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Non-Steady Behaviour of a Spiral under a Constant Current.

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Abstract. – We present an experimental study of the motion of a rotating spiral in the Belousov-Zhabotinsky reaction in an «almost open» reactor, under the influence of a constant external current. We provide a simple model based on symmetries to explain the previous experimental observations, but show that it is unable to describe the true motion of a spiral. The motion of a spiral cannot be reduced to that of its centre: global deformation modes must also be taken into account.

A rotating spiral wave is a robust two-dimensional structure observed in many non-linear pattern-forming systems, and particularly in excitable media [1, 2]. Such systems include nerve impulse propagation [3], cardiac muscle [4], the oxidation of CO on platinum [5], and reacting chemical systems like the Belousov-Zhabotinsky (BZ) reaction [6, 7]. In particular, it is generally believed that spirals in cardiac muscle play an essential role in heart diseases such as arrhythmia and fibrillation [8]. The stability and geometric shape of spiral waves are remarkable: in many cases, they are nearly Archimedean and turn at a constant rate around a stationary centre. In other conditions, they perform a compound (quasi-periodic) motion [9, 10]; even more complex behaviour has been observed [11].

In order to understand or control the dynamics of a spiral, many experiments have been performed (usually in BZ systems) where the rotational and/or temporal invariance is broken by applying an external field: AC [12] or DC electric field [13-15], periodic illumination [16], or periodic mechanical forcing [17]. In addition, experiments have focused on the motion of the spiral tip, and although distortions have been observed, they have not been quantitatively analysed. Here, we study experimentally the motion and distortion of a spiral in a *constant* electric field.

The BZ reaction is an excitable medium of chemicals in an aqueous solution, where the propagating wave is defined by the ionic concentrations of the chemical species. The effect of a DC electric field on a single wave is to increase its velocity if it is moving towards the anode, and to decrease it if it is moving away [18, 19]. Studies of spirals in a DC field have shown that it also drifts towards the anode, but with an angle: there is an induced velocity orthogonal to the field [13, 14], with a sign that depends on the chirality of the spiral. Although kinematic models find similar motion [20], the mechanism for this drift angle is not understood.

In this paper, we first show how symmetry considerations lead to equations of motion that qualitatively agree with the observation of a drift angle. We then present an experimental study of a spiral wave in the BZ reaction, under a constant electric current. We measure the position, rotation period, and changes in shape of the spiral as a function of imposed current, by fitting an Archimedean spiral to the experimental image. We find that the spiral undergoes distortions and relaxations which lead to a complex trajectory. We give a quantitative account of this behaviour, which we call the «pommel-lift» (1) effect.

Different *ad hoc* models have been proposed to explain the angle of the spiral motion with respect to an external field. We argue here that the angle is a generic feature of the motion of a rotating object coupled to a constant vectorial field. Hence, it is rather a motion *without* such an angle which should be surprising.

A decisive step towards an understanding of spiral motion in an excitable medium was made by Barkley [21], who represented the full bifurcation diagram for the spiral by the motion of its centre. Thus the dynamics of the PDEs describing excitable media is reduced to five ODEs, which include the transition to compound quasi-periodic motion known as meandering. By restricting ourselves to a simply rotating spiral, the number of ODEs reduces to three: two for the centre ($\partial_t x_c = 0$, $\partial_t y_c = 0$), and one for the phase ($\partial_t \theta = \Omega$). For a constant external field $\boldsymbol{E}$, one expects no change in the rotation rate, characterized by the pseudovector Ω, which includes the chirality. However, the translation modes need not remain stationary. The simplest way to make a true vector with $\boldsymbol{E}$ and Ω is $\partial_t (x_c, y_c) = a\boldsymbol{E} + b\Omega \times \boldsymbol{E}$, where a and b are two scalar coefficients. Their ratio defines the drift angle, which should be independent of the magnitude of $\boldsymbol{E}$ since a and b are constant to lowest order in $\boldsymbol{E}$.

Another approach starts from the reaction-diffusion equations for an excitable medium with a single activator and single inhibitor [2]. If the external field couples with different coefficients to the spatial gradients of the two variables, then its effect cannot be reduced to a simple global translation, and a drift angle is expected. Since it is known that the activator in BZ is a neutral species ($HBrO_2$) and the inhibitor is charged (Br^-), it is reasonable to expect such a description to apply.

Our experiment is done in an «almost open» reactor, in which the ferroin catalyst is fixed in a silica-polyacrylamide gel (20% silica, 5 mM ferroin) (2). The gel (0.9 mm thick) is cut into a square piece (side of about 25 mm), and sits in a plexiglas cell which is open at the top, with a reservoir on either side, as shown in fig. 1. A previously mixed solution of 150 mM H_2SO_4, 400 mM $NaBrO_3$, 30 mM malonic acid, and 5 mM sodium dodecyl sulfate (SDS) is continuously fed by four small nozzles near each corner of the gel, at a total rate of 56 ml/hour. The solution always flows from the nozzles to the edges, without backflow. SDS does not interfere with the BZ reaction [22], but reduces surface tension so that the removal of solution at the far edges of the cell is continuous, ensuring a constant liquid height of about 1 mm above the gel. This layer guarantees a homogeneous distribution of new solution. The residence time is about one minute, based on the volume of the cell containing the gel. Under these conditions, the spiral performs simple rotation. After about 4 hours, the contrast begins to fade, and the spiral's characteristics change rapidly. We thus limit our experiments to the first few hours. All experiments are performed at (23 ± 1) °C.

An important aspect of our set-up is avoiding contamination of the BZ reaction with the electrochemical byproducts of applying a current. We place a platinum wire electrode ($\phi =$

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(1) A device used to tow skiers uphill, consisting of a pole attached by a spring to a moving cable.

(2) We use a modification of the recipe found in [17], developed in our laboratory by Qi Ouyang.

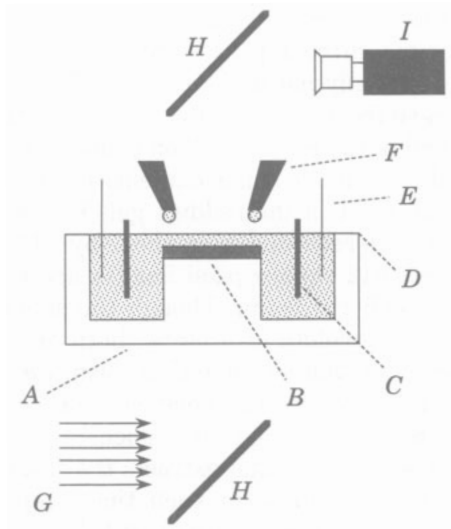

Fig. 1. – A diagram of the experimental apparatus: *A*: plexiglas cell (7 cm long); *B*: gel; *C*: glass baffles; *D*: evacuation point for solution; *E*: electrodes; *F*: nozzles for fresh solution; *G*: parallel illumination; *H*: mirrors; *I*: camera. The reaction only takes place in the gel, where the ferroin is confined; the solution filling the cell is represented in dotted grey.

= 0.7 mm, 6 cm apart) far from the gel in each reservoir. Two glass baffles glued into the reservoirs separate the electrodes from the gel, except for a narrow slot at the bottom. The solution is evacuated close to the electrodes (see fig. 1). A power supply generates a constant current (within 2%) from 9 to 80 mA, and the resistance of the whole cell is about 110 Ω, independent of the presence of the gel. The reaction is uncontaminated for current values at

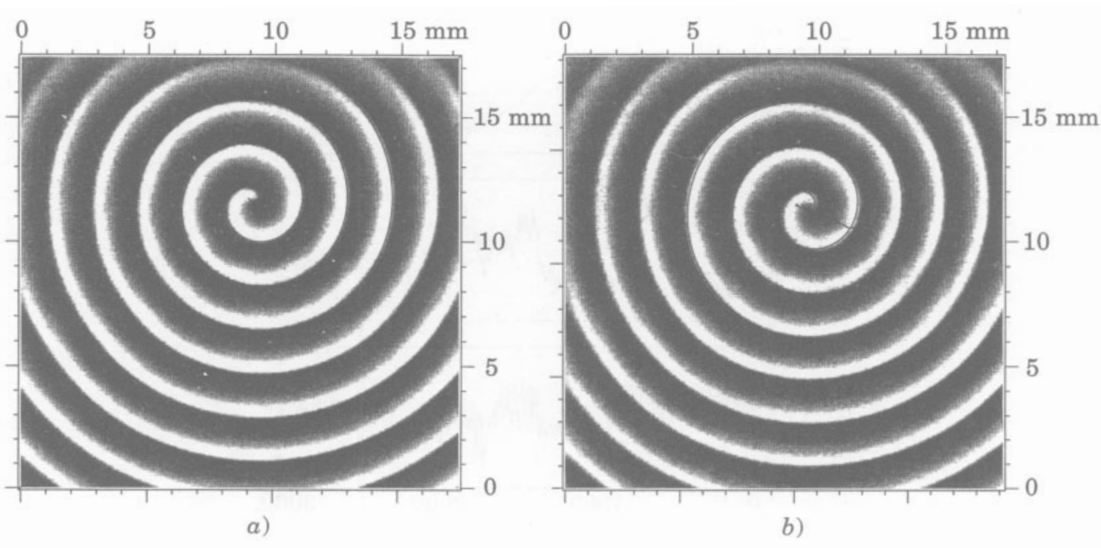

Fig. 2. – *a*) A free spiral ($I = 0$), with the best Archimedean fit superimposed. The cross marks the centre of the fitted spiral ($\alpha = 0.014$). *b*) A spiral with $I = 37.8$ mA (the anode is on the right), with the trajectory of the centre superimposed (x_c *vs.* y_c). The image shown is at $t = 700$ s, at the spiral's maximum distortion ($\alpha = 0.04$).

least up to 100 mA. From the ratio of the gel thickness to the total solution depth, we estimate that half of the applied current passes through the gel.

To create a spiral, the gel is initially put in a Petri dish filled with BZ solution. Touching it with a silver wire initiates a wave front, which is then broken with an iron wire to form a pair of spirals. The gel is then moved to the reactor. When properly done, one spiral is close to the gel edge, and can be pushed off with a small current, leaving a single spiral.

The spiral appears as a blue front in the reddish gel. The cell is illuminated from below with parallel white light; images are recorded from above by a CCD camera, with a blue filter to enhance constrast. The 726 × 512 square pixel images are videotaped for later analysis, with 1 pixel corresponding to (45.5 ± 0.1) μm. Though the actual form of spiral waves may depart from Archimedean or the involute of a circle, both are equally good fits of experimental spirals [6, 23]. Because of its simpler analytical form, we use the Archimedean shape: $r_A(\theta) = \chi(p/2\pi)(\theta - \theta_0)$, where (r, θ) are the polar coordinates, $\chi = \pm 1$ is the chirality, p is the pitch, and θ_0 is the phase at the origin; θ is such that $r_A \geq 0$. A computer program analyses the video images frame by frame and extracts the front of the spiral, which is then best-least-square–fitted to $r_A(\theta)$, providing at each time step the position of the centre (x_c, y_c), p, and θ_0. After the current has been turned off for long enough, agreement between data and fit is within experimental precision, and remains so. We call this a relaxed spiral. In fig. 2*a*), we show a typical spiral, with the Archimedean fit superimposed.

With no applied current, we measure a pitch of (1.73 ± 0.09) mm, and a period of rotation from 41 s to 46 s; these variations may be due to the long relaxation time of the spiral. Thus the propagation velocity of spiral wave fronts is $p/T = (2.4 \pm 0.2)$ mm/min. Under the same

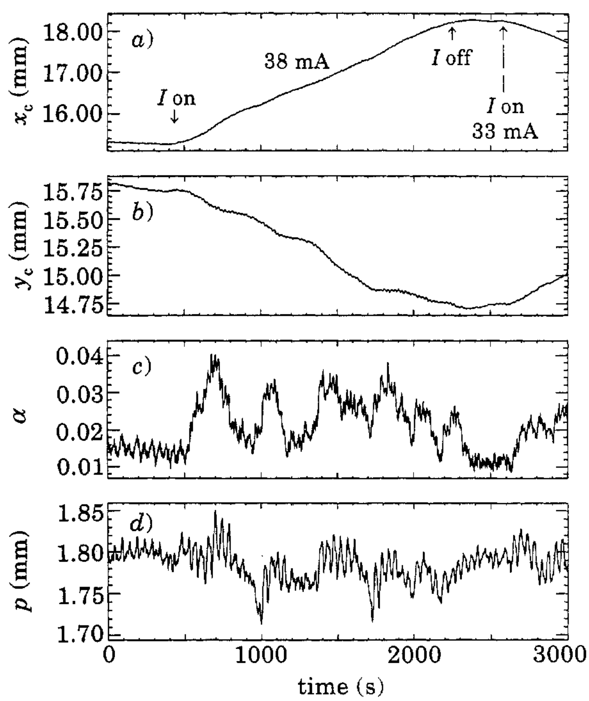

Fig. 3. – The drift and distortion of the spiral *vs.* time: *a*) x_c, *b*) y_c, *c*) distortion parameter α, *d*) pitch p. From $t = 420$ s to $t = 2220$ s, $I = 37.8$ mA. From $t = 2580$ s, $I = -33.1$ mA. Note the difference in scale between x and y. The rapid oscillations in p at the rotation frequency are an artifact of the analysing method.

conditions, a wave front with the same curvature propagates into a fully recovered medium with a velocity of $v_0 \simeq 2.7$ mm/min. Therefore the spiral wave fronts are interacting, as quantified by the ratio $\gamma = p/v_0 T \simeq 0.89$. The fact that the spiral pitch is slightly shorter than the refractory tail means that the spiral is an extended object with self-interactions.

We now analyse the effect of a constant current. As it drifts, the spiral becomes distorted: fig. 2*b*) shows a spiral after about 300 s of current (38 mA). The distortion is visible in the compressed pitch to the lower right of the centre, and the elongated pitch to the upper left. Also shown in fig. 2*b*) is the total trajectory of the centre during 30 minutes of current, as it moves from upper left to lower right; the compression of the spiral is in the direction of motion. The response of the spiral is shown quantitatively in fig. 3. A current of 38 mA is applied from $t = 420$ s to 2220 s. At $t = 2580$ s, a current of 33 mA is turned on, with opposite polarity. The motion of the spiral centre parallel to the electric current ($x_c(t)$, fig. 3*a*)) is straightforward: a rapid approach to constant velocity. The motion of the centre perperdicular to the current ($y_c(t)$, fig. 3*b*)) is more complicated. The velocity is not constant, and oscillations are evident. The mean pitch also changes in a complicated way ($p(t)$, fig. 3*d*)), although its initial response to the current is always a decrease: the spiral is compressed by the imposed current.

To quantify the distortions of the spiral, we introduce the parameter $\alpha \equiv \langle (r(\theta) - r_A(\theta))^2 \rangle^{1/2}/p$, the r.m.s. of the deviations along the actual spiral from the best Archimedean fit, normalized by the pitch. This parameter typically varies from about 10^{-2} for relaxed spirals to about 10^{-1} for our most distorted spirals. In fig. 3*c*), one sees from $\alpha(t)$ that under constant current, the spiral undergoes a series of distortions and relaxations roughly every 400 s (~ 10 periods). The correlation between these oscillations and those of the y_c component suggests a relation between the compressional modes of the spiral and the variations in its drift. This dynamical behaviour is reminiscent of the motion of a pommel-lift (see [1]) pulling a skier uphill: the average motion is constant, but there are oscillating elongations and contractions. Though the trajectory makes an angle with the direction of the field, it is in practice not possible to define a unique angle at each current.

As a function of current, the drift velocity parallel to the current v_x varies linearly (fig. 4). Although the drift perpendicular to the current (v_y) is not constant, the total drift speed also appears linear with current, in agreement with previous measurements, because v_y is in general much smaller than v_x. In many cases the spiral centre continues to move in the

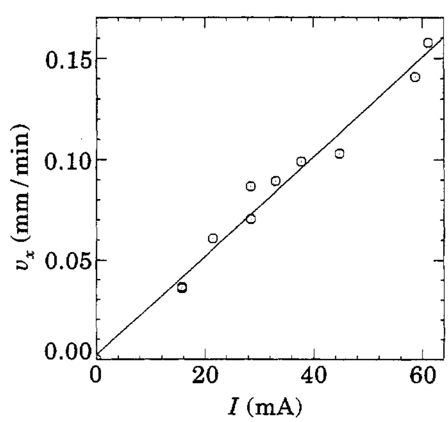

Fig. 4. – The spiral drift velocity parallel to the applied current (v_x) in mm/min, as a function of applied current I in mA.

y-direction after the current has been turned off. Preliminary measurements of the rotation period indicate that it decreases with applied current.

In conclusion, although general symmetry arguments justify the drift angle of a spiral in a constant electric field, the experimental picture is more complicated. When a constant current is applied to a spiral in the BZ reaction, the spiral undergoes successive deformations and relaxations. These deformations appear coupled to the perpendicular trajectory, which could account for the non-steady motion of the centre. Since the drift of the spiral excites its internal modes, the observed behaviour may also be connected to meandering. From these observations, it appears that spiral dynamics cannot be fully described by treating only their centre or tip, but there is as yet no satisfactory way to represent or fit a distorted spiral. A model relating the spiral motion to the distortions is also lacking.

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