CHEMICAL IMPULSES IN THE PERPENDICULAR JUNCTION OF TWO CHANNELS*

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It is known that nonlinear chemical systems may be used for direct information processing. In this paper we study properties of the perpendicular junction of two channels filled with an excitable medium as a function of a time difference of arriving impulses. It is shown that depending on this difference the cross junction works as a coincidence detector or as a signal switch.

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1. Introduction

The idea of information processing with purely chemical devices is attractive for both scientific community and leading hi-tech companies. Two most promising ways of constructing such devices come from the molecular chemistry approach (the information is processed by individual molecules) [1] and from the nonlinear chemical reactions [2,3]. Both possibilities are currently extensively investigated.

In the study presented below we are concerned with a model chemical system which can be used for direct information processing. Our system utilizes chemical reactions occurring in an excitable chemical medium. The idea seems both interesting and feasible due to the analogy with the processes occurring in neural systems of living organisms. Such processes as decision taking or memorizing may be considered as examples of similar chemical information processing, because the neural signals can be described by traveling impulses in an excitable chemical medium. The successful applications of chemical waves in image processing (including both contrast

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and contour analysis) have been reported in [2]. In contrary to conventional computers the chemical device should be able to work without any ruling clock [4], which prescribes subsequent elementary operations to particular silicone sub-units.

The topology and geometry of the chemical medium are important factors in the construction of the desired device. Chemical realizations of a diode [5], basic logical gates (AND, OR, NOT) [6,7] and a simple memory device [7] have already been proposed. Even more advanced systems, performing complex logic operations, have been constructed [3]. Some of those ideas have also been checked in laboratory experiments [3,6]. They are all based on a carefully chosen reaction scheme and the geometry of the system in which reactions occur. Thus, investigations of the propagation of chemical waves in excitable systems with various geometry are crucial for building the "chemical computer". In this paper we present the properties of a junction of two perpendicular channels (we call it a cross junction).

2. The model of spatially extended chemical excitable system

We call a chemical system excitable if it has a single stable stationary state of a special kind. Small perturbations of this state decay exponentially in time. However, if the amplitude of a perturbation exceeds a certain critical value, then the perturbation increases leading to macroscopic changes of the concentrations of chemical components. Finally the system returns to its stable stationary state after a non-trivial evolution in the phase space. If the system is distributed in space and locally perturbed, then the local excitation may propagate to the neighboring areas, while the points initially excited relax to the stationary state. Thus, for the proper choice of parameters a traveling impulse may appear in the system.

In this study we use a FitzHugh–Nagumo type model [7–9], consisting of two variables: u and v. The time evolution of their values is given by the following set of equations:

$$\tau \frac{\partial u}{\partial t} = -\gamma \left[ku(u-\alpha)(u-1)+v\right] + D_u \nabla^2 u \,, \tag{1}$$

$$\frac{\partial v}{\partial t} = -\gamma u \,. \tag{2}$$

The variables u and v in Eqs. (1), (2) may be uniquely related to concentrations of active chemical reagents and can be either negative or positive. One can also say that Eqs. (1), (2) describe the time evolution of a "chemical" system with two components. One of them, described by u, is the "activator", because it speeds up its own production, and the other, described by

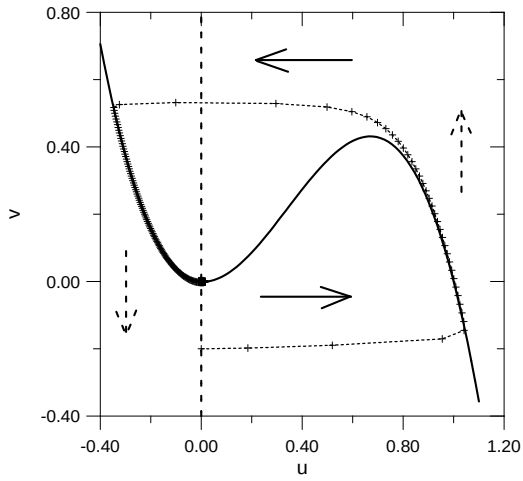

Fig. 1. The isoclines of Eq. (1) without the diffusion term (solid line) and of Eq. (2) (dashed vertical line identical with the v axis) for the values of parameters given in Sec. 2. Solid and dashed arrows indicate the driving forces of u and v respectively. The isoclines have only one common point $u = v = 0$, indicating the stationary homogeneous solution of Eqs. (1), (2). The dotted line with symbols shows the evolution of the system in the phase space after a sufficiently large perturbation (initially $u_i = 0.0$ and $v_i = -0.2$).

v, is the "inhibitor", because it slows down the production of u. In this sense the values of u and v stand for the "concentrations" of the corresponding "reagents". In the spatially distributed system we assume that the inhibitor is always immobilized (it never diffuses). The system (1), (2) is excitable for the following parameters describing reactions: $\gamma = 1$, $\tau = 0.03$, $k = 3.0$, $\alpha = 0.02$ [7]. The isoclines of Eq. (1) without the diffusion term (solid line) and of Eq. (2) (dashed vertical line identical with the v axis) for these values of parameters are shown in Fig. 1. Solid and dashed arrows indicate the driving forces of u and v respectively. The isoclines have only one common point $u = v = 0$, indicating the stationary homogeneous solution of Eqs. (1), (2). The dotted line with symbols shows a possible evolution of the system in the phase space after a sufficiently large perturbation. This is where the excitability of the system can be observed. The diffusion coefficient of activator D_u equals 0.00045 because for this value well defined pulses propagate in the system [7].

We also consider another type of medium (the diffusion one), in which no reactions occur and only diffusion of the activator u is possible. This medium is described with the following set of equations [7]:

$$\tau \frac{\partial u}{\partial t} = D_u \nabla^2 u \,, \tag{3}$$

$$v = 0 = \text{const.} \tag{4}$$

with $k = 0.03$ and $D_u = 0.00045$ [7], as previously.

3. Cross junction and its properties

Let us consider a plain (two dimensional) system with geometry shown in Fig. 2, where the brighter areas correspond to the excitable field and the darker areas — to the diffusion one. In the excitable field the time evolution of the concentrations of u and v is given by Eqs. (1), (2) while in the diffusion field — by Eqs. (3), (4). We assume that activator u can freely flow between excitable and diffusion fields. The excitable and diffusion fields in the device shown below form two perpendicular "channels", in which traveling impulses can propagate. To simplify the description that follows let us assume that the excitable areas of our cross junction are numbered as in Fig. 2.

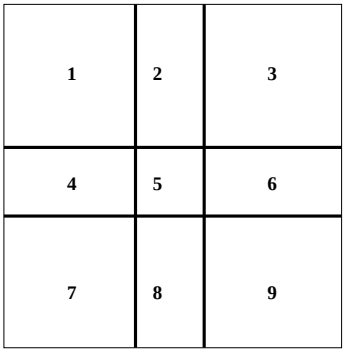

Fig. 2. Geometry of the considered device: brighter areas correspond to the excitable field and the darker areas — to the diffusion one. The excitable areas are numbered to simplify the description in text.

The properties of a pulse propagating on a semi-plane of an excitable field joined with another semi-plane of the diffusion field were studied in [7]. When a wave propagated perpendicularly to the line of junction of the semi-planes it penetrated the diffusion field up to certain distance d_c deep. Here penetration means that if another active medium was closer than d_c from the boundary then an excitable pulse was activated on it by the primary

wave. On the other hand a wave propagating parallel to the line of junction penetrated the diffusion field up to $0.94\,d_c$ deep only. It means that a layer of a width $d\,(0.94\,d_c < d < d_c)$ is transparent for a pulse propagating perpendicularly, but impenetrable for a pulse propagating parallel to it.

This result implies that if the width of the stripes of the diffusion field d_d is between $0.94\,d_c$ and d_c then the cross junction presented above may have interesting properties. It can be expected that:

(i) A single impulse propagating in one of the channels is able to propagate along its own channel, but it cannot move "sideways" (because it cannot cross the stripe of the diffusion field parallel to the direction of its motion, as $d_d > 0.94\,d_c$).

(ii) The impulse is able to propagate through the junction of two channels (because it is able to cross the stripe of the diffusion field perpendicular to the direction of its motion, as $d_d < d_c$).

For the values of parameters given in Sec. 2 the appropriate width of diffusion field is $d_d \approx 0.16$.

In our study the pulses are initiated on the border by decreasing the value of the inhibitor v inside the channel to $v_i = -0.2$, the answer of a homogeneous system with respect to such excitation is shown in Fig. 1. The stationary shape of the impulse is obtained after $t_s \approx 2.50$ and the velocity of the free propagation of such an impulse is $v_s \approx 0.77$. Thus after propagation over the distance of 2 units the impulse retains its stationary form.

Fig. 3 presents a typical stationary shape of a traveling impulse in terms of the variable u. Brighter areas correspond to the higher values of u. The thin stripes of the diffusion field are located between the pairs of dashed lines (the stripes are not filled black, so that the picture is more clear). One can see characteristic "whiskers" of the impulse, slightly sticking outside the stripes of the diffusion field. The collision of those "whiskers" explains part of the characteristics of the cross junction given below (cases A and C). Such stationary shape of a pulse does not depend on the mechanism of initialization.

If the width of the excitable field inside the channel (d_e) is large enough $(d_e > 0.4)$ we do not observe its influence on the amplitude and velocity of a pulse. In very narrow channels $(d_e < 0.1)$ both amplitude and velocity are visibly smaller than those characterizing a pulse in a wide channel.

The features of a pulse mentioned above are important for an impulse propagating through the junction. The data of penetration of diffusion filed given by Motoike and Yoshikawa in [7] applies to pulses propagating in wide channels. If the width of the channel is smaller than 0.7 then the penetration depth for a transverse propagation decreases. The width of the excitable field inside each channel is also important for an impulse to

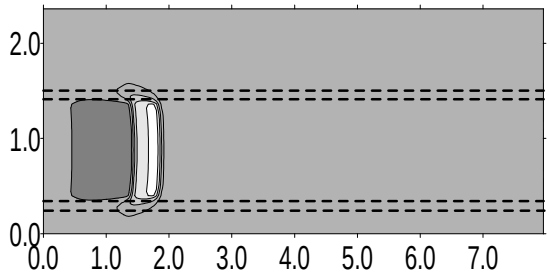

Fig. 3. Typical stationary shape of a traveling impulse in terms of the variable u. Brighter areas correspond to the higher values of u. The thin stripes of the diffusion field are located between the pairs of dashed lines (the stripes are not filled black, so that the picture is more clear).

propagate through the junction, because the impulse needs enough space between the two stripes of the diffusion field on its way to cross both of them. In other words, to obtain the propagation of an impulse from area 4 to 6 in Fig. 2 — the area 5 cannot be too small. We have found that in order to obtain the junction with features *(i)* and *(ii)* the inner diameter of the channel d_e should not be lower than *ca.* 0.8.

All the results presented below are obtained by integrating Eqs. (1), (4) numerically. The calculations were carried out with an implicit method based on the Crank–Nicolson discretization of the Laplace operator [10]. We use the values of parameters given in Sec. 2, $d_d = 0.16$ and $d_e = 1$. The time step of the integration is $dt = 0.005$. In calculations we consider three sizes of the square the junction is built on ($5 \times 5, 7 \times 7$ and 8×8), for which we use square lattices of $250 \times 250, 350 \times 350$ or 400×400 points respectively. Thus, the space step in all these simulations $dx = dy = 0.02$ remains constant. The results for different system's sizes are consistent. The boundary conditions are free between the excitable and diffusion fields and no flux on the border of the square.

To find the time characteristics of the cross junction we use the following technique. In each of the input channels (area 4 and 8) one input signal (an impulse) is placed. The impulses are initialized on the border of the square by decreasing the value of v inside the channel to $v_i = -0.2$. For various experiments the channels are located in different parts of the square, so that the impulses travel over various distances before they reach the cross junction. This way, due to a constant velocity of the propagation of impulses ($v_s \approx 0.77$), we are able to adjust the time difference Δt between their arrival at the junction. Depending on Δt we obtain seven types of the junction's response, illustrated in Figs. 4–10. The figures marked as (a) to (f) show the time evolution of the activator in the junction (the exact timing of snapshots (a) to (f) is given in figure captions).

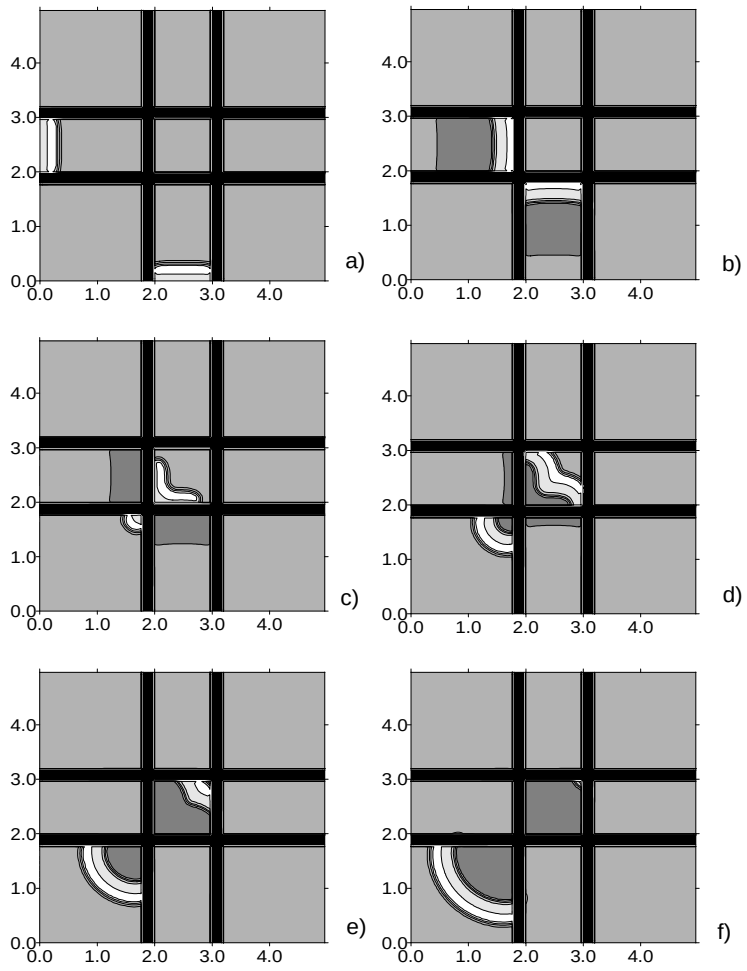

Fig. 4. The "AND" logical gate for $\Delta t = 0.00$. The output signal is produced in area 7 between the two input channels (4 and 8). The consecutive snapshots correspond to (a) $t = 0.5$, (b) $t = 2.5$, (c) $t = 3.5$, (d) $t = 4.0$, (e) $t = 4.5$, (f) $t = 5.0$.

Due to the symmetry of the system let us only discuss the case in which the first pulse is initiated in area 4 and the later in area 8. The behavior of the cross junction may be classified in the following way:

A. For a very small time difference between incoming signals ($\Delta t \leq 0.26$ the cross junction acts as the "AND" logical gate. The output signal is produced in area 7 between the two input channels (4 and 8) if and only if there are two input signals arriving at the junction within the given time difference. We think that the output signal appears when the "whiskers" of the two traveling impulses meet (see Fig. 4).

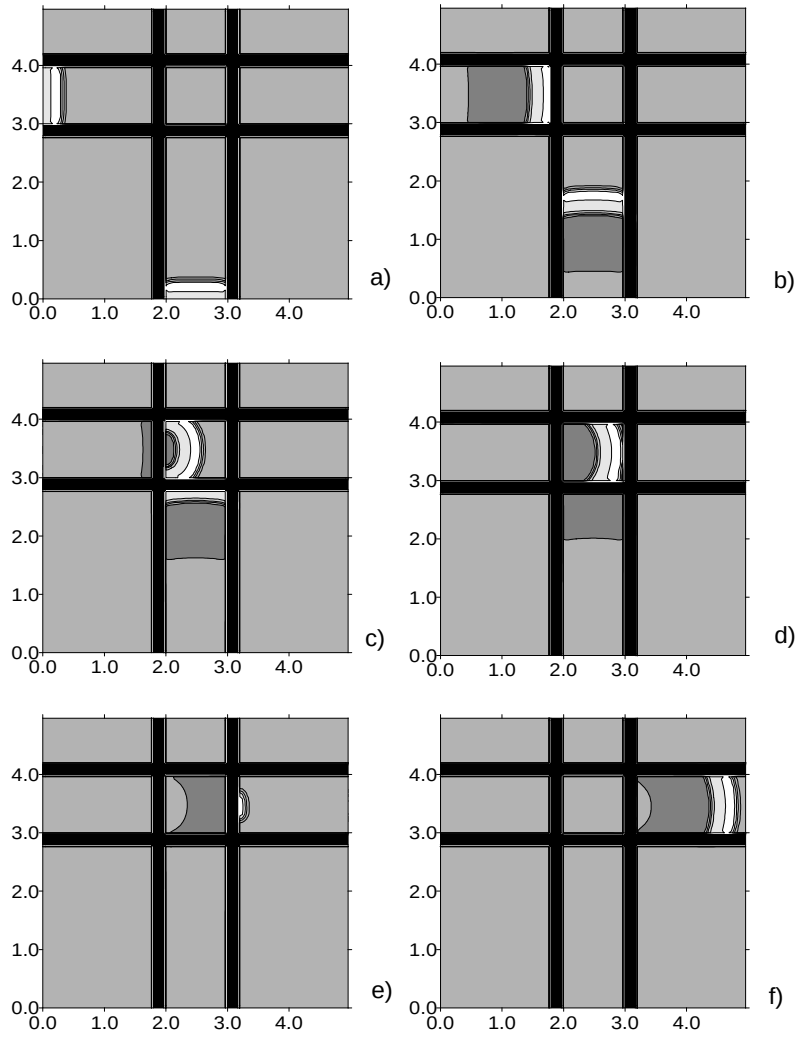

Fig. 5. Only the earlier signal survives (within its own channel; area 6) for $\Delta t = 1.29$. The consecutive snapshots correspond to (a) $t = 0.5$, (b) $t = 2.5$, (c) $t=4.0$, (d) $t=4.5$, (e) $t=5.5$, (f) $t=7.5$.

B. For the time difference in the range $0.28 \leq \Delta t \leq 3.45$ only the earlier signal survives and propagates within its own channel — area 6. Here the cross junction works (as expected) as a coincidence detector in which the earlier impulse switches the central field into the refractory regime and blocks the propagation of the second signal (see Fig. 5).

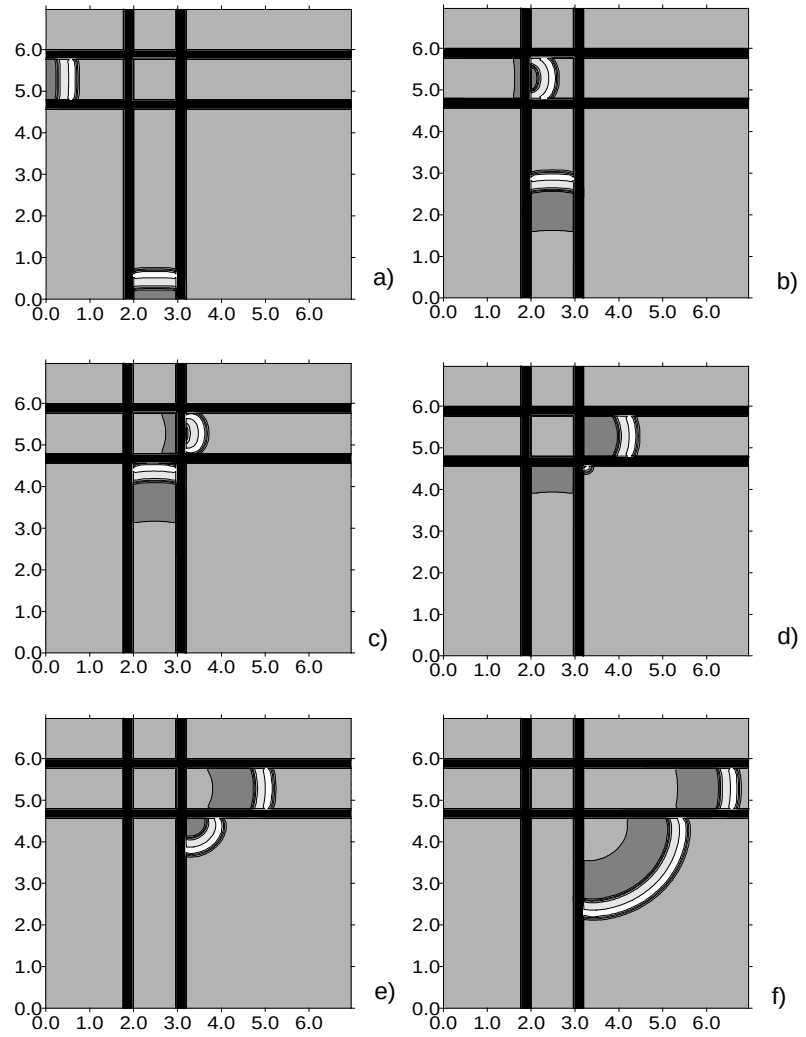

Fig. 6. The earlier signal survives in its own channel (area 6) and in addition a new output signal is produced area 9 for $\Delta t = 3.61$. The consecutive snapshots correspond to (a) $t=1.0$, (b) $t=4.0$, (c) $t=6.0$, (d) $t=7.0$, (e) $t=8.0$, (f) $t=10.0$.

C. When $3.48 \leq \Delta t \leq 3.79$ the earlier signal survives in its own channel (area 6) and in addition a new output signal is produced in the lower right-hand corner of the device (area 9). In our opinion the new output signal appears due to the same mechanism as in the case A *i.e.* due to the coincidence of the "whiskers" (see Fig. 6).

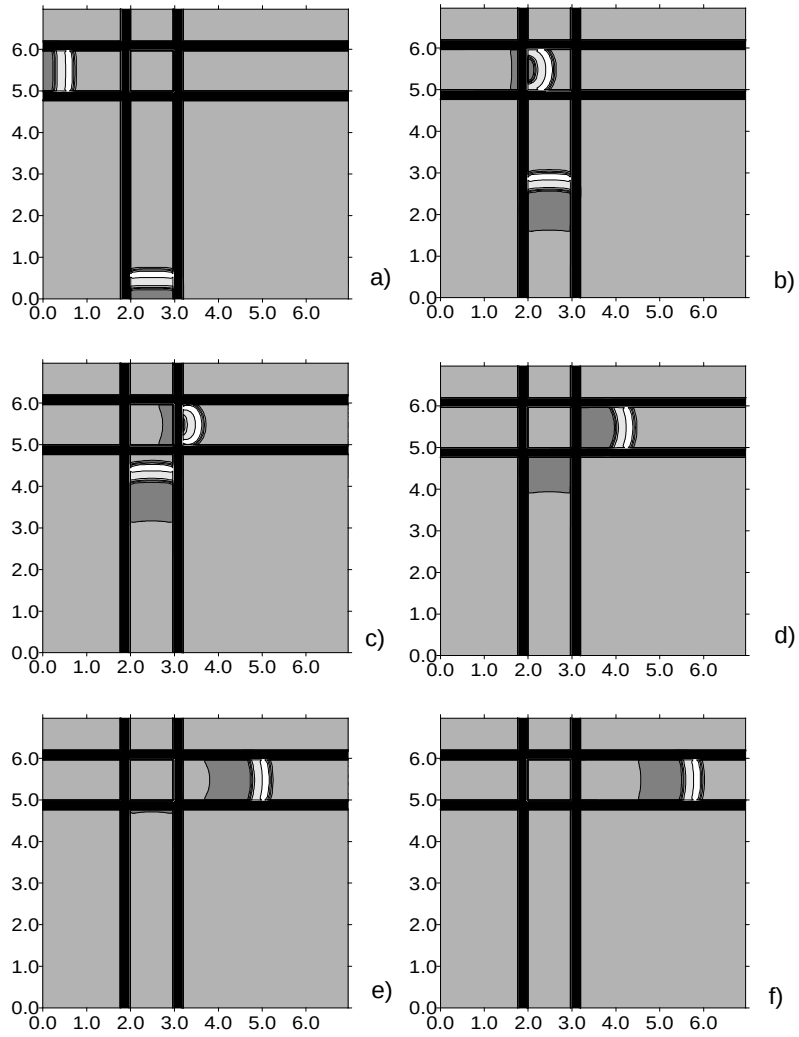

Fig. 7. For Δt=3.87 only the earlier incoming signal survives (area 6). The consecutive snapshots correspond to (a) $t = 1.0$, (b) $t = 4.0$, (c) $t = 6.0$, (d) $t = 7.0$ (e) $t = 8.0$ (f) $t = 9.0$.

D. For $3.81 \leq \Delta t \leq 4.22$ the evolution is as in the case B: only the earlier incoming signal survives and finally propagates in area 6 (see Fig. 7).

E. For $\Delta t \approx 4.25$ the earlier incoming signal follows the way within its channel (area 6), while the other signal is switched from the original path and follows the first signal (see Fig. 8).

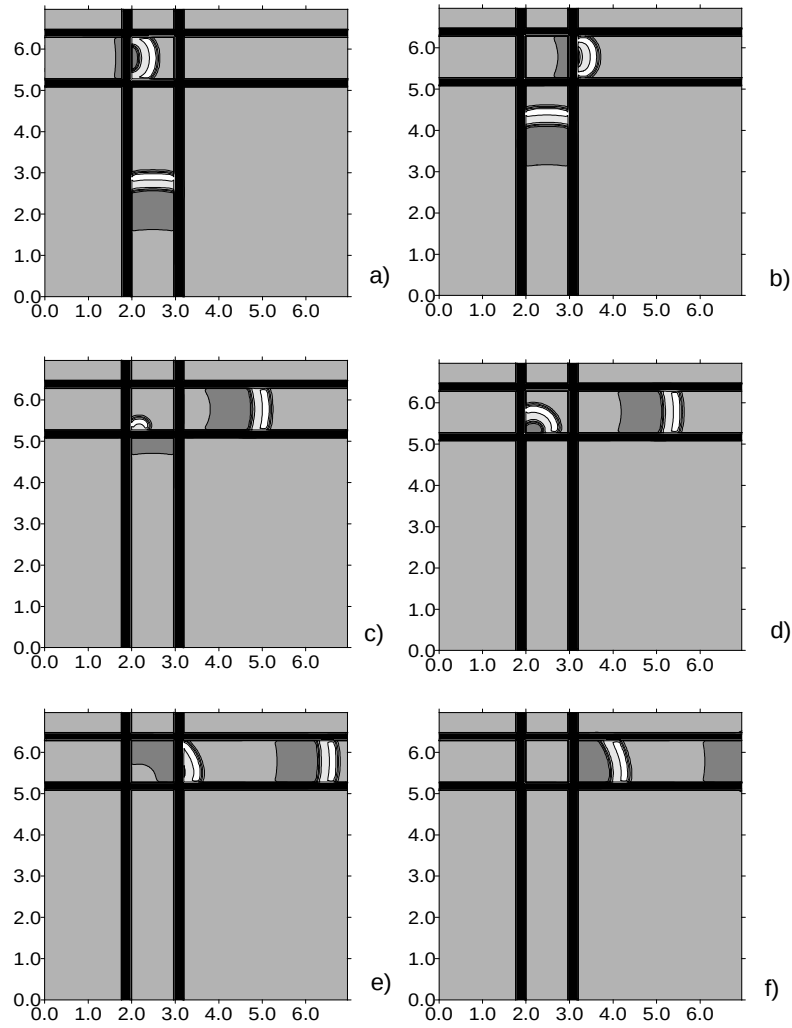

Fig. 8. For Δt=4.25 the earlier incoming signal follows the way within its channel (area 6) while the other signal is switched from the original path and follows the first signal (area 6). The consecutive snapshots correspond to (a) $t = 4.0$, (b) $t = 6.0$, (c) $t = 8.0$, (d) $t = 8.5$, (e) $t = 10.0$, (f) $t = 11.0$.

F. For $4.28 \leq \Delta t \leq 5.41$ the earlier signal passes unchanged through the junction, while the later signal initiates output impulses both within its own channel (area 2) and in the horizontal channel (area 6) (see Fig. 9).

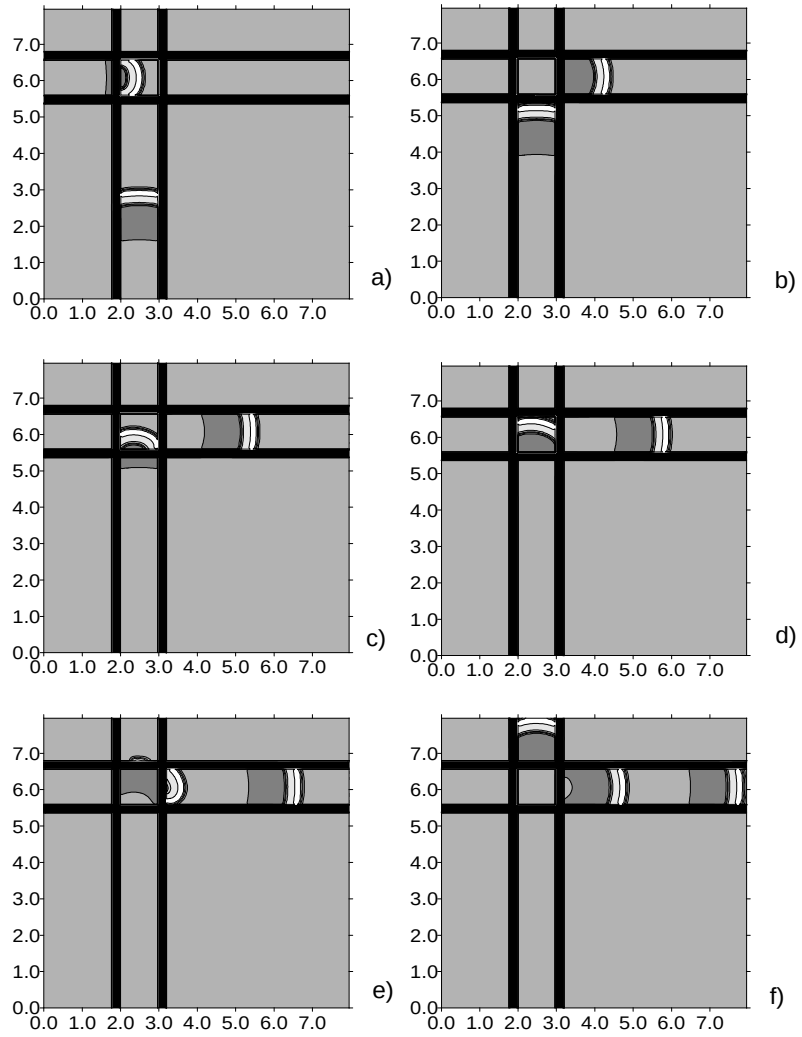

Fig. 9. For $\Delta t = 4.64$ the earlier signal goes unchanged through the junction, while the other signal initiates output impulses both within its own channel (area 2) and in the horizontal channel (area 6). The consecutive snapshots correspond to (a) $t = 4.0$, (b) $t = 7.0$, (c) $t = 8.5$, (d) $t = 9.0$, (e) $t = 10.0$, (f) $t = 11.5$.

G. For $\Delta t \geq 5.43$ the impulses do not "feel" each other any more. Each of them simply propagates in the channel it was initialized in (areas 6 and 2 respectively) (see Fig. 10).

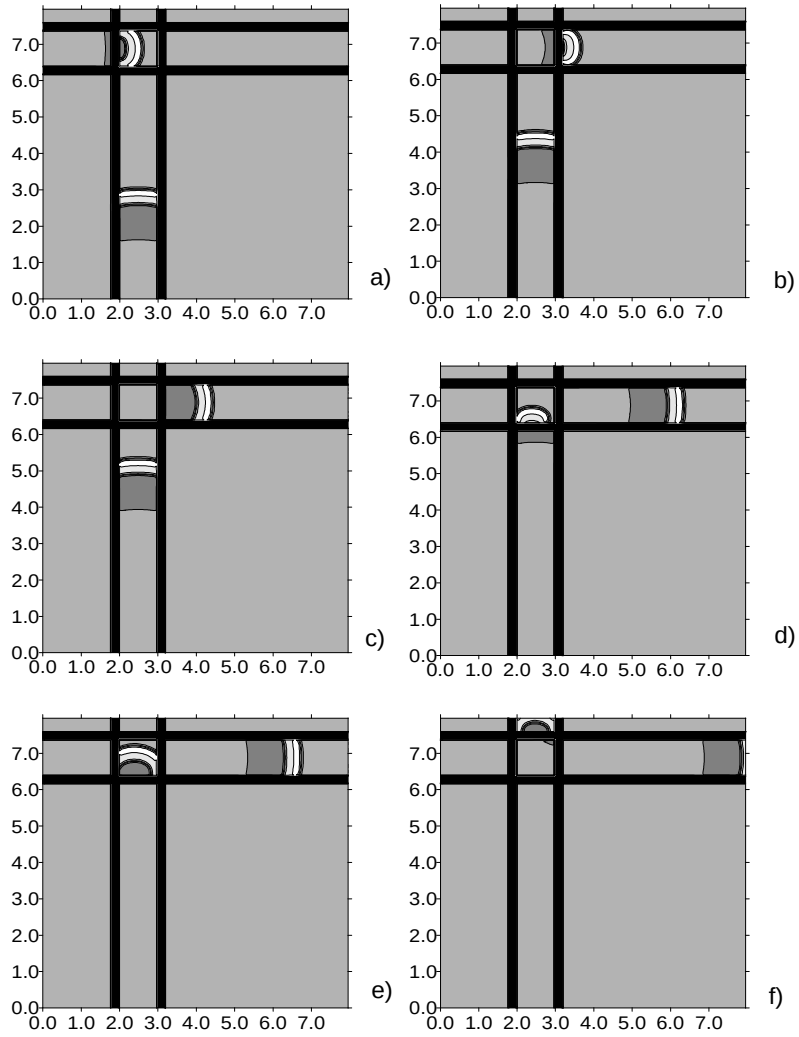

Fig. 10. For $\Delta t = 5.67$ the impulses do not "feel" each other any more. Each of them simply propagates in the channel it was initialized in (areas 6 and 2 respectively). The consecutive snapshots correspond to (a) $t = 4.0$, (b) $t = 6.0$, (c) $t = 7.0$, (d) $t = 9.5$, (e) $t = 10.0$, (f) $t = 12.0$.

4. Conclusions

We have applied the FitzHugh–Nagumo type model to study the information processing in excitable and diffusion media which form a cross junction with respect to signals coming from perpendicular directions. In our model information is associated with a traveling impulse. We have found that if

the second signal arrives later than 5.43 time units after the first one, its propagation is not disturbed by the one which comes earlier. On the other hand the first impulse "kills" the signals arriving before 4.22 time units. For the time difference between the signals Δt, $\Delta t \in [4.22, 5.43]$ the cross junction works as a logical switch; it redirects the later signal to the direction of the earlier one. We have also found that if the additional excitable fields around the original cross junction are considered (see areas 7 and 9 in Fig. 2) then the excitation of pulses in these fields brings more information about the signal coincidence. Therefore at the basis of FitzHugh–Nagumo model we have shown that a cross junction may work as a coincidence detector, a logical switch and also as a signal frequency multiplier if a signal is divided and redirected to the junction with the appropriate phase shift.

We believe that such a junction can be easily constructed experimentally. In the laboratory practice a chemical excitable medium is usually based on the Belousov–Zhabotinsky (BZ) reaction [11] with the appropriate choice of catalysts. Methods of obtaining the desired topology and geometry of the medium and procedures how to create and control traveling impulses in it are already well established [12,13]. In order to see what might be a spatio-temporal scale of a realistic cross junction we have considered the Rovinsky–Zhabotinsky model of BZ reaction [13–16] with the parameters given in [17]. The calculations have not been completed yet, but it seems that the size of the device is of the order of centimeters and the corresponding time unit — of the order of minutes. These values indicate that our chemical switch cannot compete in speed with the modern silicon devices, but nevertheless it may provide an interesting alternative for chemical information processing.

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REFERENCES

[1] P. Ball, *Nature* **406**, 118 (2000).

[2] L. Kuhnert, K.I. Agladze, V.I. Krinsky, *Nature* **337**, 244 (1989).

[3] O. Steinbock, P. Kettunen, K. Showalter, *J. Phys. Chem.* **100**, 18970 (1996).

[4] K. Yoshikawa, I. Motoike, K. Kajiya, *IEICE Trans. Electron.* **E80-C**, 931 (1997).

[5] K. Agladze, R.R. Aliev, T. Yamaguchi, K. Yoshikawa, *J. Phys. Chem.* **100**, 13895 (1996).

[6] A. Toth, K. Showalter, *J. Chem. Phys.* **103**, 2058 (1995).

[7] I. Motoike, K. Yoshikawa, *Phys. Rev.* **E59**, 5354 (1999).

[8] R. FitzHugh, *Biophys. J.* **1**, 445 (1961).

[9] J.S. Nagumo, S. Arimoto, Y. Yoshikawa, *Proc. IRE* **50**, 2061 (1962).

[10] B. Legawiec, private communication. See also B. Legawiec, D. Ziółkowski, *Inżynieria chemiczna i procesowa* **2**, 293 (1988) (in Polish).

[11] A. Zaikin, A.M. Zhabotinsky, *Nature* **225**, 535 (1970).

[12] A. Lazar, Z. Nosztczius, H.-D. Foersterling, Z. Nagy-Ungvarai, *Physica D* **84**, 112 (1995).

[13] A.B. Rovinsky, A.M. Zhabotinsky, *J. Phys. Chem.* **88**, 6081 (1984).

[14] A.B. Rovinsky, *J. Phys. Chem.* **90**, 217 (1986).

[15] T. Kusumi, T. Yamaguchi *et al.*, *Chem. Phys. Lett.* **271**, 355 (1997).

[16] R.J. Field, E. Körös, R. M. Noyes, *J. Am. Chem. Soc.* **94**, 8649 (1972).

[17] M. Frankowicz, A.L. Kawczyński, J. Górecki, *J. Phys. Chem.* **95**, 1265 (1991).