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# On architectures of circuits implemented in simulated Belousov–Zhabotinsky droplets

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ABSTRACT

When lipid vesicles filled with Belousov–Zhabotinsky (BZ) excitable chemical medium are packed in tight assembles, waves of excitation may travel between the vesicles. When several waves meet in a vesicle some fragments may deflect, others can annihilate or continue their travel undisturbed. By interpreting waves as Boolean values we can construct logical gates and assemble them in large circuits. In numerical modelling we show two architectures of one-bit half-adders implemented in BZ-vesicles.



## 1. Introduction

The design of logical gates in chemical systems can be traced back to the early 1990s when Hjemfelft et al. suggested a theoretical coupled mass flow system for implementing logic gates and finite-state machines (Hjelmfelt and Ross, 1995) and Lebender and Schneider proposed logical gates utilising a series of flow rate coupled continuous stirred tank reactors and a bistable chemical reaction (Lebender and Schneider, 1994). No experimental prototypes were implemented at that time. Mass-kinetic based computing is appealing theoretically but laboratory experiments are cumbersome to undertake. The implementation of mass-kinetic networks is inefficient as most designs require the use of programmable pumping devices. In 1994 the Showalter Laboratory presented the first ever experimental implementation of logical gates in the Belousov–Zhabotinsky (BZ) system (Tóth et al., 1994; Tóth and Showalter, 1995). The logical gates were based on the geometrical configuration of channels in which excitation waves propagate. The ratio between the channel diameter and the critical nucleation radii of the excitable media allowed various logical schemes to be realised. These original findings led to several innovative designs of computational devices, based on geometrically constrained excitable substrates. Designs incorporating assemblies of channels for excitation wave propagation were used to implement logical gates for Boolean and multiple-valued logic (Sielewiesiuk and Górecki, 2001; Motoike and Adamatzky, 2005; Górecki et al., 2009; Yoshikawa et al., 2009). All these chemical computing devices were realised in geometrically constrained media where excitation waves propagate along defined catalyst loaded channels or tubes filled with the BZ reagents. The waves perform computation by interacting at the junctions between the channels. Despite its apparent novelty the approach is just an implementation of conventional computing architectures in novel materials—namely excitable chemical systems. There is however another way to undertake computation—by employing the principles of collision-based computing (Adamatzky, 2003). Wave-fragments collide in a 'free' space and change their velocity vectors as a result of the collision. When input and output waves are interpreted as logical variables, the site of the waves' collision can be seen as a logical gate (Adamatzky et al., 2005). We explore the collision-based approach in the paper.

## 2. BZ-vesicles

A BZ-vesicle is a spherical compartment encapsulating BZ medium in an lipid membrane (NeuNeu., 2010; Szymanski et al., 2011; King et al., 2011); the BZ-vesicles can be arranged into a regular lattice. We physically imitate BZ-vesicles by projecting patterns

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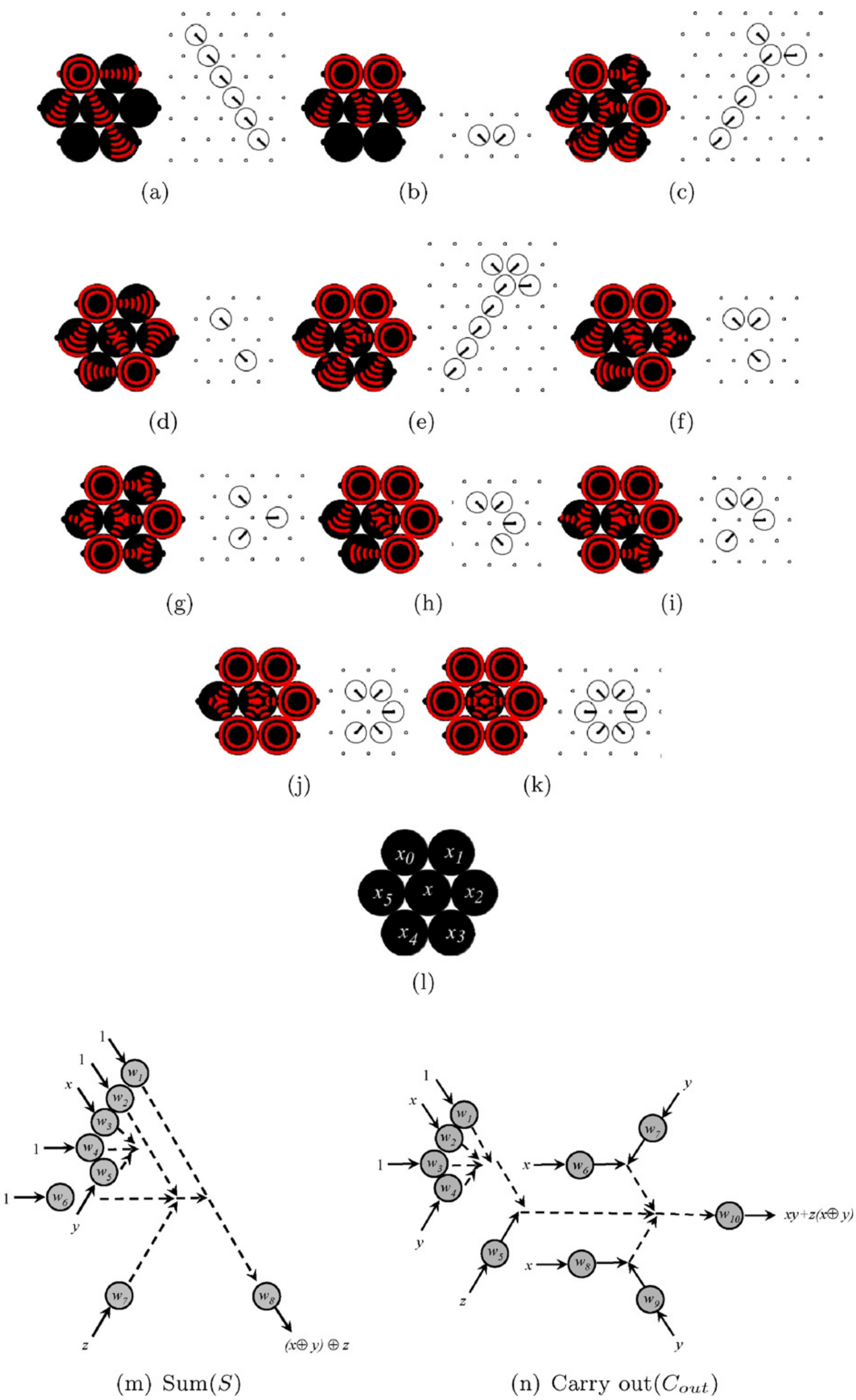


**Fig. 1.** Time lapse trajectories of wave interactions in BZ-vesicles and their use in one-bit half-adder design. Representative scenarios are given for (a) one input wave, (b)–(d) two input waves, (e)–(g) three input waves, (h)–(i) four input waves, (j) five input waves, and (k) six input waves. (a–k) Left image is wave patterns, right image is a schematic diagram. (l) Structure of disc-compartment neighbourhood of packed BZ-vesicles. (m) Sum and (n) carry out results of the adder.
From (Adamatzky et al., 2011)

and boundaries on a flat chemical reactor, and simulate the vesicles assembles in computer models.

When the BZ reaction is in a sub-excitable mode asymmetric perturbations lead to the formation of propagating localised excitation, or excitation wave-fragments. Wave-fragments of this type may travel in a predetermined direction for a finite period of time. If wave-fragments kept their shape indefinitely, we would be able to build a collision-based computing circuit of any size. In reality, the wave-fragments are inherently unstable: after some period of conserved-shape-distance travelled a wave-fragment either collapses or expands.

A way to overcome the problem of wave-fragment instability would be via the subdivision of the computing substrate into interconnected compartments, so called BZ-vesicles, and allowing

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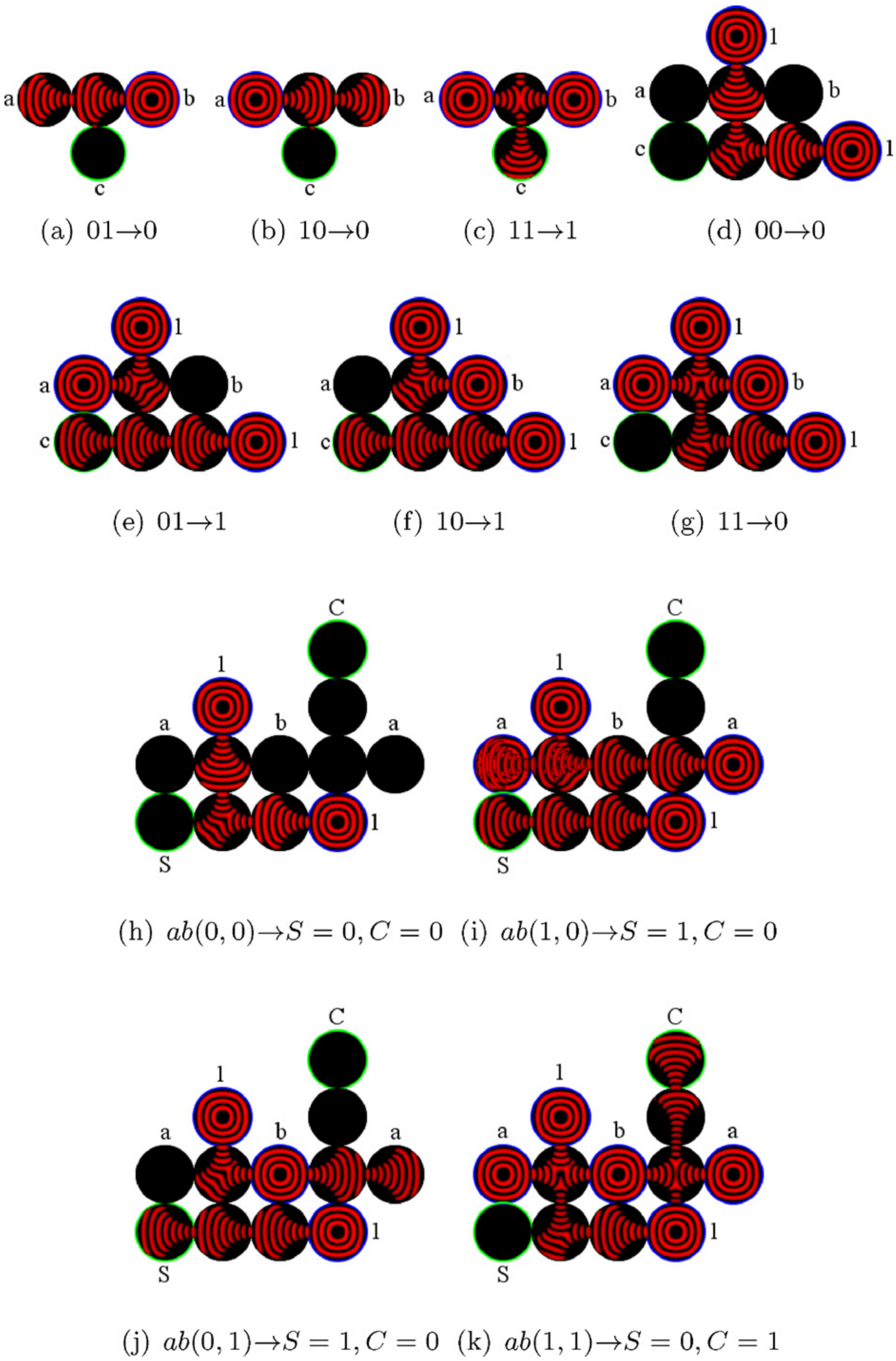


**Fig. 2.** Gates and circuits implemented with BZ-vesicles. (a–c) Two input AND gate ($c = a \cdot b$) where inputs $a$, $b$ are top left and right discs (blue rings) and output $c$ is the bottom central disc (green ring). (a) ($a$, $b$)(0, 1) A wave from input $b$ propagates uninterrupted and terminates in the opposing input disc $a$. (b) ($a$, $b$)(1, 0) Likewise, a wave from input $b$ propagates uninterrupted and terminates in the input disc $b$. (c) ($a$, $b$)(1, 1) Waves from both input discs $a$ and $b$ collide in the central disc and eject two perpendicular waves, one of which propagates into the output disc ($c$). (d–g) Two input XOR gate ($c = a \oplus b$) where inputs are middle row left and right (blue rings), the output disc is bottom left (green ring) and there are two source inputs, centre top and bottom right (blue rings). (h–j) Half adder circuit ($S = a \oplus b$, $C = a \cdot b$) where inputs are located along the central row, $a$ far left, $b$ central and then $a$ repeated far right (blue rings), two source inputs located top left and bottom right (blue ring). The output cell is bottom left (green ring). The circuit is constructed by combining an XOR gate and the AND gate (top right). The issue of the signal passing problem is obviated by replicating one of the inputs '$a$'.

waves to collide only inside the compartments (Adamatzky et al., 2011). Each BZ-vesicle has a membrane that is impassable for excitation. A pore, or a channel, between two vesicles is formed when two vesicles come into direct contact. The pore is small such that when a wave passes through the pore there is insufficient time for the wave to expand or collapse before interacting with other waves entering through adjacent pores, or sites of contact.

A spherical compartment – BZ-vesicle – is the best natural choice as it allows for effortless arrangement of the vesicles into a regular lattice, has an almost unlimited number of input/output states and also loosely conforms to a structure likely to be achieved in experiments involving the encapsulation of excitable chemical media in a lipid membrane. Scoping experiments with droplets of BZ-mixture covered with phospholipid L-$\alpha$-phosphatidylcholine provided evidence of excitation propagation between BZ-vesicles in direct physical contact (Szymanski et al., 2011).

Through the paper we use the two-variable Oregonator equation (Field and Noyes, 1974) adapted to a light-sensitive analogue of

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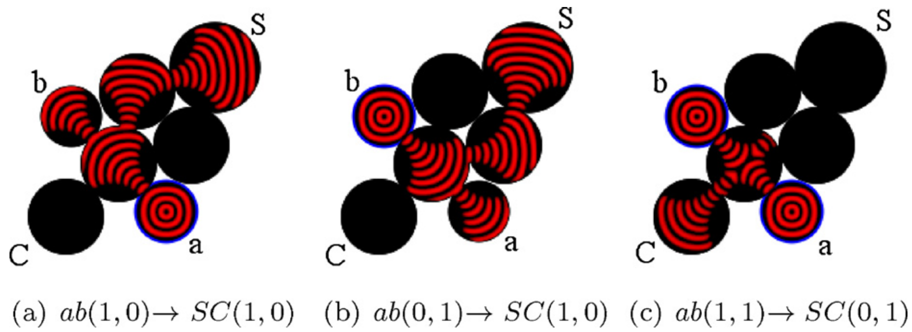


**Fig. 3.** Composite half adder circuit ($S = a \oplus b$, $C = a \cdot b$) where inputs and outputs are all connected to a central reactor disc which can achieve both the AND and XOR function. The two *half* outputs from the XOR operation are recombined with an OR operation with addition discs in the top right. The circuit employs 3 methods of signal modulation, connection angle, disc size and aperture efficacy.

the Belousov–Zhabotinsky (BZ) reaction with applied illumination (Beato and Engel, 2003):

$$\frac{\partial u}{\partial t} = \frac{1}{\epsilon}\left(u - u^2 - (fv + \phi)\frac{u-q}{u+q}\right) + D_u \nabla^2 u$$

$$\frac{\partial v}{\partial t} = u - v$$

The variables $u$ and $v$ represent the local concentrations of activator, or excitatory component, and inhibitor, or refractory component. Parameter $\epsilon$ sets up a ratio of time scale for the variables $u$ and $v$, $q$ is a scaling parameter dependent on the rates of activation/propagation and inhibition, $f$ is a stoichiometric factor. See detailed discussion of realistic parameters in Gorecki et al. (2011). Constant $\phi$ is the rate of inhibitor production. In the light-sensitive BZ $\phi$ represents the rate of inhibitor production which is proportional to the intensity of illumination. We integrate the system using the Euler method with five-node Laplace operator, time step $\Delta t = 0.005$ and grid point spacing $\Delta x = 0.25$, $\epsilon = 0.022$, $f = 1.4$, $q = 0.002$. The equations effectively map the space-time dynamics of excitation in the BZ medium and have proved to be an invaluable tool for studying the dynamics of collisions between travelling localised excitations in our previous work (Adamatzky, 2004; Adamatzky and De Lacy Costello, 2007; Toth et al., 2009; De Lacy Costello et al., 2009). We simulate a vesicle filled with BZ solution as a disc with radius $R$ centered in $(x_0, y_0)$. Sites inside the disc are excitable, sites outside the disc are not excitable. We imitate a wave-fragment entering the vesicle by exciting (assigning values $u = 1$) grid nodes inside the small disc with radius $r$, centered in $(x_0 + (R - s)\cos(\theta), y_0 + (R - s)\sin(\theta))$. The following parameters are used in the illustrations: $R = 100$, $r = 5$, $s = 5$, $\theta \in [0, 2\pi]$. Time lapse snapshots provided in the paper were recorded at every 150 time steps, and grid sites with excitation level $u > 0.04$ were displayed.

## 3. Binary Adder Made of Uniform BZ-vesicles

Let us consider how to build a one-bit half-adder using BZ-vesicles of the same size. BZ mixtures inside each vesicle is connected to the mixtures in neighbouring vesicles with the pores of the same size. The assembles of BZ-vesicles are fully synchronised.

Let several wave-fragments enter a vesicle. If at least two wave-fragments have opposite velocity vectors all wave-fragments annihilate. Otherwise the wave-fragments merge and the velocity vector of the newly formed wave-fragment is a sum of the velocity vectors of the incoming wave-fragments (Adamatzky et al., 2011). We illustrated this by a representative scenario of waves interactions inside a single vesicle (vesicle $x$ in Fig. 1l) shown in Fig. 1. If just one neighbour of vesicle $x$ is excited the vesicle $x$ acts as a conductor and signal amplifier: the wave simply passes through the vesicle $x$ slightly increasing in size and exits through the pore opposite to the wave's entry pore. Thus, in Fig. 1a north-west neighbour (vesicle $x_0$ in Fig. 1) is activated. Excitation enters vesicle $x$, wave-fragment travelling south-east is formed (Fig. 1a). The wave-fragment is transmitted to the south-east neighbour (vesicle $x_3$ in Fig. 1l) of vesicle $x$.

There are three possible scenarios when two neighbours of vesicle $x$ are excited. In a situation when excited neighbours of $x$ are also each others closest neighbours (e.g. vesicles $x_0$ and $x_1$ in Fig. 1l) the wave-fragments generated by them merge inside vesicle $x$ (Fig. 1b). The velocity vector of a newly formed wave-fragment is a sum of the vectors of the two original wave-fragments. Vectors of the original wave-fronts orientate towards the exit pores opposite to the excitation entry pores. The vector of a newly formed wave-fragment aims between the pores, hence no excitation leaves vesicle $x$. For example, in (Fig. 1b) north-west and north-east neighbours of vesicle $x$ are excited. Two wave-fragments enter vesicle $x$: one fragment travels south-east, another south-west. The wave-fragments merge and form a new wave-fragment which travels south. This fragment collides with the part of vesicle $x$'s wall lying between the south-west and south-east pores. The fragment is annihilated as a result of the collision. Other scenarios of wave-collisions are discussed in Adamatzky et al. (2011).

A binary adder based on BZ-vesicles uses collisions between propagating wave-fragments to perform addition of three one-bit binary numbers $x$, $y$ and $z$. The signal $z$ depicts $C_{in}$. There are many versions of particular implementations of a full one-bit adder. We adopted the most common one, where the adder outputs the sum of the signals $S = (x \oplus y) \oplus z$ (Fig. 1m) and carry out value $C_{out} = xy + z(x \oplus y)$ (Fig. 1n). Positions of inputs in Fig. 1mn are shown by thick solid circles. If an input equals FALSE the circle contains only central dot, if the input equals TRUE the circle contains a vector indicating the sites state. Results of the sum circuit are represented by wave-fragment travelling south-east whilst the output of the carry out circuit is represented by a wave-fragment travelling east. The circuit calculating $S$ is packed in array of $6 \times 10$ BZ-vesicles, and the circuit calculating $C_{out}$ occupies a sub-array of $7 \times 13$ BZ-vesicles. Let us discuss functioning of circuit $S$ (Fig. 1a) in detail.

If all three inputs – in the sum circuit – are FALSE no waves are initiated in sites $w_3$ and $w_5$ and $w_7$ (Fig. 1m). The wave-fragments representing constant TRUE are placed in sites $w_1$ and $w_2$ (south-east travelling wave-fragments), $w_4$ and $w_6$ (wave-fragments travelling east). Wave-fragment $w_2$ collides with wave-fragment $w_4$, both wave-fragments annihilate (step three of cellular automaton simulation, Fig. 1m). Wave-fragments $w_1$ and $w_6$ travel a bit further but collide with each other and annihilate during the fifth step of the automaton simulation. All outputs are FALSE therefore.

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If carry in value $z$ is TRUE and other input values are FALSE (Fig. 1m) then north-east travelling wave $w_7$ annihilates east travelling wave $w_6$. Therefore wave $w_1$ continues travelling south-east, thus representing TRUE value of $(x \oplus y) \oplus z$.

In situation $(x, y, z)$ = (FALSE, TRUE, FALSE) wave $w_5$ collides with wave $w_4$ (Fig. 1m), both wave-fragments annihilate. Therefore wave-fragment $w_2$ continues undisturbed on its trajectory to the south-east where it collides with wave-fragment $w_6$. With both waves representing east travelling constant TRUE cancelled wave-fragment $w_1$ reaches an output. Similarly for input $(x, y, z)$ = (TRUE, FALSE, FALSE) wave-fragments $w_3$ and $w_4$, and $w_2$ and $w_6$ annihilate in collision with each other (Fig. 1m). Therefore wave-fragment $w_1$ travels undisturbed. Other combinations of inputs can be considered in a similar manner.

In the carry out circuit the computation starts in a group of sites marked $w_1, \ldots, w_4$ in (Fig. 1n): site $w_2$ represents $x$, site $w_4$ represents $y$, and sites $w_1$ and $w_3$ represent constant TRUE. The sub-circuits' output is $x \oplus y$. Wave-fragments $w_1$, travelling south-east, and $w_3$, travelling east, are always present in the system. If only one of the inputs $x$ or $y$ has TRUE value, e.g. south-east travelling wave-front $w_2$ in Fig. 1n), then wave-fragment representing this input collides with wave-fragment $w_3$ (constant TRUE) and both wave-fragments annihilate. If both inputs $x$ and $y$ are TRUE (Fig. 1n) then waves $w_2$ and $w_4$ collide with each other and wave $w_3$, merge together and produce a new wave travelling east. This new wave collides with wave $w_1$, and both wave-fragments annihilate. Thus, sub-circuit $(w_1, \ldots, w_4)$ computes $x \oplus y$. Output wave-fragment of sub-circuit $x \oplus y$ collides with wave-fragment $w_5$, which represents carry in value $z$. These wave-fragments collide at angle 120°, therefore they merge and produce new wave-fragment (travelling east) when they collide. Thus an intermediate result $z(x \oplus y)$ is calculated.

Two small sub-circuits – $w_6$ and $w_7$, and $w_8$ and $w_9$ – are arranged symmetrically north and south of the trajectory of the wave-fragment, which represents $z(x \oplus y)$. Each of the sub-circuits $(w_6, w_7)$ and $(w_8, w_9)$ produces $xy$. The final wave-fragment $w_10$ is only produced when either only wave-fragment $z(x \oplus y)$ travels east (Fig. 1n), or all three wave-fragments—wave-fragment $xy$ (output of sub-circuits $(w_6, w_7)$) travelling south-east, wave-fragment $xy$ (output of sub-circuits $(w_8, w_9)$) travelling north-east, and wave-fragment $z(x \oplus y)$ all collide (Fig. 1n).

## 4. Elaborate Arrangements of BZ Discs

In the previous section we have shown how arithmetical circuits can be implemented in a regular arrangements of BZ vesicles, as if they were packed in a hexagonal lattice. Let us assume now that we can manipulate BZ vesicles and are capable for arranging them in the irregular structures. Fig. 2 illustrates how AND and XOR gates can be created using interconnected BZ discs. In the AND gate the collision between the two inputs results in two perpendicular fragments, one of which develops in the output cell to produce the result (Fig. 2a–c). The XOR gate is used to detect a difference between signals, producing an output when inputs alternate regardless of the composition of the difference (Fig. 2d–g) (Holley et al., 2011).

A one bit half adder created from BZ discs can also be constructed from connecting a BZ disc AND gate and XOR gate. Fig. 2h–j shows the BZ disc conjunction for the half adder circuit. The input $a$ needs to be repeated on the other side of input $b$ in order for this circuit to work. This is necessary in order to overcome the *signal passing problem*, a universal problem for systems where signals propagate along specific planar channels. There are two ways to overcome this problem, either add identity to the signals in such a way that signals can share the medium or, share the medium at different times. How two or more waves could be identified and share the same space in this BZ system remains unclear because of the diffusive nature of the reaction. However sharing a channel medium in time is possible if the time difference between signals is large enough to prevent the refractory tail from one extinguishing the other. In this instance of the 1 bit HA an alternative solution would be to avoid the signal passing problem by combining both the AND and XOR function into one poly-functional vesicle (Holley et al., 2011).

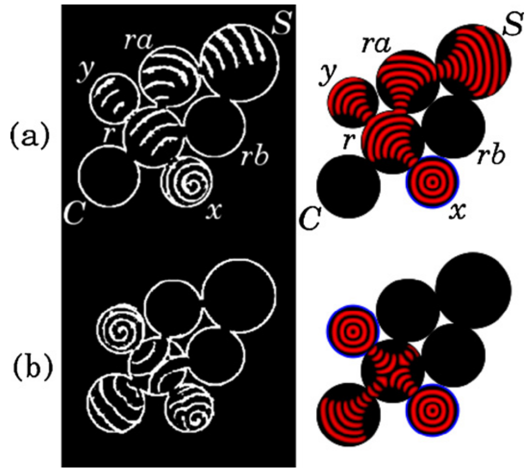


**Fig. 4.** Experimental chemical implementation of the one bit half adder circuit ($S = x \oplus y$, $C = x \cdot y$). Left column (inverse graphics) represents experimental analogue of the simulation in the right column. The central reactor disc perform both XOR and AND logic functions to create the desired logic. (a) XOR component, $S$ = 1, $C$ = 0, $(x, y)$ = [0, 1]||[1, 0]. (b) AND component, $S$ = 0, $C$ = 1, $(x, y)$ = [1, 1].

Fig. 3 demonstrates a half adder design where most of the processing occurs in one central reactor disc. The central disc achieves the AND function (Fig. 3c) and the XOR function (Fig. 3b and c). Considering the central disc principally in terms of an AND gate; then the XOR function can be derived from the AND gate response to input sets $(a, b)(0, 1)$ and $(a, b)(1, 0)$. The outputs of which are curved around into an OR gate in the $S$ output disc creating the XOR function.

Our disc designs have not been confined simulation. Several of our circuits have successfully migrated to experimental implementations on a photo sensitive BZ system with a catalyst loaded gel. Fig. 4 illustrates our experimental implementation of the HA circuit derived from simulated HA design (Fig. 3). Chemical BZ waves (left column, inverse graphics) are captured reacting in parallel to the simulation (right column). Although the parameterisation of the simulation creates different wave dynamics the projected geometry in each case is proportionally identical resulting in the same functionality (Holley et al., 2011). See full details of experimental implementation, including protocol for the experiments, in Holley et al. (2011).

## 5. Conclusion

In the Belousov–Zhabotinsky (BZ) medium in a sub-excitable state localised travelling excitation waves are formed. We interpret these localisations as quanta of information, values of logical variables. When two or more localisations collide they annihilate or form a new localisations. We interpret post-collision trajectories of the localisations as the results of a computation. We demonstrate that by colliding wave-fragments in an encapsulated excitable chemical medium we can realise a number of logical gates. Implementation of universal logical gates is a noble task however it is just a first step towards designing general purpose computers based on assemblies of BZ-vesicles. Assembling gates into a logical or arithmetical circuit is a paramount task, particularly when signals are transmitted via such sensitive and intrinsically unstable dissipative localisations. We categorised all interactions between wave-fragments in the idealised BZ-vesicles and designed a full

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one-bit binary adder in an array of tightly packed vesicles. We also demonstrated by directly manipulating the BZ-vesicles we can arrange in size-optimal logical circuits with duplicated inputs.

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