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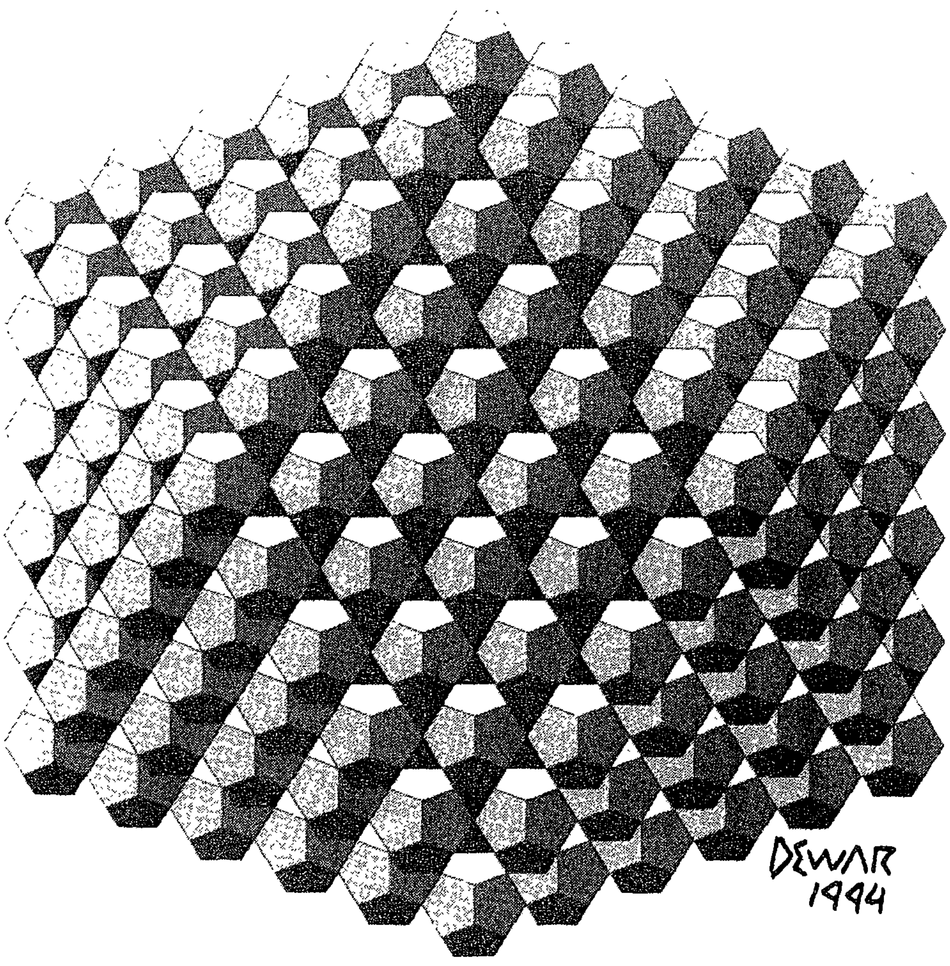

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SPIRAL WAVE DYNAMICS

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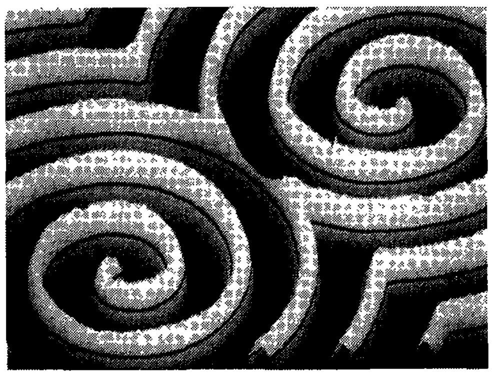
Excitable systems play an important role in the field of spatio-temporal selforganization. In biology, they serve as media for signal transmission, for instance in pulse propagation along nerve fibers or across the chambers of the heart. Selforganization, in general, occurs by symmetry-breaking transitions from homogeneous to structured states. In excitable media the coupling of nonlinear reactions with diffusion can result in the formation of traveling waves which, in two-dimensional extended media, have circular or spiral-shaped geometries. Two model systems are especially suited for in depth investigations of the wave dynamics: a chemical system, namely the excitable Belousov-Zhabotinsky reaction, and a biological system, which is the cellular slime mold Dictyostelium discoideum during early stages of cell aggregation.



Spiral symmetry in the Belousov-Zhabotinsky reaction

We have studied the formation and dynamic evolution of spiral waves in both systems in quantitative detail by applying computerized video imaging techniques. Emphasis was given to elucidate the structure and the properties of the spiral core. Spirals in well excitable preparations of the Belousov-Zhabotinsky reaction, as observed by light absorption, have approximately Archimedian shape. Their tip rotates on a circle along the boundary of the core (diameter ≈ 0.7 mm) inside which the amplitude of excitation decreases towards the rotation center, which is a singular point (diameter $< 10\ \mu m$) with quasi-stationary chemistry. When decrasing the excitability of the solution, the steady and rigid tip rotation undergoes a transition to a compound motion, close to that of an epicycloid, and the spiral shape becomes non-Archimedian. Other transitions to more complex quasiperiodic and possibly chaotic motion are also observed [1,2].

The dynamic behaviour of propagating chemical waves was also investigated under the influence of an externally applied electric field. Depending on polarity and strength, this field leads to acceleration, slowing down, reversal or splitting of wave fronts due to its action on the drift of ions participating in the reaction [3]. Spiral waves are subjected to a drift in space accompanied by spiral deformation and changes in the wavelength, analogous to the Doppler shift. Due to a drift component perpendicular to the field direction spirals of opposite chirality can be forced to collide with each other. This results in mutual annihilation or complex interaction of their cores [4].

An efficient tool for external control of spiral wave parameters is found in a light-sensitive, ruthenium-catalyzed preparation of the Belousov-Zhabotinsky reaction. If the ruthenium complex is photochemically excited, it catalyzes the production of the inhibitor species of the reaction, bromide. Thus, externally applied illumination suppresses the excitabiltiy of the medium. A thin beam of laser light was used to produce an unexcitable disk that can serve as an artificial core of a spiral. The radius of the disk determines the spiral wavelength and spirals with continuously changing pitch are easily produced. They contain all the information necessary to derive the disperion relation of the medium. Such disks are also used to anchor meandering spirals to rigid rotation and to produce multi-armed spirals that rotate at equal distances around the disk boundary. After switching off the laser beam, these multiple tips move synchronously into the disk area where they collide with each other and form interesting interaction patterns [5,6].

If illumination is applied globally to the photosensitive reaction layer, one can control the dynamics of spirals in the entire system by periodic modulation of the light intensity. The modulation of excitabilty forces meandering spiral tips to phase-locked motion, a spectrum of open and closed hypocycloidal trajectories, and complex multifrequency patterns. These experimental results are reproduced by numerical simulations with a two-variable Oregonator model which was extended by a term describing the light-induced bromide production. They also show the existence of entrainment bands in the plane of modulation period and amplitude [7].

The slime mold *Dictyostelium discoideum* is well suited for the study of cellular communication and a widely investigated biological example of a self-organizing nonlinear system. About ten thousand individual amoebae of this myxomycete undergo aggregation to form a single organism (a pseudoplasmodium). Subsequently, they differentiate to form a fruiting body bearing spores. The depletion of their food source serves as signal to trigger the onset of the corresponding developmental cycle. The aggregation is coordinated and organized by traveling waves of cyclic adenosinemonophosphate (cAMP). During this phase the majority of the population rests in an excitable state, while other cell groups serve as periodic pacemakers by producing cAMP according to an oscillatory mechanism. By means of diffusion their neighborhood becomes excited and relays the biochemical signal. A propagating wave results which shows, in general, the geometry of concentric circles (target patterns) or rotating spirals. These structures are usually investigated by darkfield photography [8].

An important particularity of the Dictyostelium system is the existence of an additonal non-diffusive transport mechanism: chemotaxis forces the cells to a movement in the direction of the strongest increase of cAMP-concentration. We analyzed the chemotactic motion in quantitative detail by applying a mutual-correlation method for the determination of velocity fields. The direction of the velocities of amoebae and chemical wave propagation are antiparallel, their magnitudes having a ratio of approximately 1:10. The centers of spiral patterns have characteristic properties of attracting domains towards which finally all cells flow as a response to the signal sent out by the travelling wave of increased messenger concentration.

At a later stage in the development of *Dictyostelium discoideum* a slug forms as a migratory stage. Its anterior part consists of prestalk cells that ultimately build the stalk of a fruiting body, while the prespore cells in the remainder differentiate to spores. The individual cells in these slugs follow also well-defined patterns of motion. We found that at this stage the chemotactic cell response is controlled by a three-dimensional scroll-shaped wave of messenger concentration in the highly excitable prestalk zone of the slug that decays in the less-excitabe prespore region into planar wave fronts. In the interface of both regions a complex twisted scroll wave is formed that reduces the wave frequency in the prespore zone [9]. These results yield an explanation of collective self-organized cell motion in a multicellular organism.

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NATURAL BEAUTY OF ARTIFICIALLY TEXTURED SURFACES: MORPHO-BUTTERFLY COLORING WITH PARTICLE ARRAYS

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Is there any minimum unit in the nature's beauty? Let us first think about jewels such as ruby or diamond. Owing to the historic studies of optics and optical properties of crystals, now we understand the origin of the beauty generated by these jewels. The knowledge combined with the high-pressure and high-temperature technologies, now enable us to fabricate and deliver various jewels at a reasonable cost without loosing their fascination. There is another type of beauty in jewels which is not yet fully understood and artificially reproduced. That is iridescent coloring observed in opal or cat's eye. The opal-like coloring is also observed in the surfaces of creatures such as insects.

We can remember that in our childhood we were completely enthralled by strange insects, such as beetles and butterflies. The fascination of their colorful surfaces is now revealed to arises from their fine surface textures which are microscopically difined and then difficult to be mimicked by our current technologies. But we can again raise the question, as in the past we put to the jewels, whether our science and technology can break the code and artificially reproduce fine textures of creatures surface. A study to imitate the textured surface and reproduce the beauty of a kind of butterfly, morpho, made famous by its fascinating iridescent colors has been thus boosted. Morpho has very brilliant wings able to catch our eyes from as far away as 1 km. There are people who believe its beauty is beyond that of the black opal. The natural wing of a morpho-butterfly has a periodic structure, as determined by optical and electron microscopy. Each 100-μm continuous parallel gratings. This structure could be closely duplicated using fine particles and their regular arrays.

Ordered arrays of colloidal particles coated on surfaces are usable in industry either as a diffraction grating, an optical storage medium or an interference layer. Formation of ordered colloidal particle arrays has been studied and fabricated by spin coating [Deckman et al., 1983, 1988], or mechanical or chemical packing of colloidal particles at the air-water interface [Dunsmuir et al., 1989]. Here we propose a novel fabrication method for preparation of colloidal particle arrays with crystalline order. The essence of our fabrication of two-dimensional (2D) assembly of particles lies in the use of stable wetting film that is made on the substrate surface as shown in Figure 1 [Nagayama, 1993]. This wetting film plays two important roles here; 1) 2D liquid medium where particles can be carried by water flow to the crystal boundary for growth (convective self-assembly, Figure 1C) [Denkov, et al., 1992, 1993; Dushkin, et al., 1993[a], 1993[b]; Dimitrov, et al., 1994], and 2) interparticle attractive force induced by the surface tension for the particle packing (lateral capillary force, Figure 1D) [Kralchevsky, et al., 1992, 1994; Velev, et al., 1993]. First, particles undergo the Brownian motion in the thick film (Figure 1A). When the wetting film becomes as thin as the particle size by the removal of water, an ordered 2D domain starts to grow on the surface (Figure 1B).

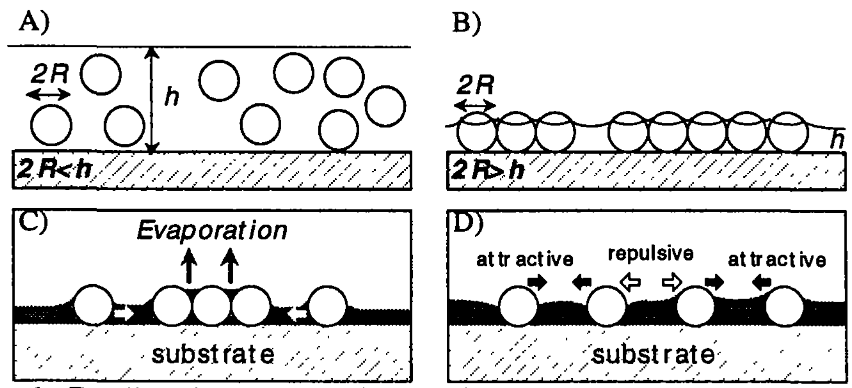

Figure 1 Two-dimensional assembly of particles in the wetting film [Nagayama, 1993].

A) Particles undergo the Brownian motion in the liquid layer of which the thickness is much larger than the particle size.

B) Particles start to assemble in the wetting film as the thickness of the film becomes comparable to or slightly smaller than the particle size.

C) Convective flow responsible for the assembling of particles stimulated by the liquid evaporation.

D) Lateral capillary forces responsible for the hexagonally closed packing of particles.

For a wettable solid plate vertically dipped in a suspension of colloidal particles, monolayers and successive multilayers of particle arrays spontaneously start to form near the three-phase contact line on the plate surface (Figure 2) [Dimitrov & Nagayama, 1995]. The formation of particle array films can be explained again by the mechanism mentioned above. This procedure was applied to different suspensions of polystyrene particles with diameters from 0.079 to 2.106 μm. Wetting films were formed due to the capillary rise along the plates. When the plates were fixed, mono-, bi-, trilayers, etc., started to form in the vicinity of the plate-suspension-air contact line. We controlled the growth of particle arrays that have multiple layers, and especially of monolayer arrays, by withdrawing the substrate plates from the suspensions with an adequate rate.

Regular structure of textured surfaces and the colors of our 953-nm particle array films was compared to those of the wings of a Morpho butterfly, *sulkowsky* (Figure 2) [Dimitrov & Nagayama, 1995]. Our experiments showed that for the same incidence angle of illumination, the dispersed light coloring of the textured surfaces mainly depends on the texture periodicity, and on the orientation of the surface domains. When illuminated by white light, the particle arrays we produced mimic the iridescent coloring of Morpho-butterfly wings. Microscopic observations showed that the similarities in the coloring of both particle arrays and butterfly wings are due to the same special periodicity of the surface texture, which is on the order of or slightly above the wavelength of visual light. Our film fabrication using colloidal particles and the film iridescent coloring will open a new avenue for the additive color processing nowadays, which are used in a wide range of arts and technologies.

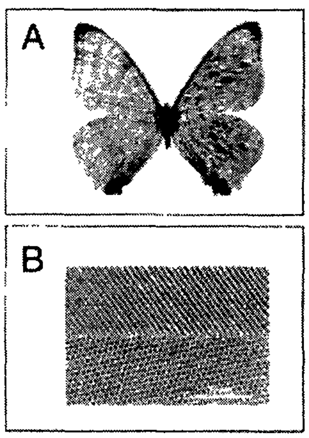

Figure 2 Coloring of textured surfaces with μm-scale periodicity

A) Morpho butterfly (*sulkowsky*) with original (left) and artificial wing (right).

B) Scanning electron microscope image of the grating on a scale from the Morpho butterfly wing (top) and monolayer particle arrays mimicking the wing grating (bottom).

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