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Simple and Complex Spiral Wave Dynamics

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Simple and complex spiral wave dynamics†

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Spiral waves rotating in an excitable medium present a classical example of unusual nonlinear phenomena in distributed systems. In this paper we discuss the results of experimental studies of spiral wave dynamics in homogeneous excitable media which are modifications of the Belousov–Zhabotinsky system. A variety of dynamical régimes from very simple and well ordered to irregular complex ones are described that are created under different experimental conditions. Spiral wave dynamics is considered in stationary media with different excitability, under the influence of the boundary conditions, and under a periodic modulation of a parameter of the medium. The experimentally observed patterns are compared with the data of computer simulations on the basis of equations representing the properties of excitable media.

1. Introduction

The propagation of waves through an excitable medium is a fascinating example of a nonlinear phenomenon in non-equilibrium systems. A localized disturbance can switch on the excitation processes resulting in a jump from a resting to an active state in a small segment of the medium. Because of the interaction between neighboring segments these jumps can propagate through the medium as excitation waves.

In the one-dimensional case such waves are propagating without any decrement of the amplitude and the velocity until they reach the boundary of the medium. If the excitable medium has the shape of a ring channel, the excitation wave can run along this ring for an infinitely long time. The simplest analogue of such a rotation in a two-dimensional medium is a wave circulating around some obstacle (Wiener & Rosenblueth 1946). In this case, the wave front has the form of a spiral that rotates with a constant angular velocity around a fixed core. Still the dynamics of such a rotation is rather simple and can be described, in fact, in terms of a one-dimensional wave phenomenon.

A qualitatively new régime can be observed in two-dimensional homogeneous media without artificial obstacles. Figure 1*a* shows an example of the spiral wave rotating in such a two-dimensional system. In this completely homogeneous medium its tip describes autonomously a circle at the boundary of the spiral core (see figure 1*b*, *c*). These waves were discovered in a thin layer of a chemical solu-

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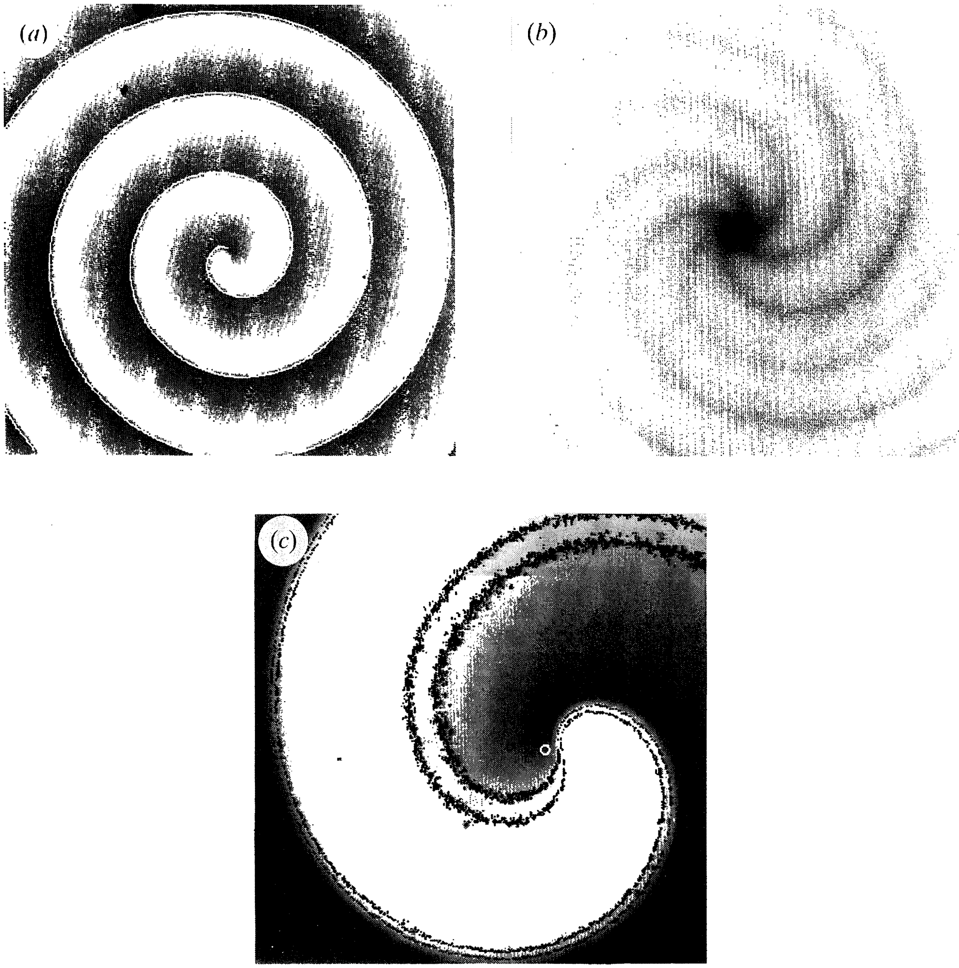

Figure 1. (*a*) Snapshot of a rigidly rotating spiral wave with archimedian shape in the ferroin-catalysed BZ reaction. Image size $7 \times 7\ mm^2$. (*b*) Digital overlay of six snapshots showing the spiral core as a dark circular area. (*c*) Close-up view of the spiral tip with its rotation axis indicated by a small white circle.

tion (Zhabotinsky & Zaikin 1973; Winfree 1972) and are also observed in many natural objects, such as cardiac tissue (Davidenko *et al.* 1992), chicken retina (Gorelova & Bures 1983), and the cellular slime mold Dictyostelium discoideum (Gerisch 1965).

A spiral wave rotating in a homogeneous medium has no fixed core. Hence the spiral wave is not anchored anywhere on the plane and its core can move through the medium. These additional degrees of freedom result sometimes in the appearance of very complicated dynamical régimes which are currently the object of intensive investigation. For instance, the composition of the rotation of the tip around such a core and the core motion along a circular path should result in cycloidal trajectories of the tip (Zykov 1986; Jahnke *et al.* 1989). In fact, it was shown that in many cases a cycloid gives a good approximation for the experimentally observed compound tip motion (Plesser *et al.* 1990; Skinner & Swinney 1991).

PHILOSOPHICAL TRANSACTIONS OF THE ROYAL SOCIETY A MATHEMATICAL, PHYSICAL & ENGINEERING SCIENCES

In this context the Belousov–Zhabotinsky (BZ) reaction proves to be a very effective tool for an experimental investigation of the nonlinear dynamical régimes in active media. Practically, a large number of quantitative experimental studies on wave phenomena have appeared since devices for two-dimensional spectrophotometry have become available (Müller *et al.* 1986). Owing to the precise recording of the time series of images that contain information of the spatial distribution of chemical compounds, the extraction of detailed spatio-temporal parameters is now feasible.

This paper describes recent results concerned with the investigation of the dynamical régimes in the BZ reaction. All the data discussed below are related to the case of homogeneous excitable media. The data obtained demonstrate that even in this simplest case the motion of the spiral wave can be very complicated. We consider three different factors causing such complexity. First, we give a short review of the present experimental results demonstrating the diversity of spiral wave dynamics under variation of the chemical composition of the BZ solution. Subsequently, we describe the influence of the boundary of the medium by considering spiral waves rotating in a small disk of an excitable medium. The last section is devoted to a periodic modulation of some of the parameters of the medium. In conclusion, we compare the experimental results with the data of computer simulations of spiral waves.

2. Rigid and compound rotation

The experimental investigation of different kinds of spiral wave régimes was carried out in the cerium-catalysed modification of the BZ reaction (Nagy-Ungvarai *et al.* 1993). The initial concentrations of the reagents in the solution were: 0.3 M $NaBrO_3$, 0.366 M malonic acid, 0.09 M brommalonic acid, 0.006 M $Ce(SO_4)2$. The concentration of H_2SO_4 was one of the control parameters and was varied in the range from 0.11–0.41 M. The solutions were filtered with Millipore filter (0.45 μm), thermostated at 25 ± 0.1 °C and placed in a flat Petri dish (diameter 70 mm) covered with a glass plate leaving an air gap of a few millimetres above the layer surface. The thickness of the BZ layer was kept as thin as 0.3 mm to avoid three-dimensional effects. The light absorption which depends on the concentration of the oxidized catalyst Ce(IV) was measured by UV-sensitive two-dimensional digital spectrophotometry at 344 nm. The oxidized front of the waves appear as dark bands due to higher absorption of Ce^{4+}.

The excitability of the solution is not a constant in the used reactors, but decreases with time. This process is rather slow compared with the rotation period of the spiral. It turns out that one can observe a remarkable diversity of the spiral wave motion during the aging of the BZ solution (Nagy-Ungvarai *et al.* 1993).

The rigid rotation of the spiral wave is typical for BZ solutions with a high concentration of sulphuric acid. The spiral wave rotates around a circular core with a constant angular velocity. The shape of the wave front does not change with time and is very close to an archimedian spiral which asymptotically is an involute of the spiral core. This case corresponds to a rather large propagation velocity and hence to a highly excitable medium.

It is known that decreasing the excitability leads at first to cycloidal rotation which can be described as a quasiperiodic motion (Plesser *et al.* 1990). In the

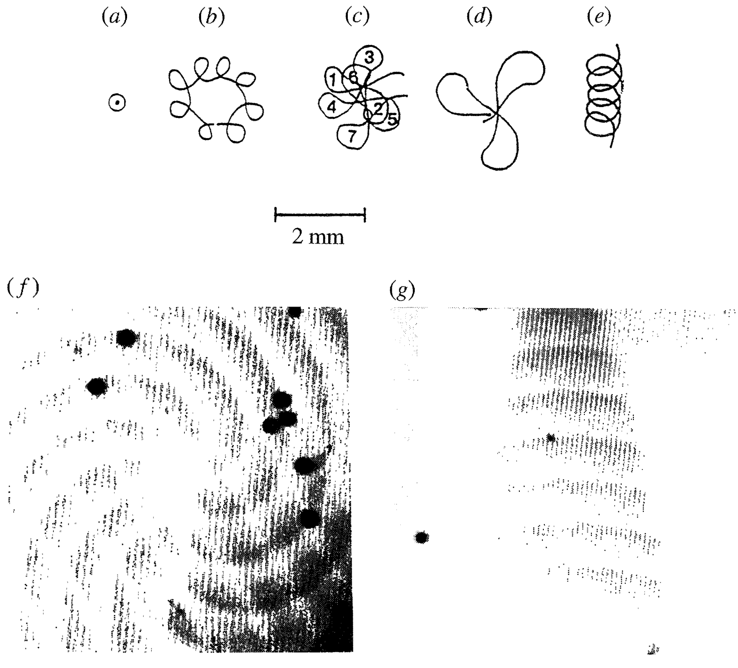

Figure 2. The trajectories of the wave tip for different excitability of the medium (*a*)–(*e*). Overlay of digital images in the cerium-catalysed BZ reaction showing one loop of complex motion (*f*). Shrinking of open end wave in a weakly excitable medium (*g*).

experiments under consideration there are two factors that lead to the decrease of the excitability. The first factor is the initial concentration of sulphuric acid and the second one is the aging of the solution. In figure 2*a*–*e* several types of the tip trajectories observed during the experiments are presented. The experiment was started with sufficiently high concentration of sulphuric acid and a circular trajectory of the tip was recorded (figure 2*a*). The decreasing of the excitability with time destroys the rigid rotation of the spiral wave and leads to the appearance of a compound motion (figure 2*b*). Then the motion becomes more complex (irregular or even chaotic) (figure 2*c*). Figure 2*f* shows in detail the wave motion during one loop of such a complex motion. But for very low excitability the tip motion becomes again rather regular with cycloid trajectories (figure 2*d*, *e*). Near the limit of excitability which restricts the ability of the medium to support the wave propagation one can observe even rigid rotation, but with an abnormally large core (Nagy-Ungvarai *et al.* 1993). For an excitability lower than this limit an open wave end does not curl up to form a spiral but shrinks with time (see figure 2*g*). Under these conditions a self-sustained propagation of the excitation waves is impossible.

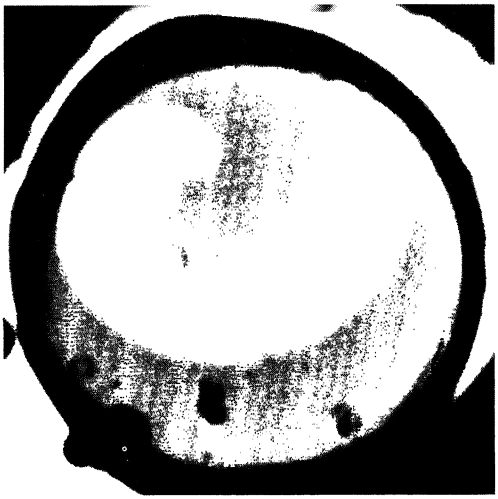

Figure 3. Spiral wave rotating in a small disc of an excitable medium with a diameter of 2 mm.

3. Spiral wave in a small disc

Usually the radius of the spiral wave core is very small with respect to the size of the Petri dish and the boundary conditions do not influence the tip motion in common chemical experiments. A special technique was used to study the role of the boundary of the excitable medium.

We used the ferroin-catalysed BZ solution with the following concentrations of the reagents: 0.33 M $NaBrO_3$, 0.24 M malonic acid, 0.06 M NaBr, 0.41 M H_2SO_4 and 0.003 M ferroin. The possibility of hydrodynamic disturbances was inhibited by gelling the reagent with agarose gel. The two-dimensional distribution of the intensity of the transmitted light was registered by a video recorder (Umatic, Sony). The oxidized fronts of the waves appear as bright bands due to low absorption of ferricin. The traces of the spiral tip were then analysed visually on a video screen.

We cut a small disc (diameter about 3 mm) of the gel by a special tool (a cylinder with a thin wall). For the cut we selected a piece of the medium surrounding the tip of a spiral wave created in the Petri dish. Then the circulation of the excitation wave occurred in a small disc of the gel (see figure 3). Significant oxygen influence on the reaction (which also inhibits the reaction) was prevented by covering the dish by a thin layer of transparent, chemically inert silicon oil.

The following evolution of the spiral is subjected to a strong influence of the boundary due to the small size of the disc and depends on the initial location of the wave inside the disc. As a rule, the boundary attracts the spiral wave core. Sometimes it leads to a fast motion toward the boundary and the death of the spiral. But rather often spiral waves started to move along the system boundary. In this case the spiral tip describes a quite complicated trajectory (see figure 4). In principle, the shape of the trajectory presented in figure 4 is very similar to an epicycloid. One can distinguish the motion around a circular core and the drift of the core along the boundary. During the drift the characteristic distance from the centre of the core to the boundary is about 0.3 mm. This drift occurs with a velocity of about 0.15 mm min^{-1}.

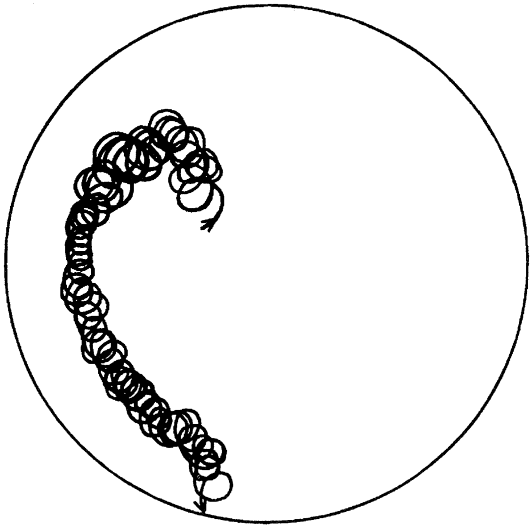

Figure 4. Trajectory of the tip of the wave rotating in a small disc with a diameter of 3 mm.

4. Spiral wave under external forcing

To study the behaviour of the spiral waves under periodic modulation of the medium parameters we used the light-sensitive BZ-reaction (Krug *et al.* 1990). The properties of this Ru(bpy)$^{32+}$-catalysed reaction are very close to those of the common BZ as long as the ruthenium–bipyridyl complex remains in its reduced and electronically unexcited state. Once the complex is photochemically excited, it slowly catalyses the production of the inhibitor bromide. It thus suppresses the excitability of the medium and allows to control the spiral wave parameters by externally applied illumination (Agladze *et al.* 1987).

In our experiments (see Steinbock *et al.* 1993) the catalyst (4 mM) was immobilized in a silica-gel matrix (thickness 0.7 mm, diameter 7 cm). The concentrations of the other reactants were: 0.09 M NaBr, 0.19 M $NaBrO_3$, 0.17 M malonic acid, and 0.35 M H_2SO_4. The temperature was kept fixed at (23 ± 1 °C). White light illuminating the entire observation area was polarized by a rotating polarization filter and directed to the active medium by reflection from a tilted glass plate. Due to the rotation of the polarization vector the intensity of the reflected light was modulated sinusoidally with time (0.49–1.36 mW cm^{-2}). The absorption of the observation light was detected by a charge-coupled-device camera (Hamamatsu C3077) at 490 nm, stored on a video recorder and finally digitized by an image-aquisition card. The temporal trace of the wave tip was detected visually with a reticle in digitized images.

At constant light intensity (0.93 mW cm^{-2}) the tip of the created spiral wave describes the trajectory presented in figure 5. The trajectory is almost a five-lobed hypocycloid. The wave period is measured at the centre of this curve as $T_0 =$ 24.5 s. When rotating the polarization vector by a fixed angle, one can change the stationary level of the illumination in the range from 0.49 to 1.36 mW cm^{-2}. But this leads only to small variations of the rotation period and slightly deforms the shape of the tip trajectory.

A periodic modulation of the applied light intensity within the same range

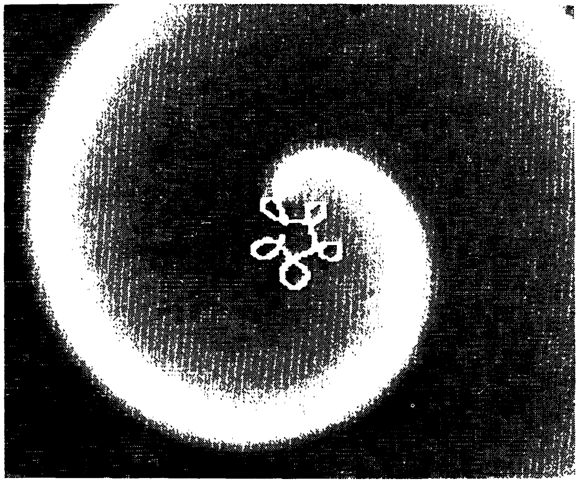

Figure 5. Spiral wave in the ruthenium-catalysed BZ reaction at a steady level of light intensity (0.93 $mW\,cm^{-2}$). The tip trajectory (overlaid white curve) is similar to a five-lobed hypocycloid. Image size 3.8 mm by 3.0 mm.

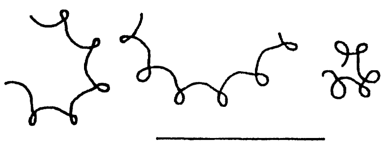

Figure 6. Sequence of tip trajectories measured under sinusoidal modulation of light intensity (between 0.49–1.36 $mW\,cm^{-2}$) with period T_m: 32.9 (*a*), 34.5 (*b*), 52.2 (*c*). Scale bar: 0.2 mm.

was created by the rotation of the polarization vector with a constant angular velocity and causes dramatic changes in the dynamics of spiral rotation. The small variations of the trajectory shape that occur during each period of the modulation are accumulated in time. As a result, the modulation forces the tip to follow trajectories which differ significantly from those observed at constant intensities. The shape of trajectories depends strongly on the modulation period T_m.

For instance, the unperturbed trajectory can be deformed to a multilobed one occupying an extremely large area (figure 6*a*, *b*). It is important that in this case the oscillations of the tip velocity and the curvature of the trajectory are synchronized by the external rhythm. Moreover, one lobe of the trajectory corresponds to one period of external modulation.

Figure 6*c* shows a surprising trajectory with alternating distances between neighbouring lobes. In this frequency range the spiral tip describes a pair of lobes during one external period of the modulation. For modulation periods between those of figure 6*b*, *c* an irregular motion with epicyclic segments of the trajectories was observed.

5. Conclusion

The experimental data considered prove that the spiral waves rotating in a homogeneous solution of the BZ reagent exhibit a wide spectrum of different régimes in their dynamics. The simplest régime of rigid rotation can be easily destroyed by different kinds of experimental conditions. Quite generally, compound or more complicated trajectories are more common for the BZ reaction than the rigid rotation.

To reproduce the experimentally observed dynamics of spiral waves it is sufficient to use one of different available and rather simple two-component reaction-diffusion models of an excitable medium. Indeed, the transition from the rigid rotation to the compound or irregular one was reported by Zykov (1986) and by Barkley *et al.* (1990) and was described in detail by Jahnke & Winfree (1991). The compound rotation occurring during a drift along the boundary of a small disc was observed in simulations by Davydov & Zykov (1993) and by Sepulchre & Babloyantz (1993). The spiral wave dynamics detected under periodic modulation of the excitability is also in good qualitative agreement with corresponding computational results (Steinbock *et al.* 1993; Braune & Engel 1993).

It means that the details of excitable kinetics do not matter to reproduce in a qualitative manner the observed transitions from simple dynamics of spiral waves to a more complex régime. On the other hand, it is very important that one can use the same model to simulate different kinds of complex dynamics. For instance, one can use the two-component Oregonator model for the BZ reaction to obtain all of the dynamical régimes described above.

This corroborates that the observed complexity in the dynamic evolution of spiral wave is a general property of excitable media. The specific features of the chemical realization of an excitable system is probably negligible in this context. Similar dynamic régimes should be observed in other types of excitable media. On the other hand, it is obvious that the different dynamical régimes described are of similar nature. The investigation of one of them should help to understand the properties of the others. It offers a chance to elaborate a unifying theoretical description of the observed régimes as a result of a systematic study of the spiral wave complexity emerging both in experimental and simulation results.

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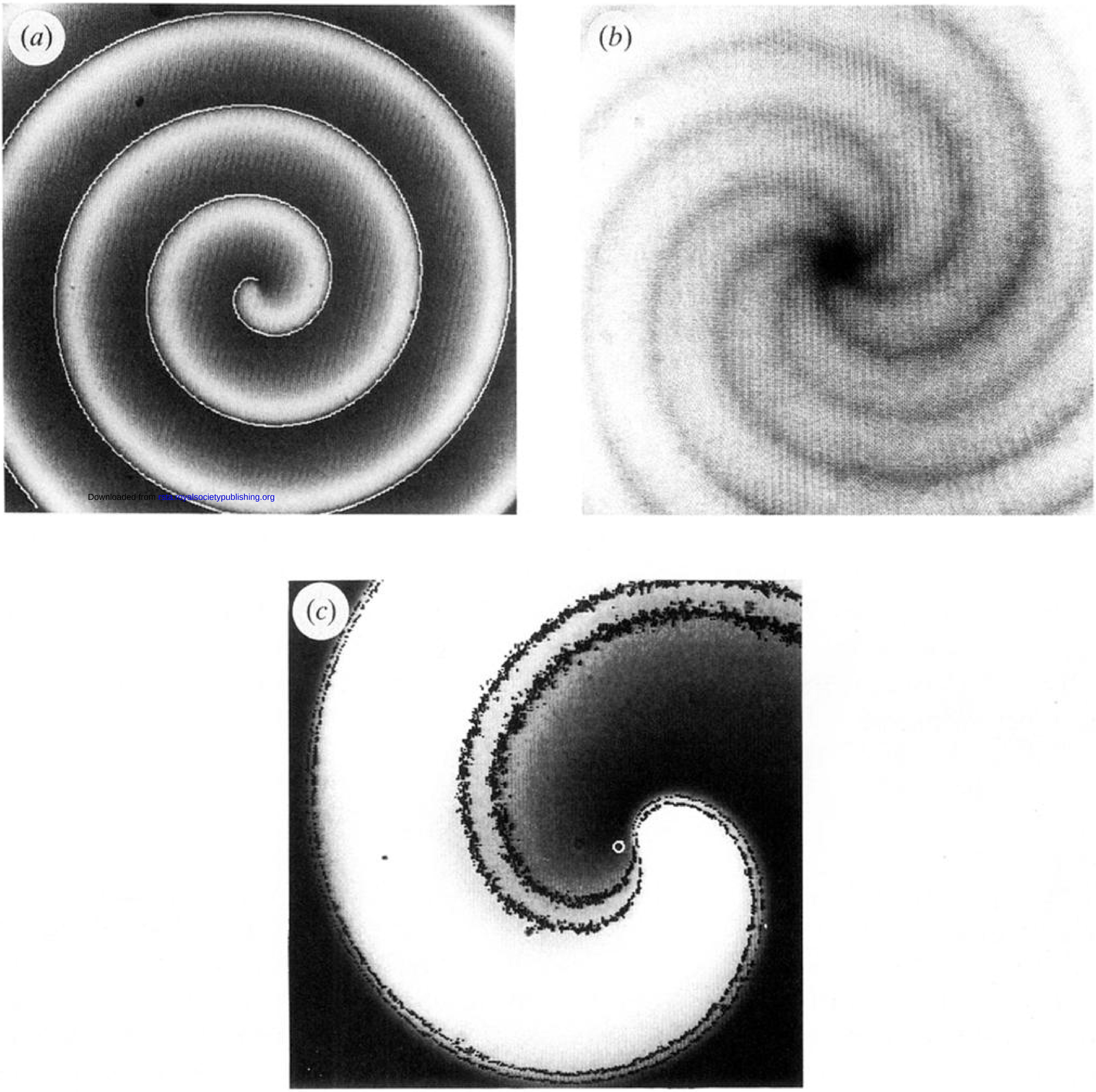

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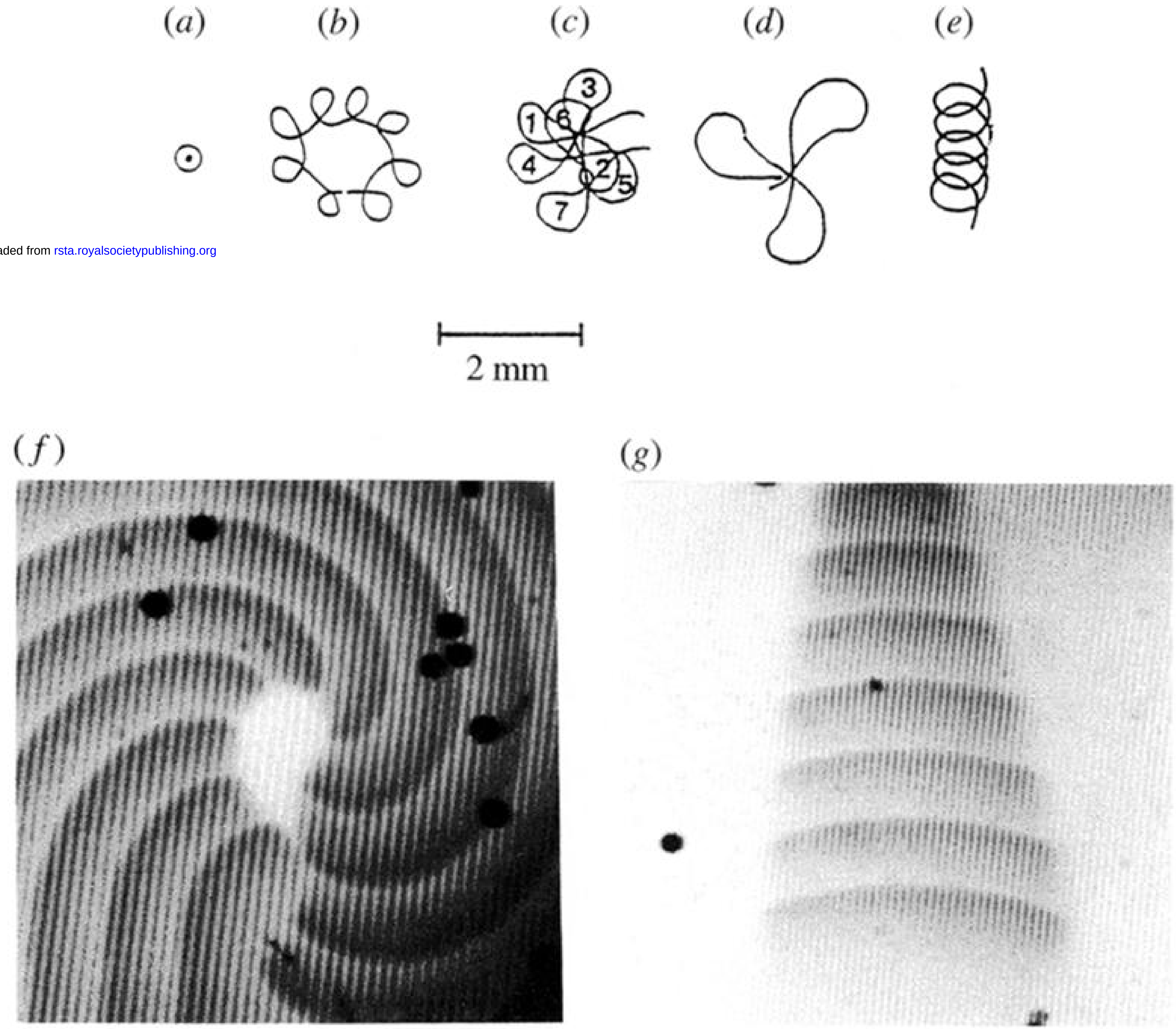

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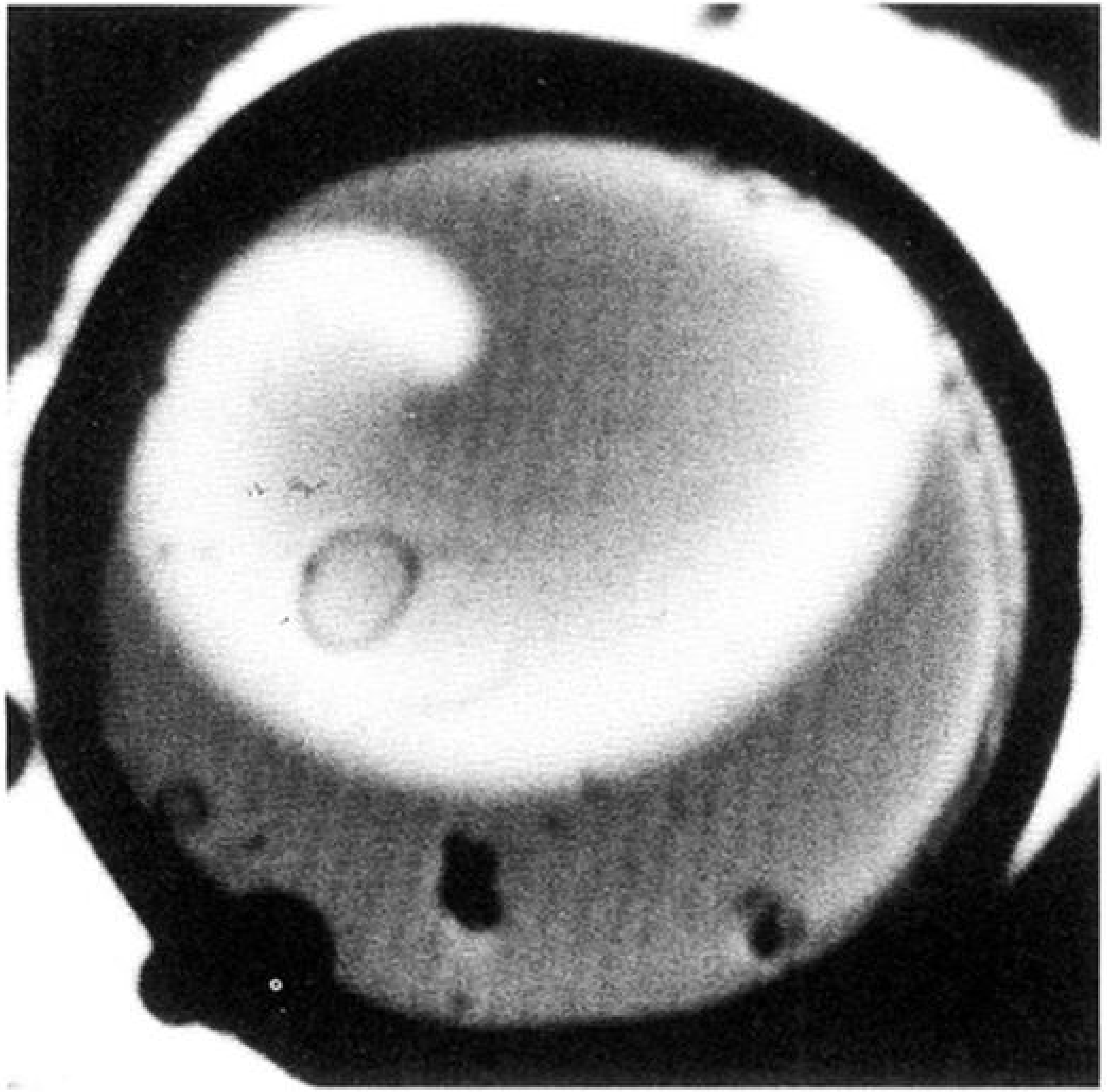

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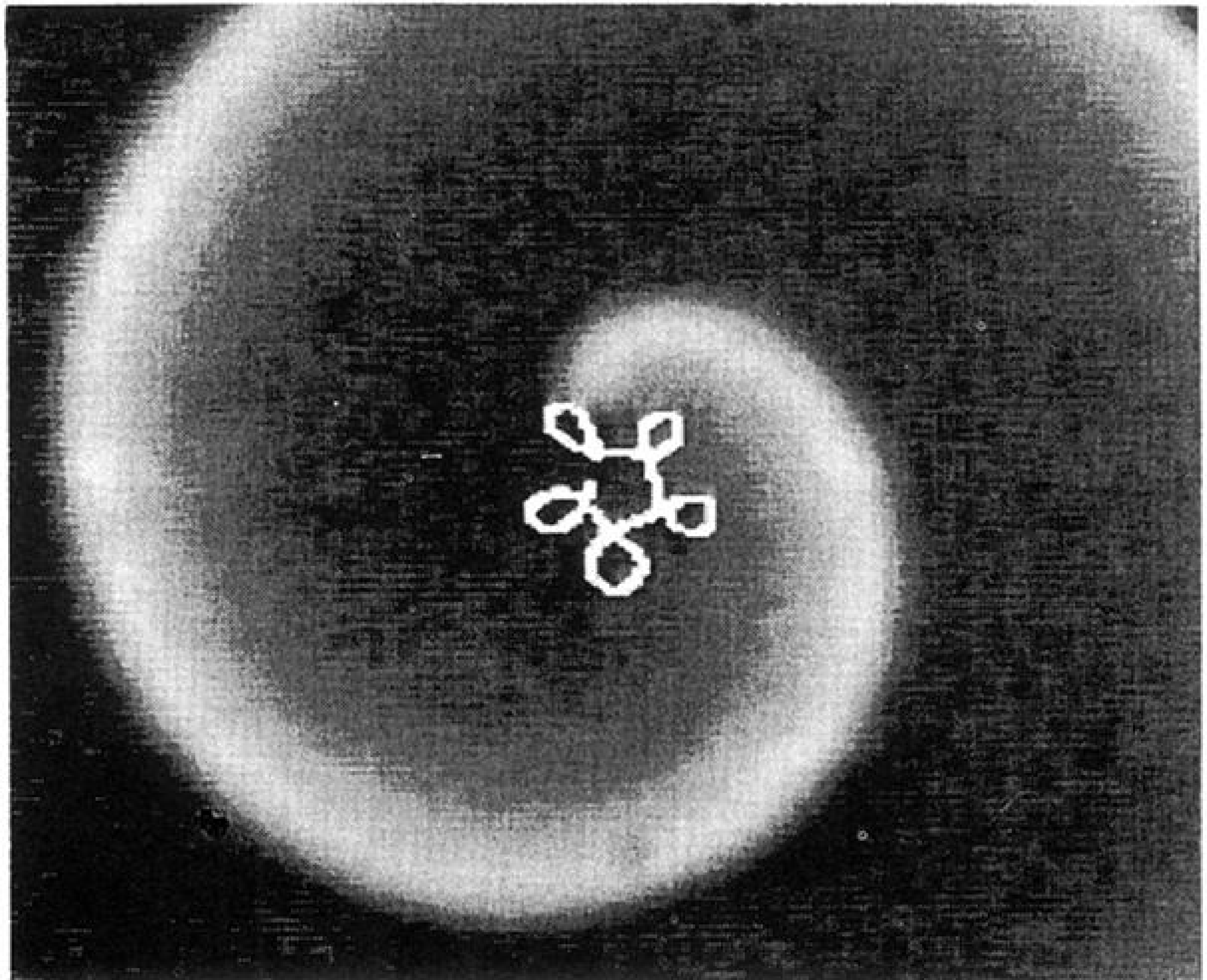

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