Different operations on a single circuit: Field computation on an excitable chemical system

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Recently, it has been proposed that various kinds of time operations can be performed using an excitable field, mainly based on computer simulation. In this study, we performed experiments toward the realization of a time operation, such as time-difference detection. We used the photosensitive Belousov–Zhabotinsky reaction as a spatially distributed excitable field. We found that a single geometrical circuit can perform different operations with changes in the intensity of light illumination. The experimental results are discussed in relation to the idea of a non-Neumann-type computational device.
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INTRODUCTION

About 50 years ago, von Neumann proposed a computational device that could perform different operations on a single electronic circuit.[1] This idea was quite innovative, since with conventional analog computers different circuits must be used to perform different operations. A Neumann-type computer can perform different computations following the instructions in a program and is equipped with a CPU, clock, and memory. Modern human activity relies heavily on this idea, along with those of Turing.[2] Although a Neumann-type computer is very powerful, there exists an unavoidable drawback, i.e., all of the operations are processed through the CPU.

Higher-order animals perform computations with a complicated network of neurons. Biological neural nets appear to be different from a Neumann computer equipped with a CPU. Recent developments in brain science indicate that the ‘‘learning’’ process changes the transmissibility of signals through synaptic junctions.[3–5] This is the fundamental idea behind an artificial neural net such as back propagation[6] and Hopfield's neural net.[7] After ‘‘learning,’’ such an artificial neural net can provide correct answers in response to different inputs. In addition to such a framework of computations that are accompanied by a change in connectivity between neurons, it is expected that some biological computations are performed even without ‘‘reconnection’’ of the circuit. Considering the reactions of animals in different situations—for example, during the day and night or before and after eating—animals most likely perform computations in a different manner using the same neural network. Most probably even under same circuit, animal neural nets can perform different computations on the same input, depending on the context. A strategy of this type is more plausible in living organisms that do not have a complex network of neurons, such as plants and bacteria.

In the present study, we examined the possibility of constructing a non-Neumann-type computer based on the characteristics of an oscillatory and/or excitable spatial field. Showalter and co-workers[8,9] have proposed a novel idea based on a logic operation of an excitable field and have reported clear experimental verification of their idea with an excitable and oscillatory chemical system, the Belousov–Zhabotinsky (BZ) reaction.[10,11] Although their idea was interesting, due to their inability to create a ‘‘diode’’ function, the signal is obliged to propagate through the network of excitable fields in a ‘‘reversible’’ manner. The direction of signal propagation has no uniqueness, and the signal undergoes backpropagation along the same route between input and output. Recently, we reported that unidirectional signal transmission with an excitable propagating wave could be generated with a spatially asymmetric connection between excitable fields separated by a diffusion field.[12,13] Such a diode function has been seen in an actual experiment with an excitable-chemical system, the BZ reaction,[12,14] and also in computer simulations.[13,15–17] With numerical simulations, we have also proposed various logic gates, such as AND, NOT, OR, etc.[13,16] However, experiments to confirm the results of simulation have not yet been performed, partly because of the technical difficulty of cutting a membrane filter to a desired shape in the excitable-chemical system. In this study, we extended our idea to develop diode characteristics and

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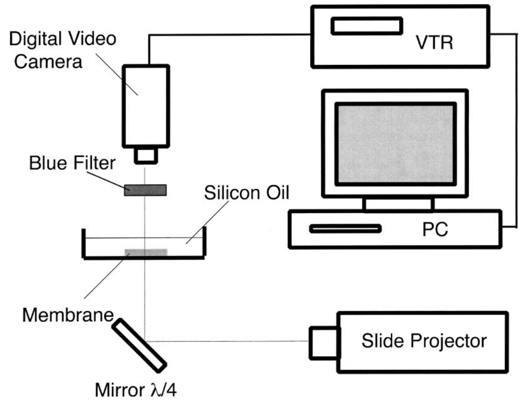

FIG. 1. Scheme of the experimental setup.

various kinds of logic gates with an excitable field. As an experimental model, we used a photosensitive version of the BZ reaction, where light illumination results in the production of bromide, which inhibits the oscillatory reaction. In other words, the degree of the excitability can be adjusted by changing the intensity of illumination.[14,17–20] Thus different kinds of computation can be processed with an excitable field of a single geometry, with suitable tuning of the light intensity as a field parameter.

EXPERIMENTAL METHODS

The reaction mixture was prepared as described previously from analytical-grade chemicals (Wako). The catalyst for the photosensitive reaction, $Ru(bpy)_3Br_2$, was synthesized and purified as in Refs. 21 and 22. The solution consisted of $[NaBrO_3]=0.15$ M, $[H_2SO_4]=0.3$ M, $[CH_2(COOH)_2]=0.2$ M, $[KBr]=0.05$ M, and $[Ru(bpy)_3Br_2]=2.04$ mM. Cellulose-nitrate membrane filters (Advantec A100A025A) with a pore size of 1 μm were soaked in BZ solution for 5 min. The membrane was gently wiped with filter paper to remove excess water and placed in a Petri dish. The surface of the membrane was immediately covered with silicon oil (Shin-Etsu Chemical Co.) to prevent it from drying and to protect it from the influence of oxygen. The experiments were carried out in an air-conditioned room at 20 ± 1 °C, at which the reaction medium showed no spontaneous excitation and no change in behavior for approximately 1 h. Excitation waves were initiated by gently touching the surface of the membrane with a 1-mm-thick silver wire.

The medium was illuminated from below as shown schematically in Fig. 1. The halogen bulb (JCD100V-300W) of a slide projector (Cabin Family II) was used as a light source, and the light intensity was varied by an external voltage controller. A black and white picture on a slide in the projector served as an illumination mask to create the appropriate boundary. The experiments were monitored with a digital video camera (Panasonic NV-DJ100) from above and recorded on a VTR (Panasonic NV-H200G). For image enhancement, a blue optical filter (AsahiTechnoGlass V-42) with a maximum transparency at 410 nm was used. The light intensity at the illuminated part was determined by a light intensity meter (ASONE LM-332).

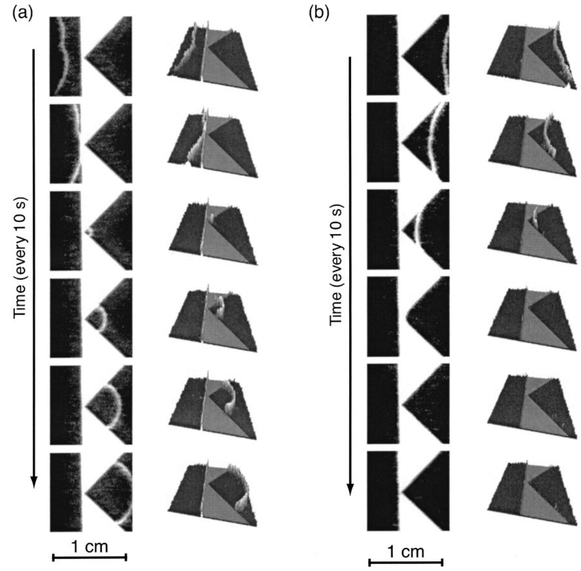

FIG. 2. Chemical diode on photosensitive BZ medium under light illumination. The light intensity in the white part was 3.23×10^4 lx. The black and white areas correspond to excitable and inhibitory fields. (a) Propagation of a chemical wave from left to right. (b) Propagation failure. The pictures on the left in (a) and (b) are video images and those on the right are quasi-three-dimensional representations, where the vertical change indicates the brightness or ''height'' of the chemical wave.

EXPERIMENTAL RESULTS

Figure 2 shows the manner of wave propagation around a ''chemical diode,'' where the white and black parts are illuminated and masked regions, respectively. In Fig. 2(a) the chemical wave passes through the gap, whereas in Fig. 2(b) the signal is not transmitted. A similar diode character has been observed by arranging a membrane filter in essentially the same geometry as in Fig. 2.[12] In the present setup, the ''chemical diode'' can only be constructed with illumination of the light-sensitive excitable field at a certain intensity.

Figure 3 shows a spatiotemporal diagram on the manner of wave propagation. At low light intensity ($I=3.07\times10^4$ lx) or at minimum photoinhibition of the excitable field, the wave propagates in both directions [Fig. 3(a)]. At intermediate intensity ($I=3.24\times10^4$ lx), the wave propagates only in one direction, from left to right [Fig. 3(b)]. At the highest intensity ($I=3.33\times10^4$ lx) or at the strongest inhibition, the wave is not propagated in either direction [Fig. 3(c)]. Thus the result in Fig. 3 clearly indicates that the manner of wave propagation can be switched merely by changing the relative excitability and without changing the spatial geometry of the ''circuit.''

Figure 4 shows an experiment on coincidence, under $I=2.76\times10^4$ lx. As shown in Fig. 4(a), when a wave propagates from left to right, the signal is not transmitted through the gap into the output channel. Figure 4(b) shows the AND operation: when two inputs arrive at the center almost simultaneously, a new wave is generated and propagated through the output channel.

Figure 5 shows an example of different operations on a single ''circuit.'' When the system exhibits low excitability with high light intensity ($I=3.53\times10^4$ lx), as shown in Fig.

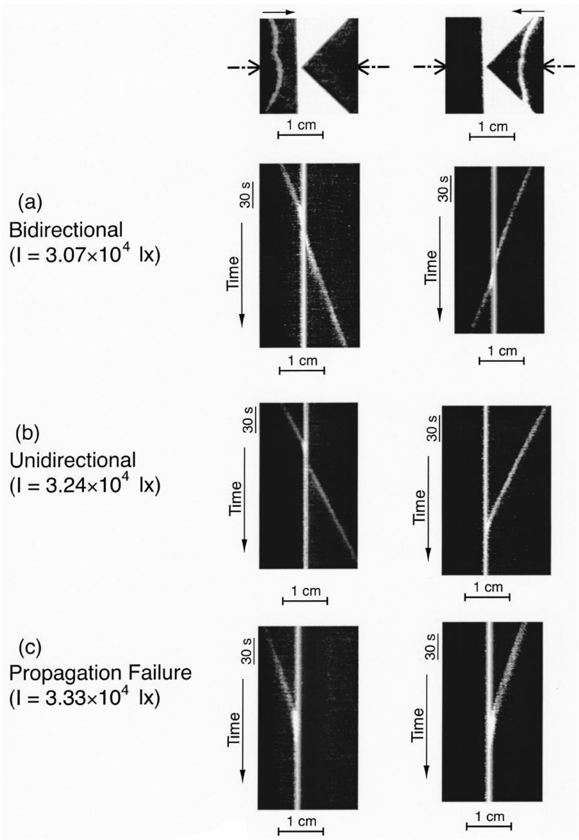

FIG. 3. Spatiotemporal representations of the manner of wave propagation, depending on the intensity of illuminated light. The geometry of the chemical diode is shown at the top, and is the same as in Fig. 2. (a) Bidirectional propagation. (b) Unidirectional propagation. (c) Propagation failure.

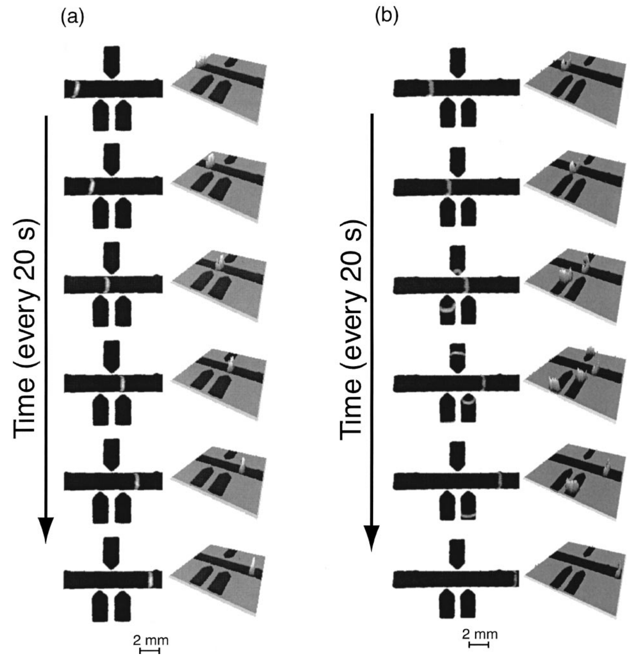

FIG. 5. Different operations with a single input. (a) At higher light intensity ($I=3.53\times 10^4$ lx), no output is generated. (b) At lower light intensity ($I=2.48\times 10^4$ lx), an output signal is seen at all three output channels.

5(a), a single chemical wave moving from left to right does not generate any signal in the output channel. In contrast, when the system is hyperexcited with low light intensity ($I=2.48\times 10^4$ lx), as shown in Fig. 5(b), a single wave causes new signals in all three output channels. Thus the system can be made either ''active'' or ''resting'' by controlling the light intensity.

By using a light intensity just above the threshold between the two modes shown in Fig. 5, one can perform an experiment on the coincidence of two opposing waves. Figure 6 shows an experimental time-difference detector, under $I=2.77\times 10^4$ lx. Similar to the results regarding the detection of coincidence in Fig. 4, when two input waves collide near a ''detector'' (the edge near the input channel), a new signal is generated in the output channel. Thus output channels I, II, and III detect time differences of $\Delta t=-11$, 0, and +11 s, respectively. On the other hand, when the time difference deviates or the collision occurs far from the edge detector, no signal is transmitted in the output channels.

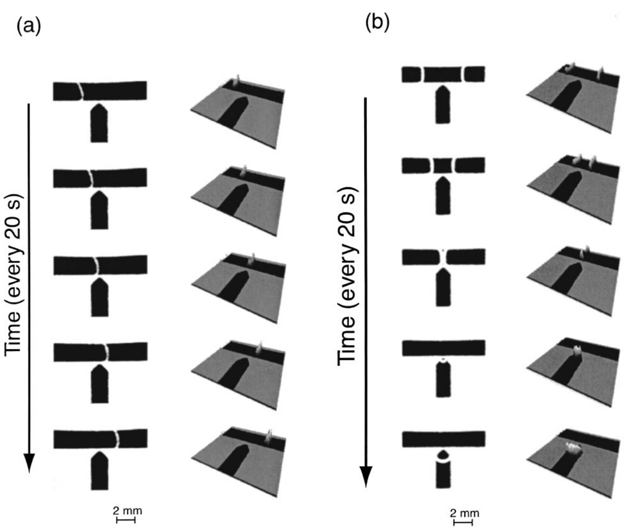

FIG. 4. Coincidence detection on photosensitive BZ medium at light intensity $I=2.75\times 10^4$ lx. (a) A single wave passing from left to right on the bar does not transmit a signal in the output channel. (b) When two waves collide near the ''gate,'' the signal is transmitted toward the output channel.

COMPUTER SIMULATION

The manner of wave propagation in the BZ reaction can be interpreted by a kinetic mechanism, the so-called Oregonator.[23] Numerical studies of these wave propagation phenomena were carried out using Tyson–Fife[24] scaling of the Oregonator model, modified to describe the photosensitive BZ reaction:[25,26]

$$\frac{\partial u}{\partial t}=D_u\nabla^2 u+\frac{1}{\epsilon_1}(qw-uw+u-u^2), \tag{1}$$

$$\frac{\partial v}{\partial t}=D_v\nabla^2 v+u-v, \tag{2}$$

$$\frac{\partial w}{\partial t}=D_w\nabla^2 w+\frac{1}{\epsilon_2}(\phi-qw-uw+fv), \tag{3}$$

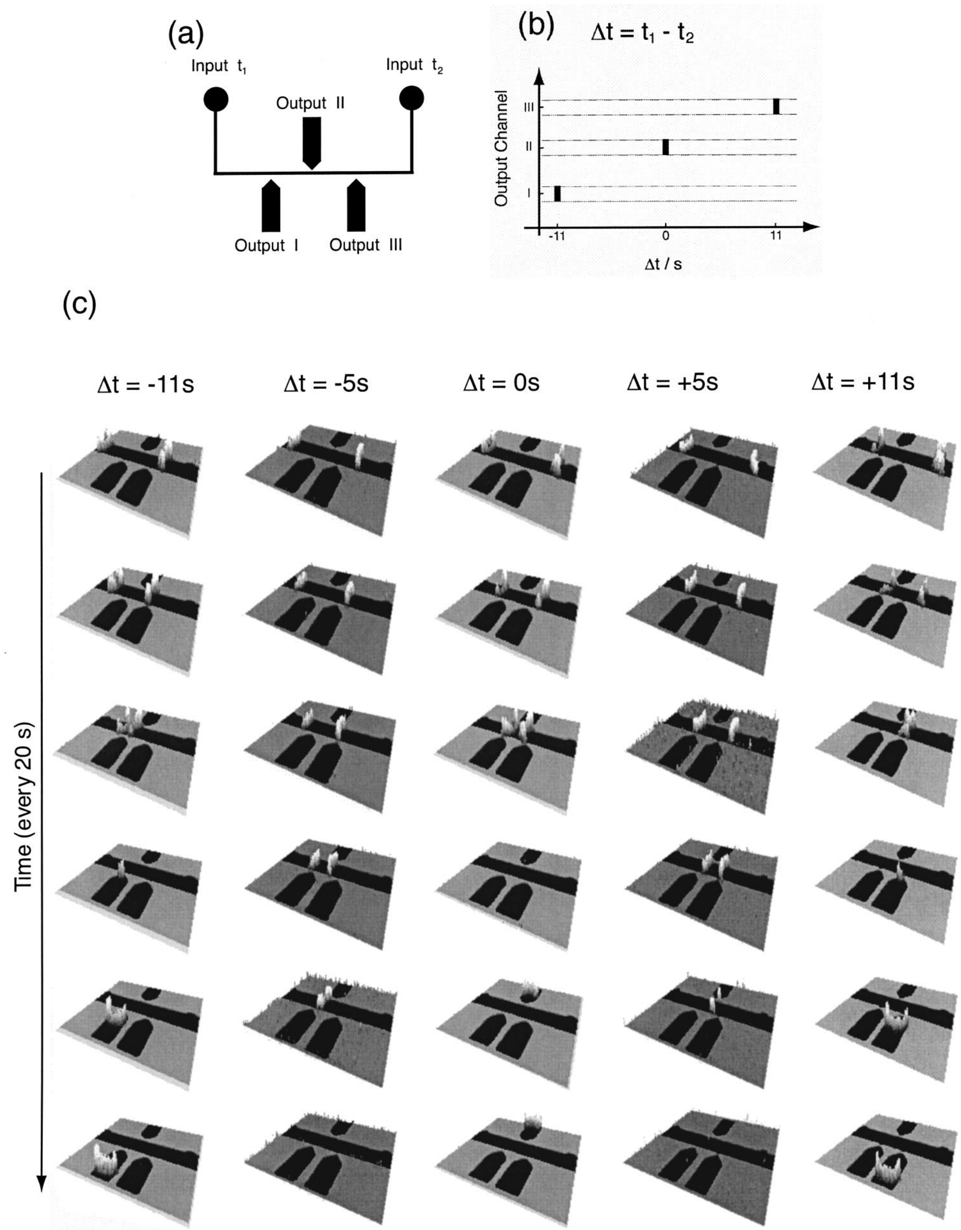

FIG. 6. Time-difference detection at light intensity $I = 2.77 \times 10^4$ lx. (a) Scheme of the time-difference detector. (b) Time difference detected at each channel. (c) Experiments at five different values of Δt. The geometry is the same as in Fig. 5.

where u, v, and w correspond to the dimensionless concentrations of $NaBrO_2$, $Ru(bpy)_3^{3+}$, and Br^-, respectively. D_u, D_v, and D_w are the diffusion coefficients for u, v, and w, respectively. ∇^2 means $\partial^2/\partial x^2 + \partial^2/\partial y^2$. The parameters $f = 1.0$, $q = 0.0015$, $\epsilon_1 = 0.03$, and $\epsilon_2 = 0.0003$ are kept constant throughout the calculations. D_u and D_w are taken to be equal with a good approximation and scaled to be unity: $D_u = D_w = 1$. As $Ru(bpy)_3^{3+}$ is almost immobilized in the membrane filter[27] adopted in the present study, D_v is set to be zero. The light intensity is proportional to ϕ.[25,26]

Numerical simulations were carried out on Eqs. (1)–(3) using the Euler method with a time step and ADI method[28] (alternating direction implicit method) with a space step. The grid size is 750×750 points in a square lattice, and the time interval is $\Delta t = 0.0003$ and the unit grid size is $\Delta h = 0.2$. We have numerically examined the effect of the grid size and confirmed that the essential manner of propagation is the same between the conditions, $\Delta h = 0.2$ and 0.04. In the present article, we show the numerical results with the grid size of $\Delta h = 0.2$. The boundary condition at the edge of the frame is taken to be no flux, while that between the excitable and inhibitory fields is free.

Figure 7 shows the results for a numerical simulation of a "chemical diode." The wave behavior was calculated from Eqs. (1)–(3), where ϕ corresponds to the light intensity. We used $\phi = 0.08$. This result reproduces well the experimental trends shown in Fig. 2.

Figure 8 shows the spatiotemporal plot of wave propagation depending on the light intensity in a numerical simulation. At a low light intensity ($\phi = 0.07$), a wave propagates through the gap in both directions. When the light intensity is increased to $\phi = 0.08$, a wave can only pass through the gap from left to right. When $\phi = 0.09$, a wave cannot propagate in either direction. These results in the simulation correspond well to the experimental observations (Fig. 3).

It may be of value to indicate some difference between

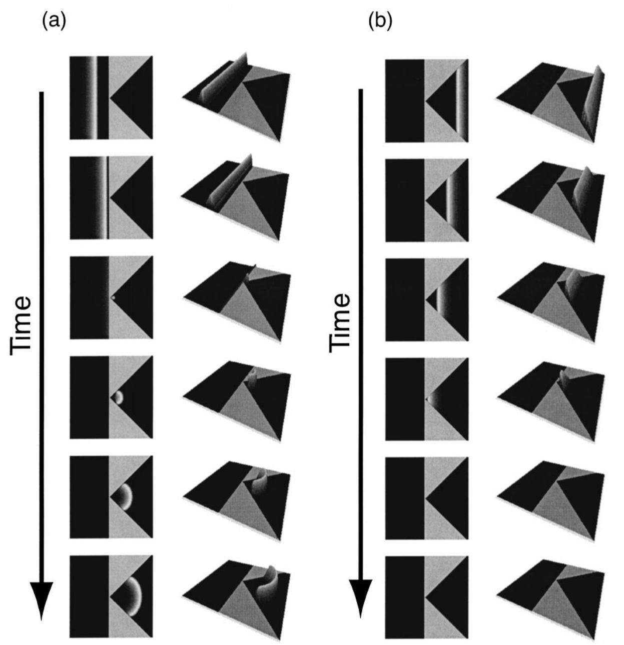

FIG. 7. Computer simulation of wave propagation on a ''diode,'' at a time interval of 1.5 using the coupled differential equations (1)–(3). (a) A signal is transmitted from left to right. (b) Propagation failure. In the pictures on the left in (a) and (b), the value of ν is indicated by the gray level where white corresponds to the maximum. The pictures on the right in (a) and (b) are the quasi-three-dimensional representations. These figures show only the center region (grid size 215×215 points) of the calculated field (grid size 750×750 points). Black and gray regions correspond to excitable and inhibitory fields, respectively, and the bright region indicates a propagating wave in quasicolor representation with the variable ν in Eqs. (1)–(3).

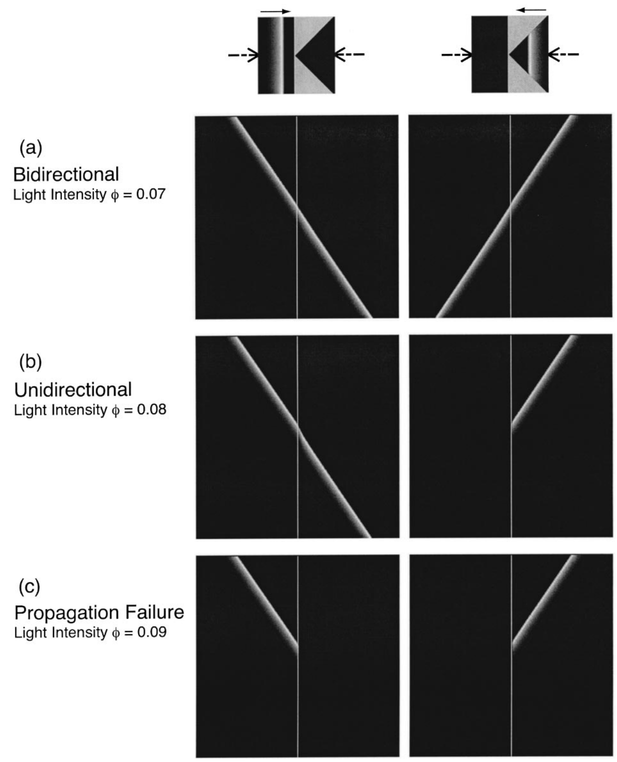

FIG. 8. Computer simulation of the manner of wave propagation with the same spatial geometry as in Fig. 7. Space-time plot of traveling waves with the variable ν in Eqs. (1)–(3) along the horizontal centerline of the diode geometry (750 points).

the experiments and numerical simulations; for example, plane waves are generated in Fig. 7, whereas in experiments as in Fig. 2, the wave fronts are curved. According to the standard theoretical consideration[29] on the stability of traveling wave, the critical radius on the wave propagation is given as $R_c = D/c$, where D and c are correspond to the diffusion coefficient and velocity of wave propagation, respectively. From the experimental observation c is around 10^{-2} cm/s. With the plausible order of D as 10^{-5} cm^2/s, R_c is estimated as 10^{-3} cm. This means that the effect of curvature may have negligible importance on the manner of wave propagation, at least in our experimental conditions. Curvature effect may become more significant on the experiments with smaller size in the experimental system.

DISCUSSION

Our results have shown that different operations can be performed depending on the excitability of a system. It is also interesting to note that the time operations can be performed without a clock. In a Neumann computer, a central clock is needed to perform time operations. In the present study, it is shown that this new idea actually works in excitable chemical medium, i.e., the BZ reaction. Thus it is becoming clear that a rich variety of operations can be performed using an excitable field, without a clock, CPU, or program. The manner of the operation of an excitable field, or field computation, can be selected by controlling the field excitability as a whole. Thus the light intensity in the present study acts as a kind of ''hormone'' to tune the activity of the system. This means that there should be no unavoidable bottleneck as seen in the usual Neumann-type computers. It is also to be mentioned that the adjustment of the excitability with light intensity means a tough reproducibility in the experiments; essentially the same manner of wave propagations is obtained even at different temperature by tuning the light intensity. On the other hand, the BZ reaction is quite fragile: the wet-reaction medium is not suitable for use in a practical computing machine. If a solid-state excitable system can be identified, it may be useful to examine the possibility of field computation with such a medium. Recently, the generation of an excitable wave has been observed on a solid system.[30] Trial on the fabrication of reaction-diffusion chip has also been reported using a semiconductor.[31] For the practical application of a ''field computer,'' it will be necessary to construct a ''circuit'' on a solid substrate. In addition, it would be interesting to consider a hybrid system with the connection of a ''field computer'' and a Neumann-type computer. That is, the output signals in a ''field computer'' can be transformed into a one-dimensional arrangement of symbols, as in a ''Turing machine.'' This indicates that the output from operations with a ''field computer'' can be further processed with a Neumann-type computer. The idea of a hybrid computing system using field and Neumann computers may be a promising target for the further development of the computational devices.

With regard to the spatial geometry of the time-difference detector shown in Fig. 6, it is interesting to note that a similar array is found in the auditory nerves of the owl.[32] It has also been proposed that cortical neurons may play a role in coincidence detection instead of integration and, thus, select for correlated input.[32,33] However, this hypothesis is still tentative,[33] due to both a scarcity of reliable experimental evidence and a lack of theoretical support. As an extension of the present study, it may be useful to look for the actual mechanism of the time operation in a real biological system.

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