonator model which takes into account the effect of feedback.

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II. EXPERIMENT

The discrete BZ reaction system was constructed using photolithography-assisted techniques [20,21]. Here the reactor was made from the elastomeric material poly(dimethylsiloxane). This methodology made it possible to freely control features of the discrete reaction system, such as the size of reactor units, the spacing between neighboring elements, and the number of elements. In the experiment, reactor units of about 430 μm in diameter and 65 μm in depth were arranged in the lattice with the spacing of 100 μm (Fig. 1), in which silica-gel matrices were prepared by acidifying the solution of 125 μl of 20 wt % Na_2SiO_3, 100 μl of 20 mM $Ru(bpy)_3SO_4$, and 100 μl of 10 M H_2SO_4. The light sensitive catalyst, tris-(2,2′-bipyridine) ruthenium (II) complex [$Ru(bpy)_3^{2+}$], was immobilized in silica-gel matrices. The reactor was placed into a chamber that was continuously fed with fresh, catalyst-free BZ solution at a pumping rate of 6 ml/h to maintain constant, nonequilibrium conditions. The initial composition of the catalyst-free BZ solution was $[NaBrO_3]=0.4$ M, $[NaBr]=0.125$ M, $[CH_2(COOH)_2]=0.4$ M, and $[H_2SO_4]=0.5$ M. Reagent grade chemicals were used without further purification. At this composition, the system was in an oscillatory regime with the period $T_p=35\pm5$ s. Here the period distribution results from some scatter in the size of reactor units and temperature fluctuations. Looking on a catalyst-doped microgel element in a reactor unit as an oscillator, a two-dimensional array of these

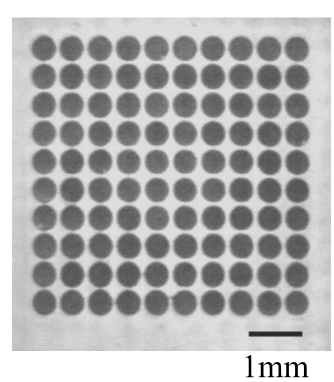

FIG. 1. Snapshot of the 10×10 microgel array with the spacing of 100 μm.

active elements can serve as a model of a network of cardiac cells in living organisms. The temperature of the BZ solution was maintained at 24 ± 0.5 °C. A computer-controlled video projector was used to illuminate the sample from below through a 460 nm bandpass filter. The color changes due to the redox reaction were detected in transmitted light by a charge-coupled device camera and transformed into the change in light intensity by the imaging system.

We controlled spatiotemporal behaviors in a 10×10 lattice of oscillators by the time-delayed feedback. The illumination intensity of the feedback control is expressed as

$$I(t) = I_0 + k[B(t-\tau) - B_0], \tag{1}$$

where I_0, k, and τ are the background intensity, the feedback gain, and the delay time, respectively. We chose a positive value for k. These are significant parameters to control a spatiotemporal coherence of the system. The intensity $B(t)$ is determined as the average of the light intensity over all oscillators in a $N\times N$ lattice:

$$B(t) = \frac{1}{N^2}\sum_{i=1}^{N}\sum_{j=1}^{N} I_{ij}(t), \tag{2}$$

where $I_{ij}(t)$ is the light intensity of the oscillator (i,j) on an eight-bit gray scale. The constant B_0 is the value of $B(t)$ at the time when all reactor units are in the reduction state, which was determined at the beginning of each experiment. In the second term of Eq. (1), therefore, the difference $B(t-\tau)-B_0$ is always positive, representing the proportion of oscillators in excited states. Thus the feedback force with positive k tends to suppress the excitability of the system. The intensity of feedback illumination was updated at the interval of 1 s and interrupted during 0.1 s every 1 s in order to capture the image of the state of the system.

In the absence of feedback, the coupling strength can be controlled by the spacing between nearest neighbors, because the coupling between oscillators is accomplished via a mass diffusion. For a large spacing, all oscillators behave such as independent oscillators, while for a small spacing they always do like a single oscillator. In the presence of feedback, the interplay between feedback force and interactions among oscillators is crucial for collective behaviors. Therefore, we chose the spacing of about 100 μm, corresponding to the intermediate coupling strength [21]. Figure 2 shows the spatiotemporal pattern of the light intensity from oscillators $(i,5)$ $(i=1\sim10)$ under the feedback control. The phases of oscillators are randomly distributed before application of the feedback. When $\tau=1$ s, the feedback scarcely exerts its effect, that is, no synchronization occurs. When $\tau=10$ s, that is, τ gets closer to the refractory period of the reaction t_r, the system gradually evolves such that all oscillators are eventually entrained, and finally full synchronization takes place. Here t_r was determined as the ensemble average of the time taken for $I_{ij}(t)$ enhanced with activation to be reduced to the steady value in a firing event.

In order to characterize the temporal coherence of oscillators in a $N\times N$ lattice, we used the coherence measure R, defined as

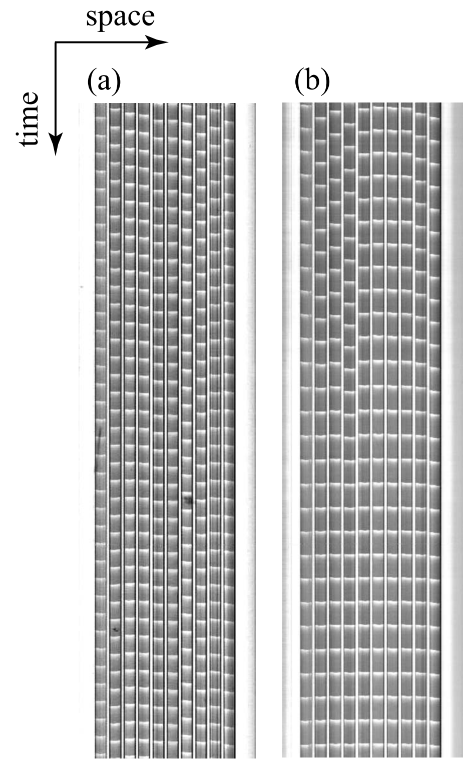

FIG. 2. Spatiotemporal pattern of the light intensity in the oscillator array $(i,5)$ $(i=1\sim10)$ controlled by the feedback with the gain $k=140$; (a) $\tau=1$ s; (b) $\tau=10$ s.

$$R = \frac{1}{N^2}\sum_{i=1}^{N}\sum_{j=1}^{N}\frac{\langle T_{ij}\rangle}{\sqrt{\langle T_{ij}^2\rangle - \langle T_{ij}\rangle^2}}, \tag{3}$$

where $\langle T_{ij}^m\rangle=(1/n_f)\sum_{k=1}^{n_f}(T_k^{ij})^m$, n_f is the number of firings, and T_k^{ij} is the time interval between the kth and $(k+1)$th firing events in the oscillator (i,j). Figure 3(a) shows the dependence of R on τ at $k=140$. One can see that R periodically varies with τ. Such an oscillatory behavior of R is consistent with the theoretical prediction [9]. Peaks appear at $\tau_1\approx10$ s, $\tau_2\approx45$ s, and $\tau_3\approx80$ s. Here it should be noted that the value of τ_1 is close to the refractory period t_r. As the time difference $\tau_{n+1}-\tau_n$ $(n=1,2)$ is almost equal to the mean period of oscillation T_p, an optimal delay time $\tau_{\rm opt}$ at which R is maximized is given by $\tau_{\rm opt}\approx t_r+(n-1)T_p$ with n being integer.

The feedback force influences not only the temporal coherence but also the spatial coherence of oscillators. To characterize the synchronization behavior between oscillators in the array, we introduce a phase of oscillator [22],

$$\phi_{ij}(t) = 2\pi\frac{t-\tau_k^{ij}}{\tau_{k+1}^{ij}-\tau_k^{ij}}, \quad \tau_k^{ij}\le t\le\tau_{k+1}^{ij}, \tag{4}$$

where τ_k^{ij} is the time of the kth firing of the oscillator (i,j). The phase difference between oscillators (i,j) and (l,m) is defined as $\Phi_{ij,lm}(t)=\phi_{ij}(t)-\phi_{lm}(t)$. As the measure of synchronization between oscillators (i,j) and (l,m), we use an index expressed as $\gamma_{ij,lm}^2=\langle\cos\Phi_{ij,lm}\rangle^2+\langle\sin\Phi_{ij,lm}\rangle^2$, where the brackets denote the average over time [23]. Then the

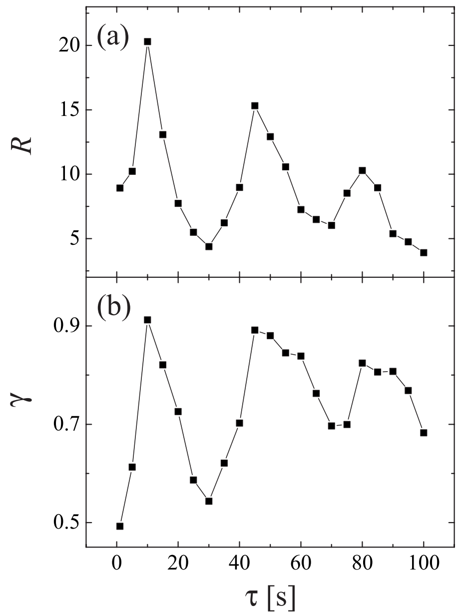

FIG. 3. (a) Temporal coherence measure and (b) phase synchronization measure as a function of τ at the feedback gain $k=140$.

degree of phase coherence in a $N \times N$ lattice can be characterized by calculating the spatial average,

$$\gamma = \frac{1}{n_p} \sum_{l=1}^{N} \sum_{m=1}^{N} \sum_{\langle ij,lm \rangle} \gamma_{ij,lm}, \tag{5}$$

where n_p is the number of coupling pairs. In particular, $\gamma=1$ if all oscillators synchronize globally, whereas $\gamma=0$ if the oscillators are uncoupled. Here the sum is over nearest neighbor sites on the lattice. The dependence of γ on τ is shown in Fig. 3(b). One can see that γ periodically changes with τ, which is entirely synchronized with R.

In order to furthermore clarify the effect of feedback, we investigated the dependence of the spatiotemporal coherence on the feedback gain k for various values of τ, as shown in Fig. 4. When τ is quite small compared to the period of oscillation, such as $\tau=1$ s, R monotonically decreases with k, while γ remains almost constant. The similar behavior is observed when $\tau=30$ s. Thus, when the feedback has the delay time independent of the characteristic time of the BZ reaction, it does not affect collective behaviors. In contrast, when τ is close to the refractory period, i.e., $\tau=10$ s, γ abruptly increases to 0.9 at k beyond about $k_c=140$, indicating that almost full synchronization occurs. This phenomenon might be regarded as a forced entrainment of 1:1 by the time-delayed feedback. This is reminiscent of classical synchronization phenomena. On the other hand, R is kept approximately constant below k_c, but it tends to decrease above k_c.

Figures 4(c) and 4(d) show a typical time series of light intensity (gray level) of the oscillator for small and large values of k. We see that firing events become more irregular with increasing k, even if τ is close to the inherent refractory period. Let us consider why such a temporal disorder grows. An increase in k monotonously increases the mean period of

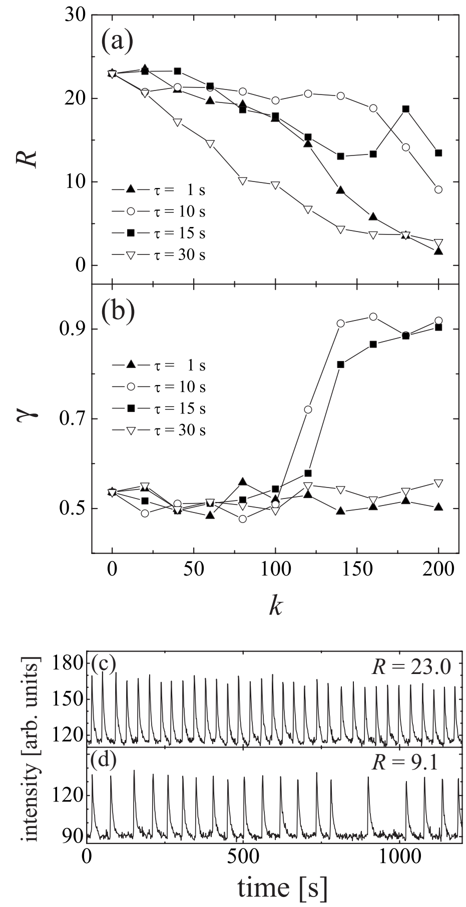

FIG. 4. (a) Temporal coherence measure and (b) phase synchronization measure as a function of the feedback gain k for various values of the delay time. (c) and (d) Time series of the gray level of the oscillator (2, 4) for $\tau=10$ s at $k=0$ and $k=200$, respectively.

oscillation T_p for every τ, as shown in Fig. 5. These behaviors result from the characteristic that feedback force tends to inhibit activity of the oscillator. The increase in T_p results in an effective increase in the refractory period of the reaction. As long as the lengthened refractory period is still close to an optimal delay time τ, the temporal coherence of oscillation remains high. Since an increase in k simultaneously strengthens a global coupling among oscillators, full synchronization

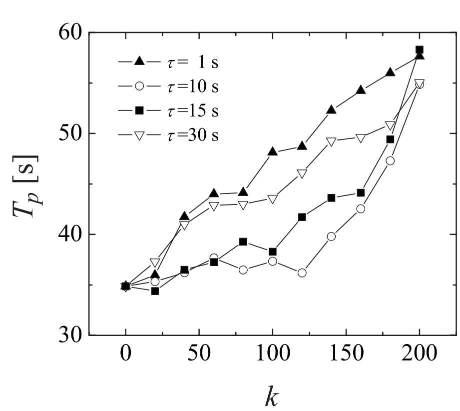

FIG. 5. Mean period of oscillation as a function of the feedback gain for various values of τ.

can occur as shown in Fig. 2(b). However, when the deviation of the refractory period from τ_{opt} gets pronounced by a further increase in k, R cannot hold a high value, as shown in Fig. 3(a). Then a fully synchronized state with low temporal coherence appears. This state might be an additional one induced only by delay feedback. According to this view, it follows that it is possible to enhance R even at large k, if the delay time larger than the inherent refractory period is used. The case of $\tau=15$ s is shown in Fig. 4. As expected, R is enhanced around $k=180$, although it monotonically decreases in the range of small k.

III. NUMERICAL SIMULATION

We modeled a two-dimensional array of limit cycle oscillators coupled to its nearest neighbors. We employed the three-variable Oregonator model modified to take into account the effects of feedback. In our experimental setup, the catalyst $\mathrm{Ru(bpy)}_3^{2+}$ is immobilized in the silica-gel matrix, so that its self-diffusion is negligible. The excitability of each oscillator is influenced by the feedback illumination, because the product of inhibitor Br^- is promoted due to the photochemical reaction of $\mathrm{Ru(bpy)}_3^{2+}$. Then the model equations are given by

$$\frac{du_{i,j}}{dt} = \frac{1}{\varepsilon}[u_{i,j} - u_{i,j}^2 - w_{i,j}(u_{i,j} - q_{i,j})] + K_u(u_{i+1,j} + u_{i-1,j} + u_{i,j+1} + u_{i,j-1} - 4u_{i,j}), \quad (6)$$

$$\frac{dv_{i,j}}{dt} = u_{i,j} - v_{i,j}, \quad (7)$$

$$\frac{dw_{i,j}}{dt} = \frac{1}{\varepsilon'}[fv_{i,j} - w_{i,j}(u_{i,j} + q_{i,j}) + \phi] + K_w(w_{i+1,j} + w_{i-1,j} + w_{i,j+1} + w_{i,j-1} - 4w_{i,j}), \quad (8)$$

where the variables $u_{i,j}$, $v_{i,j}$, and $w_{i,j}$ describe the concentrations of HBrO_2, the $\mathrm{Ru(bpy)}_3^{3+}$ catalyst, and Br^- in the oscillator (i,j), respectively. The coupling among oscillators is controlled by the coupling strength K_u and K_w $(=1.12K_u)$. ε, ε', and $q_{i,j}$ are scaling parameters and f is the stoichiometry parameter. The parameter ϕ represents the light-induced production of Br^-, expressed as

$$\phi = \phi_0 + k[V(t-\tau) - V_0], \quad (9)$$

where ϕ_0 corresponds to I_0 and k is the feedback gain. The intensity $V(t)$ is determined as the average of $v_{i,j}$ over all oscillators in a $N \times N$ lattice:

$$V(t) = \frac{1}{N^2}\sum_{i=1}^{N}\sum_{j=1}^{N} v_{i,j}(t). \quad (10)$$

The constant V_0 is the value of $V(t)$ at the time when all oscillators are in the steady state. These parameters were chosen such that the system was in the oscillatory regime: $\varepsilon=0.01$, $\varepsilon'=0.0001$, $f=1.4$, $K_u=0.1$, and $\phi_0=0$. The parameters $q_{i,j}$ were randomly chosen between 0.0125 and 0.0275 to provide the distribution of the period to the oscillator array. The computation was performed by the improved Euler method with time steps $\Delta t=0.0001$. The feedback force with the duration time $\delta=500\Delta t$ was globally applied to oscillators. The spatial separation of the oscillators was taken as $\Delta x=1$. The boundary conditions for both edge oscillators were taken to be zero flux. The mean period of oscillation T_p were distributed over the range of 1.8 ± 0.4, where the refractory period t_r was about 0.88. We used indices R and γ defined in Eqs. (3) and (5) to characterize the degree of temporal and spatial coherence, respectively.

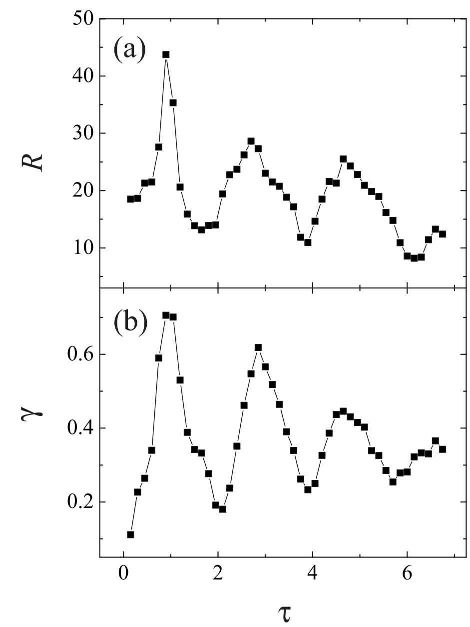

FIG. 6. (a) Temporal coherence measure and (b) phase synchronization measure as a function of τ at the feedback gain $\log_{10} k=-1.3$.

Figure 6 shows the τ dependence of R and γ at $\log_{10} k=-1.3$. We see that both change periodically and synchronously with increasing τ from $\tau=0.15$. The first maximum appears at the value of τ close to the refractory period t_r, and successive maxima appear at the interval of T_p. Thus the nth optimal delay time τ_{opt} maximizing the spatiotemporal coherence can be expressed as $\tau_{\mathrm{opt}} \approx t_r+(n-1)T_p$ $(n=1,2,\ldots)$ in a similar manner as the experimental results.

Note that the model, compared to the experiment, yields higher values of R and lower values of γ. In the experiment, the interspike interval of each oscillator fluctuates due to thermal noise. This spontaneous fluctuation makes oscillators in the intermediate coupling regime to occasionally synchronize, resulting in large values of γ to a certain extent. In the model, in contrast, the interspike interval of each oscillator is kept constant because of a limit cycle, although there exist a distribution of periods among the oscillators. Accordingly, the values of R lie much higher than in the case of the experiment. On the other hand, synchronization rarely occurs in the intermediate coupling regime, because the interspike interval of each oscillator does not fluctuate. This results in the low values of γ.

We investigated the dependence of the spatiotemporal coherence on k for various τ, as shown in Fig. 7. When τ is very small compared to the refractory period, i.e., $\tau=0.15$, R

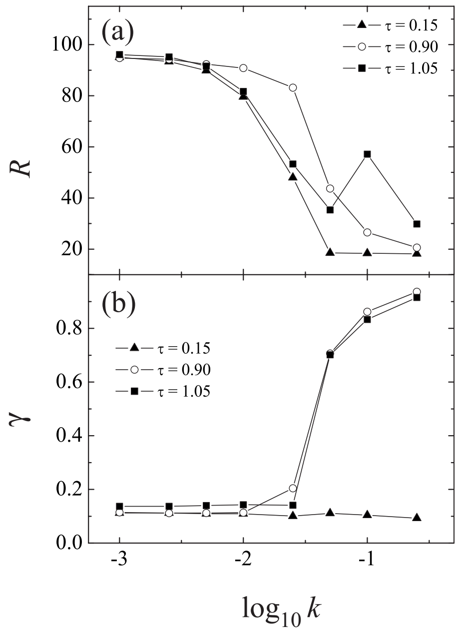

FIG. 7. (a) Temporal coherence measure and (b) phase synchronization measure as a function of the feedback gain k for various values of the delay time.

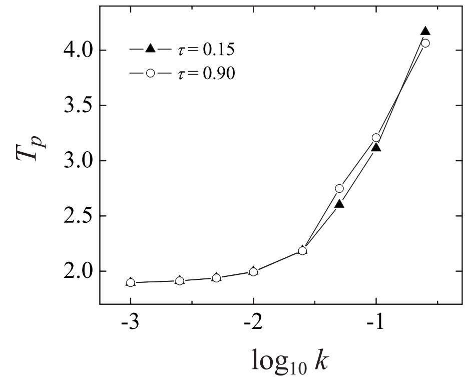

FIG. 8. Mean period of oscillation as a function of the feedback gain for various values of τ.

monotonously decreases with an increase in k, while γ remains unchanged. In contrast, when τ is close to the refractory period, i.e., τ=0.90, R is kept almost constant below $\log_{10} k_c$=−1.6, but it rapidly decreases above k_c. On the other hand, γ is rapidly enhanced when k is increased beyond k_c. These behaviors are in agreement with the experimental results.

An increase in k increases the mean period of oscillation T_p, as shown in Fig. 8, resulting in an effective increase in the refractory period. From the view that such a decrease in R originates from the large deviation of the enhanced refractory period from 0.88, we investigated the k dependence of R by setting τ to 1.05, i.e., the value larger than the inherent refractory period. As expected, R is enhanced around $\log_{10} k$=−1.0, as shown in Fig. 7. This is consistent with the experimental observation.

The periodic variation in spatiotemporal coherence can be explained according to Ref. [9]. For simplicity, we are concerned with two limit cycle oscillators with slightly different intrinsic periods, termed the oscillator 1 (OSC1) and the oscillator 2 (OSC2). Figure 9(a) shows time series of two oscillators at a certain time. In the absence of the feedback, two oscillators behave independently. In the presence of the feedback, the oscillator is forced to change the phase according to the state of the oscillator; the phase is not influenced when the state is in the refractory period, while a delay of phase occurs when the state is in the steady period, because the feedback illumination tends to inhibit activity of the oscillator. When $\tau < t_r$, the feedback events can scarcely overlap, as illustrated in Fig. 9(b). That is to say, feedback forces from both oscillators act independently. Such a feedback force can never exert a cooperative effect leading to synchronization. If $\tau \approx t_r$, the overlap of feedback events necessarily arises with the elapse of time, resulting in a matching of phases between oscillators [Fig. 9(c)].

When $\tau \gtrapprox t_r$, the situation described for $\tau \lessapprox t_r$ is repeated, because firing events are repeated on the average at T_p. Thus enhancement of coherence occurs when $\tau \approx t_r + (n-1)T_p$ $(n=1,2,\ldots)$.

In the above, we were concerned with synchronization between two oscillators. We now consider the synchronization process in a 10×10 array of oscillators with phases distributed randomly. Figure 10 illustrates the temporal evolution of the phase distribution. When $\tau < t_r$, the phase remains scattered around the circle irrespective of the time elapse. Here three clusters apparently arise, but they are quite unstable. When $\tau \approx t_r$, in contrast, phases approximately converge in a unique value at t=6. This state is robust, indicating the occurrence of full synchronization.

IV. CONCLUSION

We have experimentally and numerically investigated the effect of time-delayed feedback on both spatial and temporal

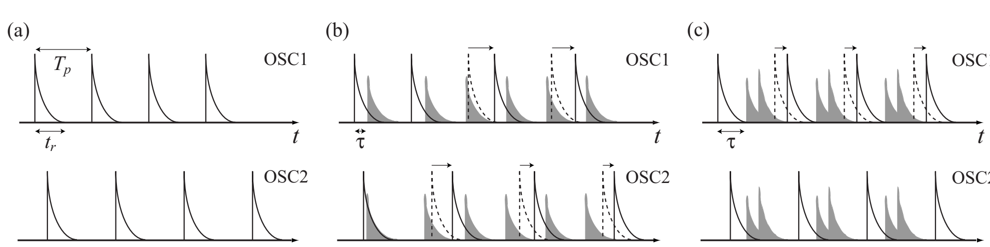

FIG. 9. Schematic time series of two oscillators, (a) in the absence of feedback force, and in the presence of feedback force with the delay time (b) $\tau < t_r$ and (c) $\tau \approx t_r$. Gray areas indicate profiles of feedback illumination. Arrows show delay of phase due to the feedback force

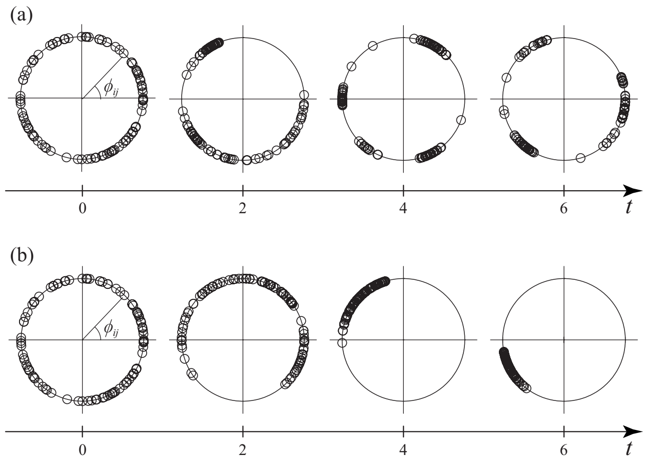

FIG. 10. Time evolution of the distribution of phases ϕ_{ij} under the time-delayed feedback with the gain $\log_{10} k=-0.60$, where (a) $\tau=0.15$ and (b) $\tau=0.90$.

coherence in a two-dimensional array of self-sustained oscillators based on the BZ reaction. We have demonstrated that it makes possible to freely increase or decrease the spatiotemporal coherence by adjusting the delay time. The feedback with an appropriate gain can maximize both spatial and temporal coherence when the delay time is close to the refractory period. However, too strong feedback decreases the temporal coherence, even if the delay time is optimal, although the fully synchronized state is maintained.

Coupling among oscillators in the present system is achieved by both mass diffusion and global feedback. Hence the interplay between both effects plays a significant role in organizing observed dynamics. When the spacing between oscillators is large to such an extent that a local coupling is negligible, only global coupling becomes feasible through feedback. Further inspection in this connection is in progress.

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