Array-enhanced coherence resonance and phase synchronization in a two-dimensional array of excitable chemical oscillators

Taiji Okano, Akane Kitagawa, and Kenji Miyakawa*
Department of Applied Physics, Fukuoka University, Fukuoka 814-0180, Japan
(Received 2 May 2007; published 1 October 2007)

We investigate the spatiotemporal dynamics in a two-dimensional array of excitable elements subjected to independent external noise, where elements are prepared by localizing the Belousov-Zhabotinsky reaction in a gel matrix. We experimentally demonstrate that the coherence of noise-induced firings is improved with increasing the array size, i.e., the occurrence of array-enhanced coherence resonance. Furthermore, it is found that synchronization among oscillators which are barely coupled can be achieved via coherence resonance. Experimental observations are approximately reproduced in a numerical simulation with a forced Oregonator reaction-diffusion model.

DOI: 10.1103/PhysRevE.76.046201 PACS number(s): 05.45.Xt, 05.40.−a, 82.40.Bj

I. INTRODUCTION

Noise-induced effects in nonlinear systems have recently attracted much attention. One of the most interesting effects is stochastic resonance (SR), which is characterized as an enhancement of a system response to a weak input signal due to a moderate noise intensity [1–5]. Another effect is the resonancelike phenomenon induced in the system without external periodic forcing, where the coherence of noise-induced firings is enhanced for an optimal noise intensity, called coherence resonance (CR) [6] autonomous SR [7,8], or internal signal SR [9]. The phenomenon of CR has been experimentally confirmed in various systems [10–12]. The frontier of interest in noise-induced effects has shifted to spatially extended systems, where some new spatiotemporal dynamics appears, such as the noise-supported wave propagation [13,14], the array-enhanced propagation [15], and noise-sustained pulsating pattern formation [16]. When SR or CR oscillators are coupled to homo-or heterogeneous arrays and subjected to independent local noise, nontrivial behaviors also appear, called array-enhanced stochastic resonance (AESR) [17–20] or array-enhanced coherence resonance (AECR) [21–23], where the effects of SR or CR are further optimized.

Phase synchronization can also occur in an array of stochastic elements. It is accompanied by enhancement of the coherence of the noise-induced firings [21,22,24–27]. This is the feature characteristic of coupled stochastic oscillators. Thus noise can play a positive role in not only enhancing a resonant behavior but also inducing synchronization, in cooperation with coupling. An interesting question about such an array of stochastic elements is to what extent noise-induced effects can be enhanced by coupling. Do noise and coupling always cooperate with each other in enhancing spatiotemporal coherence? Studies of array-enhanced effects made so far are mainly confined to locally and linearly coupled arrays.

In this paper, we experimentally address these problems using a two-dimensional array of excitable elements prepared by localizing the Belousov-Zhabotinsky (BZ) reaction in a gel matrix. Here each element is subjected to independent external noise. Such a two-dimensional discrete lattice may be suitable for description of biological extended systems, such as cardiac cells and receptors. In the present system the coupling strength can be controlled by varying the spacing between elements, because of the diffusively coupled array. We find that at a fixed coupling strength the coherence of firings induced by noise are enhanced with increasing the number of elements, indicating the occurrence of AECR. Synchronization of oscillators which are barely coupled can be accomplished via coherence resonance. The observed behaviors are numerically reproduced, using a Oregonator model which takes into account the effects of noise.

II. EXPERIMENT

Our approach to the construction of the discrete BZ reaction system was based on photolithography-assisted techniques [28], which allowed the fabrication of the reactor with two-dimensionally arranged, microsized units. The reactor was made from the elastomeric material poly(dimethylsiloxane). By utilizing this methodology, we can freely control the size of reactor units, the spacing between neighboring units, and the number of units. In the experiment, we used the lattice-patterned array with reactor units of about 430 μm in diameter and 65 μm in depth (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. Looking on catalyst-doped microgels as active elements (or cells), a two-dimensional array of them can serve as a model for the study of a network of excitable neurons in living organisms. Measurements were carried out on arrays ranging in the number of elements N from 1 to 100, where all elements are considered to be nearly homogeneous. The chamber of the reactor was continuously fed with fresh, catalyst-free BZ solution at a pumping rate of 5 ml/h to maintain constant, nonequilibrium conditions. The initial composition of the catalyst-free

*Author to whom correspondence should be addressed.

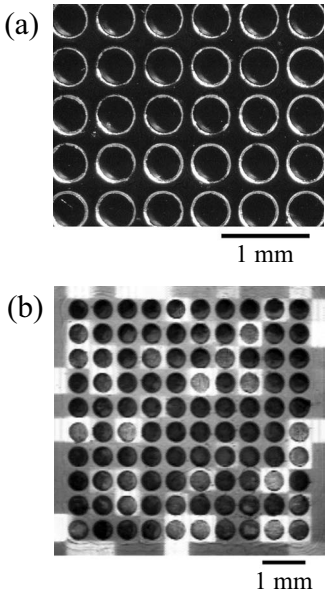

FIG. 1. (a) Patterned microreactor. Each reactor unit has a depth of 65 μm and a diameter of 430 μm. (b) Snapshot of the 10×10 microgel array superposed to the illumination pattern with random intensity values at each cell.

BZ solution was $[NaBrO_3]=0.33$ M, $[NaBr]=0.12$ M, $[CH_2(COOH)_2]=0.30$ M, and $[H_2SO_4]=0.32$ M. Reagent grade chemicals were used without further purification. 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 change 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. At this composition, the system was initially in an oscillatory regime. Increasing illuminated light intensity I beyond 6 mW drove the system to the excitable state. Then the period under the dark was estimated by extrapolation to be approximately $T_c=60$ s. In the experiment, we sustained the system in an excitable regime close to the bifurcation point by fixing I at $I_0=6.5$ mW. The pattern of illumination consisted of an array of square cells, whose light intensity was varied on an eight-bit gray scale between 0 and 255 (Fig. 1). The light intensity in the cell (i,j) is $I_{ij}(t)=I_0+\beta\xi_{ij}(t)$, where β is the noise amplitude and $\xi_{ij}(t)$ are statistically independent and random numbers equally distributed between -1 and 1, i.e., $\langle\xi_{ij}(t)\xi_{lm}(t')\rangle=\delta_{il}\delta_{jm}\delta(t-t')$ and $\langle\xi_{ij}(t)\rangle=0$. The noise pattern was updated at 4 s intervals and interrupted during 0.1 s every 1 s in order to capture a noise-free image of the system.

We evaluated the degree of temporal coherence of noise-induced firings by using the coherence measure R for N oscillators, defined by

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

where $\langle T_{ij}^m\rangle=(1/n)\Sigma_{k=1}^{n}(T_k^{ij})^m$, n 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). The coupling is accomplished via a mass diffusion, so that the spacing d between the nearest neighbors acts as a control parameter governing coupling behaviors. For very large d, all oscillators behave like independent oscillators for any noise amplitude, corresponding to the weak coupling regime. For very small d, on the contrary, all oscillators behave like a single oscillator, corresponding to the strong coupling regime. Therefore, we chose an intermediate value of d for the study of the effect of interplay between noise and coupling. Figure 2 shows the dependence of R on the number of oscillators at $d=80$ μm. We see that R reaches a maximum value R_{max} at an optimal noise amplitude β_{opt} in every array. This shows that the present oscillators have an ability to cause a coherence resonance. The period of oscillation at R_{max} approximately coincides with T_c, the period under the dark. It is important to emphasize that the degree of resonance is improved with increasing the number

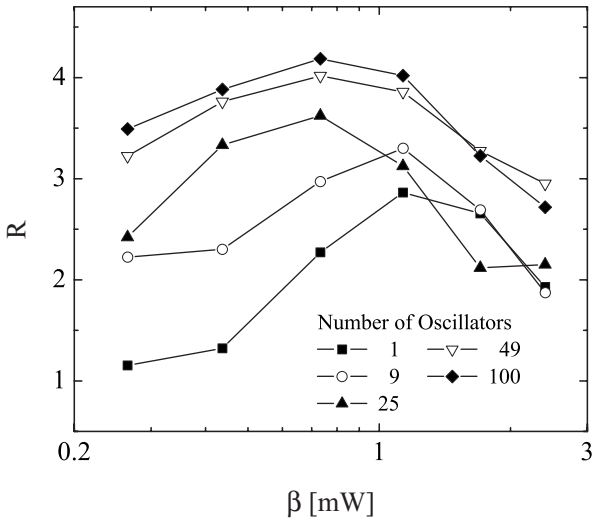

FIG. 2. Degree of coherence R as a function of the noise amplitude β at the separation $d=80$ μm for different values of the number of oscillators.

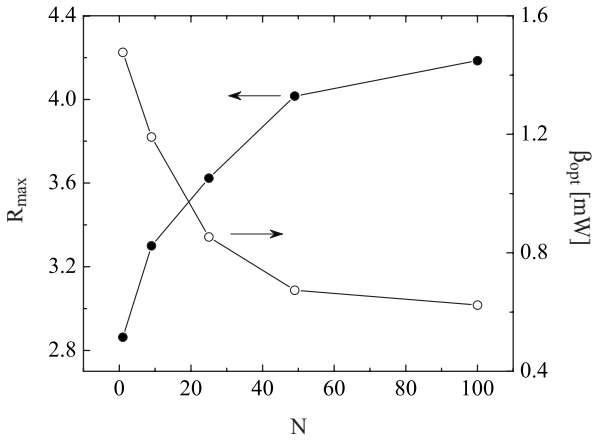

FIG. 3. Array size dependence of the maximum value R_{max} of R and the optimal noise amplitude β_{opt} as a function of the number of oscillators N at $d=80$ μm.

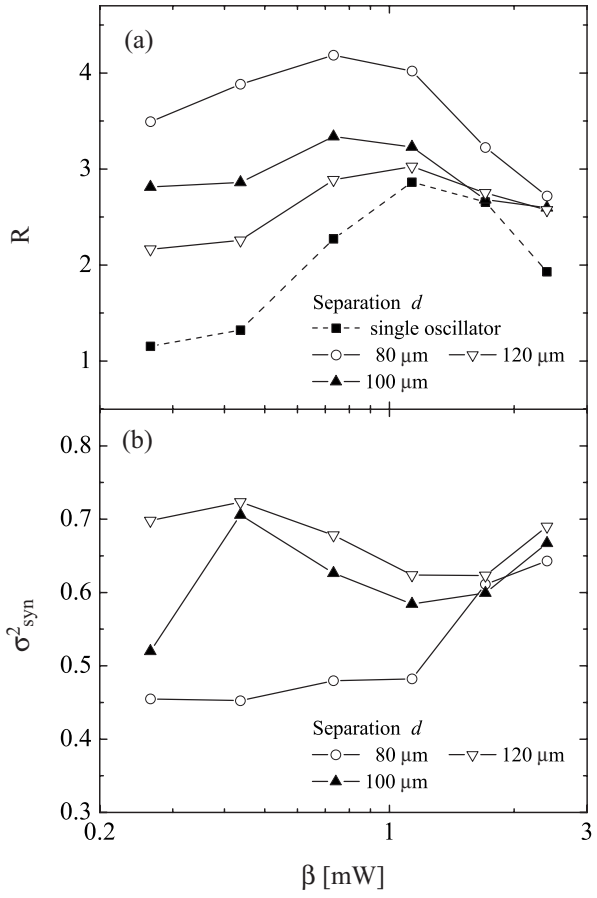

FIG. 4. (a) Coherence measure R and (b) phase synchronization measure σ^2_{syn} for various values of separation as a function of the noise amplitude β for the 10×10 array.

of oscillators from $N=1$ to 100. This indicates that temporal coherence is enhanced with an increase in array size, i.e., the occurrence of AECR. The values of $R_{\max}$ and β_{opt} depend on N, as shown in Fig. 3. With the increase of the array size, $R_{\max}$ and β_{opt} monotonically increases and decreases, respectively. However, they tend to saturate around $R_\infty \sim 4.2$ and $\beta_\infty \sim 0.61$ mW with increasing the array size, indicating that the effect of AECR is limited, even if the array size is made larger. In other words, there is the maximum coherence characteristic of the system.

Coupling an element into an array influences not only the temporal coherence but also the spatial coherence of noise-induced firings of elements. To characterize the synchronization behavior between oscillators in the array, we introduce a phase of the oscillators [22],

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

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 $\Phi_{ij,lm}=(\phi_{ij}-\phi_{lm})\mathrm{mod}\,2\pi$. We use the variance of the phase difference $\sigma^2_{ij,lm}$ as the measure of synchronization between oscillators (i,j) and (l,m). A low value of $\sigma^2_{ij,lm}$ indicates the synchronization between two oscillators. To characterize the degree of the array's phase coherence in a $M\times M$ lattice, we introduce the spatial average,

$$\sigma^2_{\mathrm{syn}} = \frac{1}{n_p}\sum_{l}^{M}\sum_{m}^{M}\sum_{i<l}\sum_{j<m}\sigma^2_{ij,lm}, \tag{3}$$

where n_p is the number of coupling pairs. In particular, $\sigma^2_{\mathrm{syn}}=0$ if all oscillators synchronize globally, whereas $\sigma^2_{\mathrm{syn}}=1$ if the oscillators are uncoupled. Here we took in account only the phase difference between the nearest neighbors.

Figure 4 shows the d dependence of the phase coherence, in addition to that of the temporal coherence, for a 10×10 lattice. For $d\geqslant140$ μm, all oscillators were uncoupled. At $d=120$ μm, the temporal coherence is improved a little, but the values of $R_{\max}$ and β_{opt} are almost the same as those of a single oscillator. This suggests that all oscillators still behave like independent oscillators because of weak coupling. This is also obvious from a high value of σ^2_{syn} on the whole. In the region of the noise amplitude close to β_{opt}, however, a minimal σ^2_{syn} appears, indicating that the phase synchronization is induced. This suggests that the phase synchronization becomes feasible through the occurrence of CR even among very weakly coupled oscillators. Thus the spatial coherence is closely related to the temporal coherence, in agreement with the results reported in Ref. [22]. When the spacing is very small, e.g., $d=80$ μm, the temporal coherence is highly enhanced. But a minimal σ^2_{syn} linked to a maximal R is not observed. Instead σ^2_{syn} takes small values in the range of small β, indicating the occurrence of synchronization, and steeply increases above $\beta\sim1.7$ mW. This behavior may be explained as follows, on the basis of a competition between noise and coupling. The small spacing between oscillators strengthens coupling interactions. Consequently, the synchronization of noise-induced firings can occur without the aid of noise, as usual events among coupled self-sustained oscillators. In the range of small β where coupling interactions surpass random forcing of noise, the phase-synchronized state is maintained. With further increasing β, however, random forcing conversely surpasses coupling interactions and destroys the synchronized state abruptly above a threshold. This may be regarded as a sort of order-disorder phase transition. At the intermediated spacing such as $d=100$ μm, both dynamics appear depending on β; the usual phase synchronization at small β and noise-enhanced phase synchronization around β_{opt}. Thus whether the effect of noise on the phase synchronization in the array is constructive or destructive depends on the coupling strength.

III. NUMERICAL SIMULATION

The system studied here is comprised of a two-dimensional square array of excitable oscillators coupled to its nearest neighbors through diffusion of chemical species. We employ the three-variable Oregonator model modified to take into account the effects of illumination noise. 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 element is influenced by the illumi-

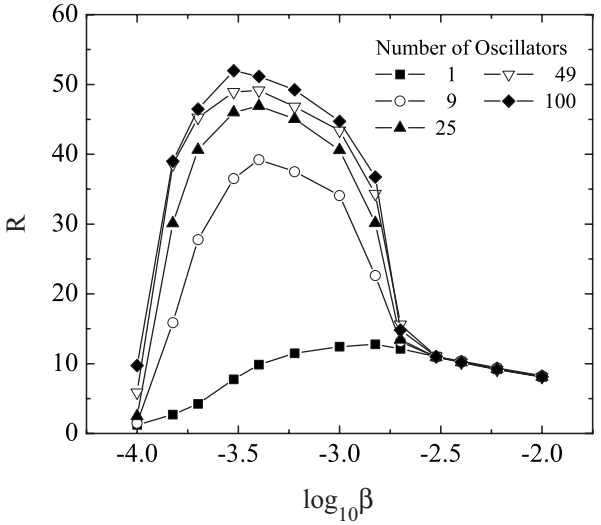

FIG. 5. Coherence measure R for various array sizes as a function of the noise amplitude β at the coupling strength $K_u=0.1$.

nation because the product of inhibitor Br^- is promoted due to the photochemical reaction of $Ru(bpy)_3^{2+}$. Then the model equations are given by

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

$$\frac{dv_{i,j}}{dt} = u_{i,j} - v_{i,j}, \quad (5)$$

$$\frac{dw_{i,j}}{dt} = \frac{1}{\varepsilon'}[fv_{i,j} - w_{i,j}(u_{i,j} + q) + \phi_{i,j}] + K_w(w_{i+1,j} + w_{i-1,j} + w_{i,j+1} + w_{i,j-1} - 4w_{i,j}), \quad (6)$$

where the variables $u_{i,j}$, $v_{i,j}$, and $w_{i,j}$ describe the concentrations of $HBrO_2$, the $Ru(bpy)_3^{3+}$ catalyst, and Br^- in the cell (i,j), respectively. The coupling among the elements is local and diffusive with the coupling strength K_u and K_w $(=1.12K_u)$. ε, ε', and q are scaling parameters and f is the stoichiometry parameter. The parameter $\phi_{i,j}=\phi_0+\beta\xi_{i,j}$ is the light-induced flow of Br^-, directly proportional to the illumination I_{ij}, where ϕ_0 corresponds to I_0 and $\beta\xi_{i,j}$ is the illumination noise with the noise amplitude β and the random numbers $\xi_{i,j}$ distributed uniformly in the interval $[-1,1]$, i.e., $\langle\xi_{ij}(t)\xi_{lm}(t')\rangle=\delta_{il}\delta_{jm}\delta(t-t')$ and $\langle\xi_{ij}(t)\rangle=0$. In the absence of the illumination, these parameters were chosen such that the system was initially in the oscillatory regime: $q=0.002$, $f=1.4$, $\varepsilon=0.01$, and $\varepsilon'=0.0001$. Increasing ϕ_0 beyond 0.00475 drove the system to the excitable state. We maintained the system in an excitable regime by setting ϕ_0 to a value near the bifurcation point, $\phi_0=0.005$. The computation was performed by the improved Euler method with time steps $\Delta t=0.0001$. The noise is independently subjected with the duration time $\delta=3000\Delta t$ to each oscillator. The spatial separation of the oscillators is taken as $\Delta x=1$. The boundary conditions for both edge elements were taken to be zero flux. We used the coherence measure R and the variance of the phase difference σ_{syn}^2 to characterize the degree of temporal coherence and phase synchronization, respectively.

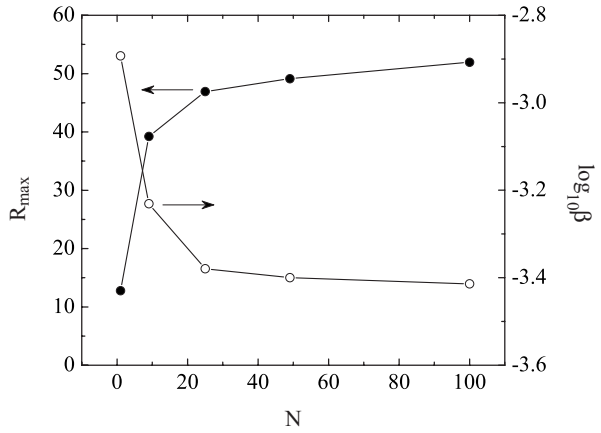

FIG. 6. Maximum coherence measure $R_{\max}$ and the optimal noise amplitude β_{opt} as a function of the number of oscillators N at the coupling strength $K_u=0.1$.

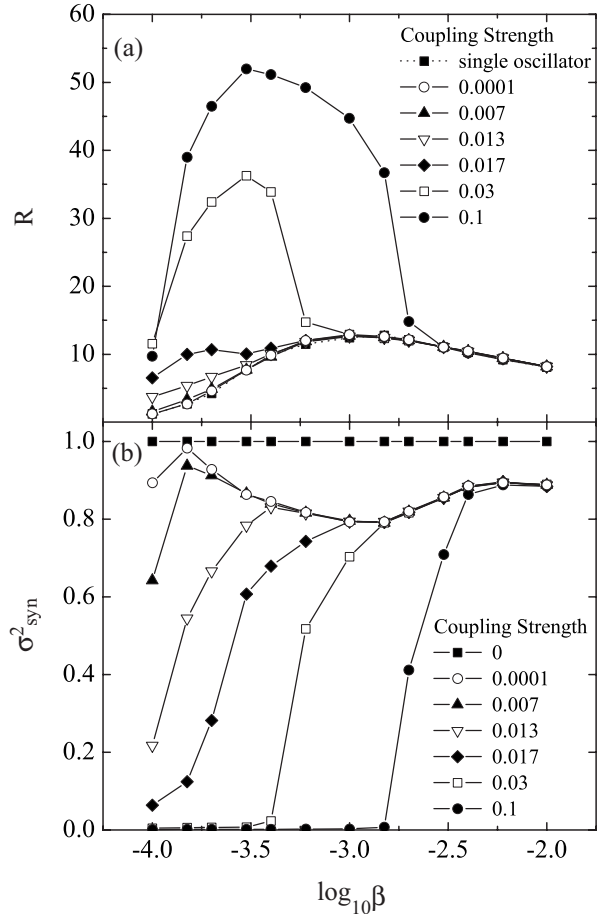

FIG. 7. (a) Coherence measure R and (b) phase synchronization measure σ_{syn}^2 on various coupling strengths as a function of the noise amplitude β for the 10×10 array.

In the limit of strong coupling, the oscillators behave as a single oscillator. To the contrary, in the limit of weak coupling the oscillators behave as if isolated. In between, therefore, we calculated R as a function of the number of oscillators. Figure 5 shows the case where the coupling strength is $K_u=0.1$. We see that, with an increase in β, R passes through the maximum value $R_{\max}$ at the optimal noise amplitude β_{opt} in every case. In addition, $R_{\max}$ increases with the array size, indicating the occurrence of AECR. Figure 6 shows the dependence of $R_{\max}$ and β_{opt} on the array size. With increasing the array size, $R_{\max}$ and β_{opt} monotonically increases and decreases, respectively. However, for large N, they tend to converge to constant values $R_\infty \sim 55.6$ and $\beta_\infty \sim 10^{-3.42}$, respectively. These behaviors are in agreement with the experimental results. We see that significant enhancement can be obtained even for a small number of elements. The saturation of β_{opt} with the array size is contrasted with the case of the array-enhanced stochastic resonance where the optimal noise amplitude increases linearly with N, although a maximum coherence measure converges to a constant value [29].

Synchronization behaviors were investigated on the array ranging in K_u from 0 to 0.1 for $N=100$, as shown in Fig. 7. For $K_u<0.0001$, all oscillators were always uncoupled for any value of β. When $K_u=0.0001$, the profile of R is almost similar to that of a single oscillator, where R takes a maximal value at the optimal noise amplitude $\beta_{\mathrm{opt}} \sim 10^{-3.0}$. As noticed by an appreciable minimum of σ^2_{syn} at β_{opt}, however, a phase synchronization, though incomplete, is improved. This is just a noise-enhanced phase synchronization, which shows that the phase coherence and the temporal coherence is closely linked to each other by the cooperation between noise and coupling. When $K_u \geqslant 0.03$, R is highly enhanced, where R takes a maximal value at the optimal noise amplitude $\beta_{\mathrm{opt}} \sim 10^{-3.5}$. Thus β_{opt} takes a value characteristic of the array size (see Fig. 6), as the effect of coupling becomes pronounced. A minimal σ^2_{syn} linked to a maximal R as observed in the weak regime does not appear. Instead σ^2_{syn} is almost zero in the range of small β, indicating that all oscillators synchronize globally. This is just similar to an usual phase synchronization of coupled self-sustained oscillators induced by strong coupling interactions. Increasing β beyond a threshold causes the abrupt transition from the globally synchronized state to the unsynchronized state, as observed in the experiment. It is thought that such a transition behavior results from a competition between noise and coupling, since the transition point shifts to a larger noise amplitude with an increase in K_u. Thus the interplay between noise and coupling does not always exert a positive effect on temporal coherence and phase coherence. In the intermediate region of $0.007 \leqslant K_u \leqslant 0.017$, the profile of R gradually deviates from that of a single oscillator with increasing K_u. At $K_u=0.017$ a tiny hump newly appears in the range of small β, where two types of synchronization described above occur depending on β. Thus the degree of coherence of the array in space and time strongly depends on the number of elements, the noise amplitude, and the coupling strength.

IV. CONCLUSION

We have experimentally and numerically investigated spatiotemporal dynamics induced in the two-dimensional array of excitable elements subjected to independent external noise. We have demonstrated evidence for array-enhanced coherence resonance and noise-enhanced phase synchronization; the maximum value of coherence $R_{\max}$ and the corresponding optimal noise amplitude β_{opt} monotonously increases and decreases with the increase of the array size, respectively. This means that the larger the number of elements, the more sensitive the system can respond to an external stimulus. However, observed array-enhanced effects tend to saturate for a further increase of the number of elements beyond 100. Noise-enhanced resonance and synchronization are closely correlated with each other. Even in the weak coupling regime where oscillators are barely coupled, synchronization can be achieved via coherence resonance. Our findings may be important in the operation of extended system such as biological receptors. We believe that the cooperation among noise, coupling, and nonlinearity of the chemical reaction plays an essential role in organizing these phenomena.

ACKNOWLEDGMENTS

This work was supported in part by a Grant-in-Aid for Scientific Research from the Ministry of Education, Culture, Sports, Science and Technology in Japan (Grant Nos. 15540377 and 18540385), and was also supported in part by the Central Research Institute of Fukuoka University.

[1] R. Benzi, A. Sutera, and A. Vulpiani, J. Phys. A **14**, L453 (1981); S. Fauve and F. Heslot, Phys. Lett. **97A**, 5 (1983); L. Gammaitoni, P. Hänggi, P. Jung, and F. Marchesoni, Rev. Mod. Phys. **70**, 223 (1998).

[2] B. McNamara, K. Wiesenfeld, and R. Roy, Phys. Rev. Lett. **60**, 2626 (1988); R. N. Mantegna, B. Spagnolo, and M. Trapanese, Phys. Rev. E **63**, 011101 (2000).

[3] J. K. Douglass, L. Wilkens, E. Pantazelou, and F. Moss, Nature (London) **365**, 337 (1993); B. J. Gluckman, T. I. Netoff, E. J. Neel, W. L. Ditto, M. L. Spano, and S. J. Schiff, Phys. Rev. Lett. **77**, 4098 (1996).

[4] A. Guderian, G. Dechert, K. P. W. Zeyer, and F. W. Schneider, J. Phys. Chem. **100**, 4437 (1996); A. Föster, M. Merget, and F. W. Schneider, *ibid.* **100**, 4442 (1996).

[5] K. Miyakawa, T. Tanaka, and H. Isikawa, Phys. Rev. E **67**, 066206 (2003).

[6] A. S. Pikovsky and J. Kurths, Phys. Rev. Lett. **78**, 775 (1997).

[7] Hu Gang, T. Ditzinger, C. Z. Ning, and H. Haken, Phys. Rev. Lett. **71**, 807 (1993); W. J. Rappel and S. H. Strogatz, Phys. Rev. E **50**, 3249 (1994).

[8] A. Longtin, Phys. Rev. E **55**, 868 (1997).

[9] Z. Hou and H. Xin, J. Chem. Phys. **111**, 721 (1999).

[10] D. E. Postnov, S. K. Han, T. G. Yim, and O. V. Sosnovtseva, Phys. Rev. E **59**, R3791 (1999).
[11] G. Giacomelli, M. Giudici, S. Balle, and J. R. Tredicce, Phys. Rev. Lett. **84**, 3298 (2000).
[12] K. Miyakawa and H. Isikawa, Phys. Rev. E **66**, 046204 (2002).
[13] S. Kádár, J. Wang, and K. Showalter, Nature (London) **391**, 770 (1998).
[14] S. Alonso, I. Sendiña-Nadal, V. Pérez-Muñuzuri, J. M. Sancho, and F. Sagués, Phys. Rev. Lett. **87**, 078302 (2001).
[15] K. Miyakawa, T. Okano, and T. Tanaka, Phys. Rev. E **71**, 066202 (2005).
[16] H. Hempel, L. Schimansky-Geier, and J. Garcia-Ojalvo, Phys. Rev. Lett. **82**, 3713 (1999).
[17] P. Jung and G. Mayer-Kress, Phys. Rev. Lett. **74**, 2130 (1995).
[18] J. F. Lindner, B. K. Meadows, W. L. Ditto, M. E. Inchiosa, and A. R. Bulsara, Phys. Rev. Lett. **75**, 3 (1995).
[19] F. Marchesoni, L. Gammaitoni, and A. R. Bulsara, Phys. Rev. Lett. **76**, 2609 (1996).
[20] M. Löcher, G. A. Johnson, and E. R. Hunt, Phys. Rev. Lett. **77**, 4698 (1996).
[21] B. Hu and C. Zhou, Phys. Rev. E **61**, R1001 (2000).
[22] C. Zhou, J. Kurths, and B. Hu, Phys. Rev. Lett. **87**, 098101 (2001).
[23] C. Zhou, J. Kurths, and B. Hu, Phys. Rev. E **67**, 030101(R) (2003).
[24] S. K. Han, T. G. Yim, D. E. Postnov, and O. V. Sosnovtseva, Phys. Rev. Lett. **83**, 1771 (1999).
[25] A. B. Neiman, L. Schimansky-Geier, A. Cornell-Bell, and F. Moss, Phys. Rev. Lett. **83**, 4896 (1999); A. B. Neiman and D. F. Russell, *ibid.* **88**, 138103 (2002).
[26] D. E. Postnov, O. V. Sosnovtseva, S. K. Han, and W. S. Kim, Phys. Rev. E **66**, 016203 (2002).
[27] M. Ohtaki, T. Tanaka, and K. Miyakawa, Phys. Rev. E **70**, 056219 (2004).
[28] B. T. Ginn, B. Steinbock, M. Kahveci, and O. Steinbock, J. Phys. Chem. A **108**, 1325 (2004).
[29] J. F. Lindner, B. K. Meadows, W. L. Ditto, M. E. Inchiosa, and A. R. Bulsara, Phys. Rev. E **53**, 2081 (1996).