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Experimental implementation of collision-based gates in Belousov–Zhabotinsky medium

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10 Abstract

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We experimentally demonstrate that excitation wave-fragments in a Belousov–Zhabotinsky (BZ) medium with immobilised catalyst can be used to build elementary logical gates and circuits. Following our previous theoretical constructions [Adamatzky A. Collision-based computing in Belousov Zhabotinsky medium. Chaos, Solitons & Fractals 2004;21:1259–4] on embedding logical schemes in BZ medium, we represent True/False values of logical variables by presence/absence of wave-fragments. We show that when wave-fragments collide with each other they may annihilate, fuse, split and change their velocity vectors. Thus the values of logical variables represented by the wave-fragments change and certain logical operations are implemented. In the paper we provide examples of experimental logical gates, and present pioneer results in dynamic, architectureless computing in excitable reaction–diffusion systems.

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21 1. Introduction

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22 Belousov-Zhabotinsky (BZ) [27] chemical system—a typical example of excitable chemical media—is proved to be 23 capable for specialised computation. The medium belongs to a class of reaction-diffusion processors, namely to its subclass of reusable processors. A reaction-diffusion processor (RD) is a real chemical medium, typically a thin-layer solu-25 tion, gel or film, which transforms data to results in a predictable, sensible and pre-programmable way. In RD proces-26 sors data are represented by perturbances of medium's characteristics, e.g. inhomogeneous concentration profile of 27 reagents, possibly induced by non-uniform illumination. Excitation waves—in reusable processors, or diffusive 28 waves—in disposable processors, travel from the disturbances, interact with each other, and produce either stationary, precipitate concentration profile (disposable processors), or a dynamic, oscillatory field, or a dissipative structure (reusable processors). The final state of the medium's spatial dynamics represent a result of the RD computation. Recently experimental prototypes of RD processors were applied to solve a wide range of computational problems, including image processing [20,2], path planning [24,10,19,4], robot navigation [5,8], computational geometry [6], counting 33 [15], memory units [17].

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19 January 2005 Disk Used

2 B. De Lacy Costello, A. Adamatzky / Chaos, Solitons and Fractals xxx (2005) xxx-xxx

A number of logical gates were implemented in excitable chemical systems [26,23] and precipitating systems [3].

5 2. Dynamical versus structural computation

Thus far all known experimental and simulated prototypes of RD processors implementing logical computation [25,26,23,3,18] mimic a conventional hardware implementation of logical circuits, with their wires and gates fixed forever. These RD processors exploit the interaction of wave fronts in a geometrically constrained chemical medium, i.e. the computation is based on a stationary architecture of the medium's inhomogeneities. Constrained by stationary wires and gates RD chemical universal processors give us little computational novelty and no dynamical reconfiguration ability because they simply offer a new material base for old computing architectures.

To free RD processors from the limitations of fixed computing architectures we adopt an unconventional paradigm of dynamical, architecture-less, or collision-based, computing [7]. The paradigm originates from computational universality of Game of Life [11], conservative logic and billiard-ball model [14] and their cellular-automaton implementations [16].

A collision-based (CB) computation employs mobile compact patterns, in our particular case they are self-localized excitations in active non-linear medium. The localizations travel in space and perform computation (implement logical gates) when they collide with each other. There are no predetermined stationary wires—a trajectory of the travelling pattern is a momentary wire—almost any part of the medium's space can be used as a wire. Truth values of logical variables are given by either absence or presence of a localization or by various types of localizations [7].

Would it be possible to realise collision-based computation schemes in RD medium? A medium is capable of collision-based computing if it exhibits mobile self-localisations in its space–time evolution. Until very recently we had little if any information about the interaction of mobile localizations in two- or three-dimensional RD media. However the works [22,12] demonstrated the existence and rich interaction of dissipative solitons in a three-component RD system. The basis for dynamical universality of RD chemical media was finally cemented when the existence of localized excitations—travelling wave-fragments which behave like quasi-particles—in photosensitive sub-excitable BZ medium was experimentally proved [21]; however, the phenomenology of localized traveling wave-fragments have never been considered in a context of novel computing architectures.

In the present paper we give experimental evidence of how logical circuits are embedded in a sub-excitable BZ medium via collisions between travelling wave-fragments.

61 3. Experimental rationale

The formation and control (modulation) of small BZ fragments has previously been undertaken in a modulated light sensitive BZ reaction [21]. This medium was considered to be sub-excitable and the direction and growth of small wavefragments could be controlled by carefully controlling a projected light intensity.

From a computational point of view these results are interesting as they provide a method for implementing collision-based logic gates and theoretical papers have shown some examples of these gates based on this system [9]. However, from an experimental point of view it poses some problems, whilst these are not insurmountable it is a fact that a high degree of control and auxillary equipment is required even to control the modulation of a single wave-fragment and this will be further complicated if two or more fragments are to be collided. There are two other foreseen difficulties with this methodology namely does the modulation of light which has to be so carefully controlled to maintain the fragment in a semi-stable state correspond to a "pseudo wired" architecture? Also if a collision was implemented in this system what light modulation would be applied (and at what point) to the resulting fragment and would this therefore compromise the computational results? If no modulation was applied presumably the resulting fragment would expand and dominate the reactor space or conversely die. There is one major advantage that this system would possess over the one described in the following sections and that is some degree of inherent controllability. However, some possible limitations of this system have been highlighted above.

We devised an alternative system whereby small semi-stable wave-fragments were readily produced in a BZ type reaction over a long period and readily underwent collisions with other fragments, often multiple sequences of collisions. Thus the experimental system we will describe serves as a proof of principle prototype for assessing collision-based computations. The control aspect is not considered in detail as we do not aim to provide an architectureless system capable of directed computation—however, obviously there is some degree of control factored into the experimental design allowing fragments to exist per se but never to dominate the reactor space. Our aim is just to attribute computational behaviour to the dynamics and interactions of wave-fragments in a truly architectureless BZ medium.

4. Experimental design

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The reactor used during the experiments is based on a ferroin catalysed analogue of the BZ reaction. A 2.5 cm by 2 cm piece of photographic paper (Kodak Ultra ISO 400) was cut and coated with 0.25 ml of a solution of 1,10-phe-86 nanthroline iron(II) sulphate complex (ferroin 0.025 M, Sigma Chemical Co.). Any excess was removed and the evenly 87 coated film left to dry in ambient conditions for 10 min. The piece of photographic paper was then added to a petri dish 88 and 2 ml of a BZ stock solution added to the top surface. The stock solution was prepared as follows and was based on 90 a recipe by [13]. An acidic bromate stock solution incorporating potassium bromate (KBrO₃) and sulphuric acid $([BrO_3^-] = 0.5 \text{ M})$ and $[H^+] = 0.59 \text{ M})$ was prepared (solution A) and stock solutions of Malonic acid 92 $[CH_2(CO_2H)_2] = 0.5 \text{ M}$ (solution B) and sodium bromide [Br] = 0.97 M (solution C) were prepared. The following 93 quantities of the stock solutions were mixed 3.5 ml solution A, 1.75 ml solution B and 0.6 ml solution C. The solution 94 was stirred until it turned from brown to colourless prior to use in the experiments. The piece of photographic paper 95 was immersed in the BZ solution for circa 5 min in which time the back surface of the photographic paper had turned from a dark brown to a deep orange colour. At this stage the photographic paper was placed between two clean glass 97 microscope slides. Some additional BZ stock solution was introduced between the slides using a pipette and the assembly was sealed using a clip taken from a gel-electrophoresis assembly (Atto corporation). The whole assembly was then viewed under a binocular microscope (Prior-James Swift) at a low magnification of ×10. Pictures were recorded using a 100 Fujifilm 2600z digital camera (zoom lens f = 6-18 mm equivalent to f = 38-114 on a 35 mm camera) using the built in macro function with a resolution of 640×480 pixels and using a transmissive lighting method. No filters were used.

102 5. Experimental results

The waves could be observed from either side of the reactor indicating that they were traveling in the bulk material (emulsion) of the photographic paper. The affect on the chemistry of the BZ reaction of using photographic paper is likely to be complex when compared to the use of conventional support substrates such as agarose or silica gels and is outside the scope of this current paper. The photographic paper will initially contain inhibitors to the reaction in the form of silver halides (chloride or bromide ions)—there will be some conversion to an activator species silver with light exposure (experiments carried out in ambient light) although this reaction is limited and reversible under acidic conditions.

The fact that wave-fragments and not the classical target waves and spirals are observed suggests that the chemical environment is inhibitory and the medium is highly constrained. The time for small traveling wave-fragments to form varied but was after approximately 15 min and the wave formations could be observed for over 2 h continuously. The average velocity of the waves was circa 1 mm/min although this increased at the point just prior to collision. It was also highly dependant on the curvature of the fragments. The number of waves or wave-fragments on average within the reactor space increased with reaction time as expected, however, at no stage did the wave-fragments dominate the reactor space as would be observed for most conventional BZ systems especially with no external modulation.

Wave fragments observed ranged from 0.4 mm in length (such as the fragment termed the "BZ bullet"—see Fig. 1) to several millimeters, however, large fragments traveling in the medium were short lived and subject to spontaneous splitting thereby forming two or more smaller traveling fragments. Small traveling fragments were also subject to extinction prior to any collision, e.g. see Fig. 1 where the straight fragment on the left hand side disappears prior to any collision with the convex fragment. Experimental observations suggest that this effect is more likely (but not solely) if the fragments are approaching a collision point.

Observations also indicate that waves traveling on the same trajectories as previous wave-fragments (within a certain time interval) are more likely to undergo extinction or spontaneous splitting. This effect can also be seen in the wake of collisions where a daughter fragment will diminish if it travels back over the path of one of the parent fragments. This is strongly indicative that the BZ reaction run in this set up has a long refractory period. The reaction has been carried out in this format on many occasions and apart from a high proportion and large variety of travelling wave-fragments it is difficult to assign any typical behaviour. The examples covered in this paper aim to give a representative sample of the types of collisions between these fragments and some computational significance. However, due to the high volume of information collected it is not possible to cover all aspects of the behaviour.

Therefore, the following section will set out some of the noteworthy features of the reaction and some typical and atypical collision types, some of which are covered in the computational schemes but others that are not.

Perhaps most noteworthy is that as mentioned a number of small sub-millimeter wave-fragments were stable (did not expand or diminish appreciably in size) over distances of many millimeteres and could therefore be observed undergoing collisions. If a specific example of the "BZ bullet" (Fig. 1) is taken its size varied between 0.3 and 0.4 mm and it

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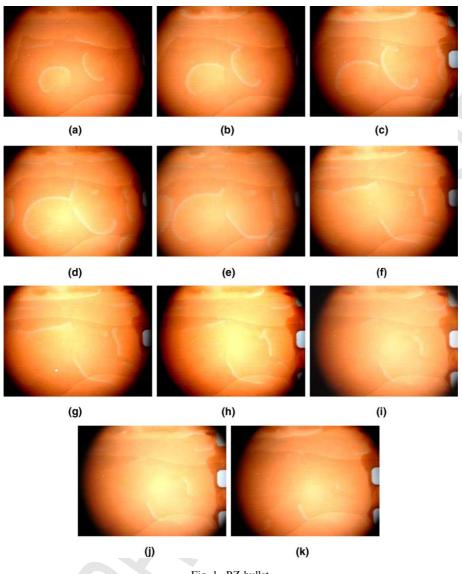


Fig. 1. BZ bullet.

136 was stable for many minutes travelling a distance of 0.4 cm at which point it underwent a collision with a larger fragment. The "BZ bullet" was originally formed from a collision between a straight fragment travelling on a north-east to south-west trajectory and a convex fragment expanding along its north-west axis the leading tips of both fragments meet and a small fragment results travelling on north-west to south-east trajectory.

The type of wave-fragments observed varied greatly with the dominant type being a convex type wave comprising two counter rotating spiral tips, waves of this type were the only fragments observed to grow significantly in the reactor space. Obviously a large number of the more unusual shaped fragments e.g. non-symmetrical, concave and small stable fragments in the reactor space were the result of previous collisions or spontaneous splitting of larger fragments. A typical collision involving these convex fragments when expanding on directly opposite trajectories (convex to convex collision) results in two small daughter fragments travelling in opposite directions at 90° angles to the original trajectories of the parent fragments (see Fig. 3).

Fig. 1 shows an alternative scenario where fragments of this type collide when not on opposite trajectories resulting in larger fused fragments whose trajectory is dictated by the surviving spiral tip.

Another scenario is when the fragments expand and collide when moving in the same direction resulting in a small daughter fragment travelling in the opposite direction to the original parent fragments and a larger fused concave frag-

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B. De Lacy Costello, A. Adamatzky / Chaos, Solitons and Fractals xxx (2005) xxx-xxx

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151 ment moving in the same direction as the parents. If they collide whilst moving on opposite diagonals (e.g north-west to 152 south-east and south-west to north-east) and are size matched then no daughter fragments may be produced.

Another typical collision is that of a convex fragment and a concave fragment (result of previous collision), this con-154 vex to concave collision (if at the point of max curvature and fragments are size matched) results in two "v" shaped daughter fragments which undergo "self-annihilation". This term "self-annihilation" is used where fragments formed due to collisions are very short lived as they contain integral parts which continue to move on opposite trajectories or have resultant parts which are directly opposed to other parts of a larger fragment (a form of self-collision). It must be noted that size matched collisions are not the norm in this case and the concave fragments tend not to be symmetrical thus convex to concave head on collisions in this case typically result in one travelling daughter fragment and one "v" shaped short lived fragment. There were also a number of linear fragments that typically either split or diminished in size as they travelled in the reactor space. Typical collisions observed for this type of fragment were usually with convex fragments. However, on occasion two size matched straight fragments will collide and annihilate completely.

Fig. 4 shows a collision of a linear fragment with a larger concave fragment resulting in two daughter fragments moving in the same direction as the larger parent fragment but moving on opposite trajectories away from the point of collision. If the larger fragment had also been straight the distance between the daughter fragments would have remained constant. The experimental observations are consistent with a typical concave fragment being formed via a convex-convex collision and the retained counter rotating spiral tips altering the trajectory of the daughter fragments when involved in the subsequent collision.

Fig. 6 shows a straight fragment colliding almost end on to a convex fragment—the result is a fusing and self-annihilation of one spiral tip which causes a shift in the trajectory of the larger parent fragment as the remaining spiral tip dominates the final trajectory of the daughter fragment. At some stage during the reaction a convex fragment may collide (convex face on) with the edge of the reactor, the result is two daughter fragments moving in opposite directions along the edge of the reactor.

As mentioned at some stages during the reaction fragments may spontaneously split—the most marked occasion is when a convex fragment splits into two daughter fragments moving in opposite directions to each other. This is almost identical to the case where two convex fragments collide convex face on (Fig. 3). It is assumed that this spontaneous splitting is due to the depletion of the activator species in the vicinity of the travelling front. This can occur when a fragment travels along a previously travelled trajectory within or close to the refractory period. It may also occur when two fragments converge suggesting competition between the two fronts for the activator species. In this reactor the inhibitory nature of the gelatin-silver halide emulsion must be considered a factor in producing a sub-excitable environment. However, another remote possibility is that local impurities in the reactor may act as a temporary barrier to diffusion. This possibility is dealt with and expanded in the next section. Perhaps more likely is that diffusion processes are extremely limited in the reactor set up described per se.

All of the aforementioned collisions and observed behaviours can be assigned some computational significance (see following sections). The experimental results fit well with a theoretical framework for collision-based computing devised in [9].

187 6. Examples of experimental gates

To describe interaction gates realized in collisions of wave-fragments we adopt formalism of [1]: 188 $\langle a_1, \dots, a_k \rangle \to \langle b_1, \dots, b_m \rangle$ as a transformation of a set of logical values of k input trajectories to a set of logical values 189 190 of m output trajectories.

The gate $\langle x, y \rangle \to \langle x \text{ AND } y, x \text{ AND NOT } y, \text{ NOT } x \text{ AND } y \rangle$ (Fig. 2) is the most common gate implemented in almost any non-linear medium with mobile self-localisations.

When two convex wave-fragments approach each other (Fig. 3(a)-(c)), they collide (Fig. 3(d)), and the convex faces of the wave-fragments merge and annihilate (Fig. 3(e) and (f); see wave-interaction scheme in Fig. 3(k)). However, the respective spiral tips of the fragments form new wave-fragments, these daughter fragments are initially concave (Fig. 3(f) and (g)), then become convex (Fig. 3(h) and (j)) and continue travelling along new trajectories (Fig. 3(f)). When one of the wave-fragments is not present, another fragment will travel along its original trajectory. So, assuming that presence of a wave-fragment along trajectory x is representative of logical TRUTH and the absence of the fragment FALSE, then unchanged trajectory of each fragment represents operation x AND NOT y and NOT x AND y, respectively. Trajectories of daughter wave-fragments represent operation x AND y (Fig. 2). This is analogous to famous Fredkin– Toffoli interaction gate [14] however the BZ gate just simulates but does not support conservativeness.

The gate has $\langle x, y \rangle \to \langle x \text{ AND } y, x \text{ AND NOT } y, \text{ NOT } x \text{ AND } y \rangle$ two output trajectories which represent values of the 203 same logical variable x AND y, so the gate can be used in signal splitting.

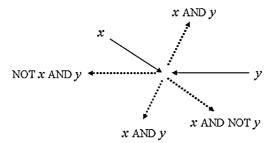


Fig. 2. Gate $\langle x, y \rangle \to \langle x \text{ and } y, x \text{ and not } y, \text{ not } x \text{ and } y \rangle$.

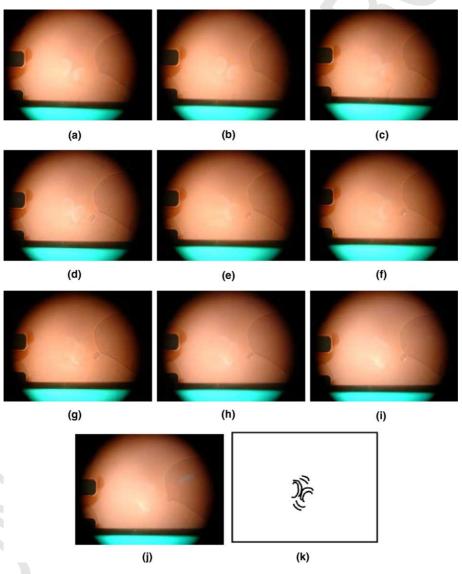


Fig. 3. Snapshots of experimental implementation of collision gate $\langle x, y \rangle \rightarrow \langle x \text{ AND } y, x \text{ AND NOT } y, \text{ NOT } x \text{ AND } y \rangle$ (see Fig. 2) in BZ medium, x = Truth, y = Truth. (a)–(j) snapshots of excitation dynamics, (k) scheme of wave-fragments collision.

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There is one more scenario of wave-fragment interaction—they just merge in one wave-fragment when they collide 205 with each other (Fig. 4(a)–(e)); see scheme of collision at Fig. 4(i).

In this case the colliding wave-fragments implement gate $\langle x, y \rangle \to \langle x \text{ AND } y, x \text{ AND NOT } y, \text{ NOT } x \text{ AND } y \rangle$ with two input and three output trajectories (Fig. 5, output trajectory labelled x AND y in the rightmost part of the gate).

A wave-fragment can be split onto two independent wave-fragments when a smaller wave-fragment collides with it (Fig. 4(f) and (h)). Thus, assuming a smaller wave-fragment (as that travelling south-west in Fig. 4), represents a constant TRUTH, and the bigger wave-fragment, x, we get a splitting of a signal x into two signals x. Via the combination, merging and splitting of wave-fragments it is apparent that almost any kind of logical circuit can be implemented.

An example of cascaded gates is shown in Fig. 5: $\langle x, y \rangle \rightarrow \langle x \text{ AND } y, \text{ NOT } x \text{ AND NOT } y, \text{ NOT } x \text{ OR NOT } y \rangle$ 213 the gate has three input trajectories, for x and y, and constant truth, and five output trajectories: two x AND y, one NOT x or not y, one x and not y, one not x and y. Space-time dynamics of the gate for x = Truth, y = Truth is shown 215 in Fig. 4.

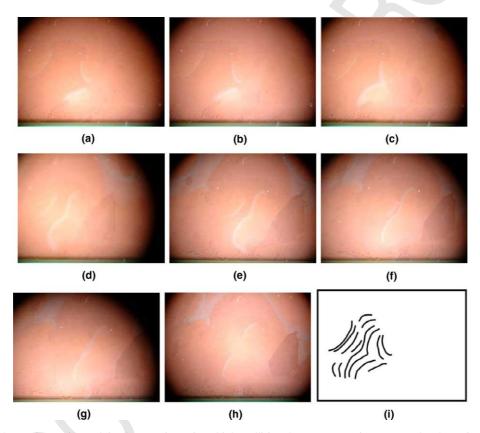


Fig. 4. Snapshots of experimental implementation of multiple collision between wave-fragments. The dynamics realized gate $\langle x, y \rangle \rightarrow \langle x \text{ and } y, \text{ not } x \text{ and } y, x \text{ and not } y, \text{ not } x \text{ or not } y \rangle$, shown in Fig. 5.

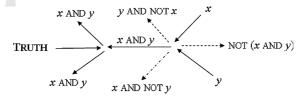


Fig. 5. Gate $\langle x, y \rangle \to \langle x \text{ and } y, \text{ not } x \text{ and } y, x \text{ and not } y, \text{ not } x \text{ or not } y \rangle$.

B. De Lacy Costello, A. Adamatzky / Chaos, Solitons and Fractals xxx (2005) xxx-xxx

Trajectories of wave-fragments are changed during collisions, so we can implement reflection (deviation of the signal trajectory) of one signal by colliding another signal into it, as show in Fig. 6. Sites of the collision correspond to positions of "momentary" reflectors.

In certain conditions two colliding wave-fragments do not change their velocity vectors but continue their travel along original trajectories and generate travelling daughter wave-fragments, as shown in Fig. 1 which may undergo subsequent collisions with other wave-fragments. An example of possible computational scheme derived from dynamics of wave-fragments in Fig. 1 is shown in Fig. 7.

There is also an option to incorporate stationary reflectors of signals in the BZ medium—the reflectors are represented by impurities. As discussed above waves are subject to spontaneous splitting in the devised reactor there are many possible reasons outlined and one includes the presence of impurities in the reactor. Presently if these impurities do act as a barrier to diffusion it is a natural and uncontrollable phenomena. However, we discussed cases whereby spontaneous splitting of certain parent fragments resulted in daughter fragments identical to those produced in collision-based gates. Therefore, as mentioned it may be possible to incorporate impurities into the BZ reactor that would cause this splitting or bring about some deviation in the trajectory (reflection) of the travelling wave-fragments. Ideally, the positioning of such impurities could be controlled during the lifetime of the reaction in order to bring about directed behaviour. This may be via the application of external fields and ideally would be a reversible and dynamic process. Another option would be to use high local light intensities—then rather than manipulating the fragments motion con-

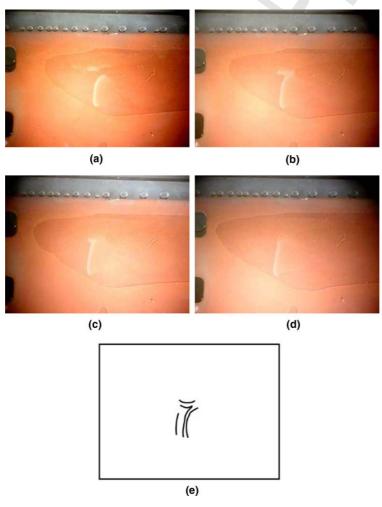


Fig. 6. Experimental implementation of signal's reflection. Trajectory of a wave-fragment representing a logical variable x is changed by colliding a control wave-fragment to the wave-fragment representing x. (a)–(d) snapshots of excitation dynamics, (e) scheme of wave-fragment collisions.

Fig. 7. A computational scheme derived from dynamics of wave-fragments shown in Fig. 1.

233 tinuously it may be possible to control it transiently to induce certain types of collision or act as a reflector etc. Work is 234 continuing in the area of controllability.

235 7. Discussion

236 We provided experimental evidence of collision-based computation in a BZ medium with immobilised ferroin cat-237 alyst. We demonstrated that under certain conditions compact wave-fragments develop in the medium. The wave-fragments travel in the medium and implement logical gates when they collide with each other. We have shown how to 238 embed non-trivial logical circuits in experimental BZ medium. The medium realises an architectureless computation 239 240 because there are no predetermined stationary wires, a trajectory of the travelling wave-fragment is a momentary wire: almost any part of the medium's space can be used as wire at some stage of the computation. To realise a Boolean log-241 242 ical gate we represent a logical TRUTH by presence of a wave-fragment and logical FALSE by absence of the wave-frag-243 ment. When two or more wave-fragments collide, they may fuse, annihilate, generate new wave-fragments or at least 244 change their trajectories or velocity vectors. Thus Boolean variables represented by the fragments are changed and the 245 computation is implemented. A spectrum of logical gates realised are similar to that implemented in numerical models 246 of soliton-based computation in bulk medium (see [7]), the key point is that we provided experimental verification of 247

The experimental setup described in the paper is particularly encouraging because the reaction can continue for over two hours, the chemical reactor never becomes overloaded with wave-fragments because conditions do not favour wave growth, the reactor resides in a reduced steady state with only minimal wave-fragments populating the space. This gives us an opportunity to employ temporal cascading of complicated logical circuits.

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19 January 2005 Disk Used

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294

B. De Lacy Costello, A. Adamatzky | Chaos, Solitons and Fractals xxx (2005) xxx-xxx

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