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The Search for Turing Structures

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Albeit their prediction by Turing, chemical periodic stationary patterns under far from equilibrium conditions, which are considered as prototypes of dissipative structures, have not yet been observed in a clear-cut way. We here assess the situation and discuss the perturbative influence of convection on their formation and selection mechanisms.

KEY WORDS: Chemical instabilities; double-diffusion; convection.

1. INTRODUCTION

Although cellular patterns in driven hydrodynamic systems have been known for nearly a century,(1,2) the problem of pattern selection is still a very active field, which allows for many new effects and surprises as structures tend to appear in an ever-increasing number of situations. Recent ones, for example, show patterning in swelling polymers due to a mechanical instability and in silicon under cw irradiation leading to ordered coexisting solid and molten regions.(3) (see also Refs. 30 and 31).

On the other hand, the observation of organized behavior (periodic oscillations, waves, etc.) in complex chemical systems is much more recent. It goes back to the work of Belousov and Zhabotinsky[3] and came as a surprise to most chemists. They were therefore at first considered as rather exotic phenomena (although they had been shown by Glansdorff and Prigogine(5) not to be in contradiction with basic thermodynamic principles), but it is now apparent that an increasing number of complex chemical networks may exhibit this behavior when functioning sufficiently far from thermal equilibrium.(6) The control of their occurrence may

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[3] See Ref. 4 for a review of the development of the Belousov–Zhabotinsky oscillator.

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furthermore be of considerable technological importance in relation with chemical engineering(7) and materials science.

Besides chemical waves (which are discussed in this volume by C. Vidal), chemical structure formation should also occur in the form of stationary space-periodic concentration patterns, as proposed by Turing.(8) Here the characteristic times of the autocatalytic chemical reactions are such that complete mixing in the reaction vessel cannot occur through diffusion solely. This allows the uniform distribution of reagents, characteristic of thermodynamic equilibrium, to become unstable at a finite distance from it, leading to a spatial patterning whose realizations in reaction-diffusion models were extensively studied by Prigogine and his collaborators.(9)

Although the analysis of theoretical models (Brusselator,(9) Oregonator,(10) etc.) corroborates the possibility of existence of such structures, few if any real stationary patterns have been experimentally reported. Besides that in yeast extracts,(11) the examples arise in systems related to the Belousov–Zhabotinsky reaction.(12–14)

Interpretation of the origin of these "mosaic patterns" is unfortunately made ambiguous because of the possible presence of convective and interfacial effects.

Some causes behind these experimental difficulties are that:

1. The analysis of the theoretical models implies that the diffusion coefficients of the activator and inhibitor substances should differ sufficiently from one another.(9,10) However, this is not usually the case among the typical species playing a role in experimental systems presenting organized chemical behavior. Indeed it is worth noting that pseudo-Turing structures have been observed, and their stability studied, in networks of mutually connected, continuous, well-stirred reaction cells where mass transfer coefficients may easily be controlled.(15)

2. For obvious reasons, evidence of chemical structures should be found in unstirred batch reactors (no feeding) in order to get rid of the interference of hydrodynamic fluxes. Therefore, because of the consumption of reagents, the system drifts in parameter space and the structures may at best appear as transients, which makes their detailed study very difficult. The study of waves (see the contribution by C. Vidal in this volume) is plagued by the same problems.

3. The avoidance (or the control) of convective currents may prove difficult in fluid-phase multicomponent reactive systems, as we shall discuss below.

In the following section, we discuss the patterns generated at interfaces, often by light irradiation. They are the chemical visualization of convective currents created either by evaporative cooling, for instance, or by the chemical processes through multidiffusion phenomena. In Section 3, the influence on a Turing instability of controlled convective flows in thin channels is discussed. This leads to the conclusion that simple Turing structures should be hard to obtain as such in fluid systems and that they might be easier to find in other contexts, some of which are presented.

2. CONCENTRATION PATTERNS GENERATED AT INTERFACES

A rich variety of processes have been shown to produce spatial structures at liquid interfaces. In a pioneering work, Möckel(16) observed the appearance of inhomogeneous concentration stripes while irradiating the system KI/CCl_4/starch in water (Fig. 1). Thereafter the generality of this phenomenon has been proved by Kagan *et al.*(17) and Gimenez and Micheau,(18) who reported the formation of photochemical structures for a

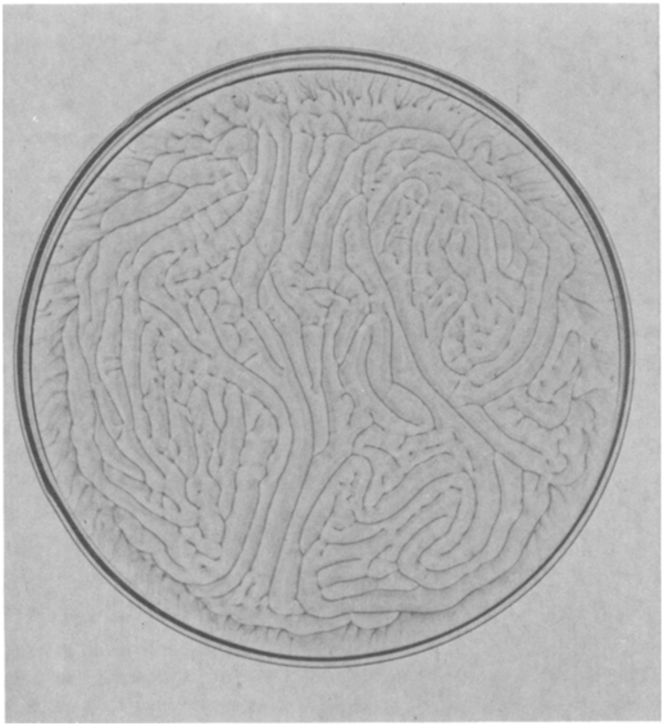

Fig. 1. Structures obtained when irradiating with UV radiation a solution of KI/CCl_4/starch in water. (Courtesy J.-C. Micheau.)

large number of photochromic (reversible) and chromogenic (irreversible) compounds. As a result, because of the diversity, the nature of the chemical reaction does not seem to play an important role in the onset mechanism. Moreover, photochemistry is even not an essential factor; patterns are also formed when gases diffusing through a liquid interface react with the solute or when the reagents in the same solvent are separated by a dialysis membrane.(19) In all these cases a thin layer of product first appears in the vicinity of the interface and then breaks down into inhomogeneous zones. The patterns do not form when convective motion is prevented from appearing; for instance, when the reactions are carried out in a gel or in a very thin layer. The interpretation of this class of spatial phenomena has led to a controversy about whether the convective motions are generated by the chemical reactions or whether the chemical reactions merely serve to visualize preexisting hydrodynamic currents.

It now appears that there is probably not a unique mechanism of patterning and that one must clearly distinguish experiments conducted in open air from those performed in a Petri dish sealed with a glass cover.

2.1. Preexisting Convection (Experiments in Open Air Conditions)

The mechanism of photolysis of the halogen compounds first studied by Möckel and Kagan *et al.* was complex and largely unknown. Afterward, Micheau and Gimenez studied simpler photochemical reactions with known mechanisms(18): first- or second-order reactions. These experiments revealed the existence of patterns whether the irradiation was performed from above or from below. The following facts strongly suggest a hydrodynamic origin for these patterns(20):

1. The wavelength of the structure increases almost linearly with the thickness of the layer.
2. The structure disappears when the Petri dish is covered and it reappears after removal of the plate.
3. Photographs of striations have been obtained before irradiation (prepatterns) using the Schlieren technique. Moreover, the patterns revealed under illumination coincide with the prepatterns.

Consequently in these experiments chemistry only serves to trace preexisting convective structures driven by the mechanism of evaporative cooling. The evaporation of the solvent effectively cools the liquid surface, thereby inducing a potentially unstable density gradient in the layer and also initiating surface tension variations. Both effects contribute to the destabilization of the conduction state. For example, linear analysis in the ideal case of a constant, time-independent temperature gradient yields the following threshold relation for the onset of convection (in the case of small Biot numbers):

$$(\mathrm{Ra}/R_0) + (\mathrm{Ma}/M_0) = 1 \tag{2.1}$$

The values of R_0 and M_0 depend on the boundary conditions. The Rayleigh (Ra) and Marangoni (Ma) numbers are two different dimensionless realizations of the same temperature difference ΔT between the bottom and the upper plate:

$$\mathrm{Ra} = \frac{\alpha g d^3\, \Delta T}{K\nu}, \qquad \mathrm{Ma} = -\left(\frac{d\sigma}{dT}\right)\frac{d\,\Delta T}{K\nu\rho} \tag{2.2}$$

where d is the layer thickness, α is the thermal expansion coefficient, ν is the kinematic viscosity, K is the heat diffusivity, ρ is the density, and σ is the surface tension. From (2.2), surface tension effects are seen to dominate in thin layers, while for thicker layers convection is driven mainly by the buoyancy effects. Concurrently a modification in the morphology of the pattern is observed. At small d, surface tension forces tend to favor hexagons (cellular structures), whereas rolls (vermiculated structures) characteristic of buoyancy forces become more and more the rule as d is increased. Both structures sometimes coexist, giving rise to quite irregular patterns. The signature of this transition appears as a discontinuity, with hysteresis effects, in the plot of the yield of the photochemical reaction versus the layer thickness.

Deformations of the free surface accompany these convective motions and modify the threshold conditions (2.1). The profile is concave when d is small and convex when the layer is deep. These ondulations can lead via the Beer–Lambert law to a spatial variation of the light intensity transmitted through a solution. For instance, Müller and Plesser(21) reported spatial modulations in the absorbance of an aqueous solution of reduced nicotinamide adenine dinucleotide (NADH), which is an important intermediate of glycolysis. These observations help to explain the results obtained when a light beam with a wavelength specific for the absorption of NADH passes through an open-to-air layer of yeast extract exhibiting glycolytic oscillations. Structures form and disappear in interval of several minutes.(11,21) They result from a strong coupling between the chemical oscillations and the network of convective cells induced by the Bénard–Marangoni instability. The convective patterns can be detected whenever the NADH concentration passes through a maximum.

Another example of the influence of convection is provided by the oscillatory behavior observed during fluorescence or phosphorescence analysis. When the vertical cell containing some organic species is illuminated at constant intensity, periodic and aperiodic oscillations (photochemical chaos) in the emitted light have been reported. It has been clearly shown that the temporal behavior is not driven by the photochemical reaction, but by time-dependent convection taking place in the vertical tube when the corresponding Rayleigh number exceeds some critical value $\mathrm{Ra}^{\mathrm{osc}}$ which strongly depends on the Prandtl number $\mathrm{Pr} = \nu/K$. The temperature gradient is generated here either by evaporative cooling in open tubes or by the heat produced by the lamp of the spectrofluorimeter. Here also photochemistry merely serves to probe hydrodynamic processes.(22,23)

These studies point out that considerable care must be taken to ensure that all preexisting hydrodynamic motions are properly taken into account in the interpretation of the symmetry-breaking instabilities observed in chemical systems.

On the other hand, photochromic compounds provide a powerful tool to characterize hydrodynamic flow.(24) For instance, the study of the drift and of the deformation of a grid obtained by photochemical printing allows one to determine the velocity field and the components of the velocity gradients in the solution. Let us also note that helicoidal convective patterns have been observed in experiments performed in vertical photochemical cells.

2.2. Convective Motion Induced by Chemical Reactions at Interfaces

Growing experimental evidence suggests that chemical reactions can also promote hydrodynamic movements(19); a well-documented example is provided by photochemical reactions in closed Petri dishes. A large number of possible mechanisms have been proposed to explain the onset of these structures. Among them the case of surface-catalyzed reactions leading to adverse density gradients has been analyzed using the methods developped for the study of the classical Rayleigh–Bénard problem.(25)

We now briefly discuss the role of double diffusion, which has long been recognized as the major mechanism leading to convection in multicomponent systems.(26) Because these patterns can form even in the presence of a hydrostatically stable density gradient, we think that this instability provides the "engine" of many chemically driven convective motions in isothermal systems. Inhomogeneous chemical reactions are indeed supposed to create only weak density gradients via the expansion coefficients. For the sake of simplicity we consider a photochemical reaction taking place in the absorbing layer of a shallow solution:

$$A + h\nu \rightarrow B$$

Furthermore, we assume that $D_A > D_B$, where D_i is the molecular diffusion constant of the species i. This reaction induces in the layer opposite gradients of reactant and product, which can then generate various instabilities.(27)

2.2.1. Fingers. For the idealized conditions corresponding to constant, time-independent gradient of concentration and free–free boundary conditions for the velocity, linear stability analysis yields the following threshold for the onset of the stationary patterning instability:

$$R_A + R_B = -R_c \qquad (R_c = 657) \tag{2.3}$$

where R_i is the solutal Rayleigh number:

$$R_i = \frac{\alpha_i g d^3 \Delta x_i}{\nu D_i} \tag{2.4}$$

Here x_i is the mass fraction of the constituent i and

$$\alpha_i = \frac{1}{\rho}\left(\frac{d\rho}{dx_i}\right)_T = \rho(v_s - v_i)$$

where v_i is the specific volume.

Beyond this threshold, fingers perpendicular to the interface develop, tending to increase their length and width. In relatively thick layers, long fingers can appear. In that case the following criterion for the formation of finite-amplitude fingers can be derived by assuming that the motion is essentially vertical(28):

$$\alpha_A\left(\frac{\partial x_A}{\partial z}\right) D_B + \alpha_B\left(\frac{\partial x_B}{\partial z}\right) D_A > 0 \tag{2.5}$$

When the system is irradiated from above one has

$$\partial x_A/\partial z < 0 \quad (\Delta x_A > 0); \qquad \partial x_B/\partial z > 0 \quad (\Delta x_B < 0)$$

If we furthermore suppose that the global density gradient is small, $\Delta x_A = \Delta \approx -\Delta x_B$, Eqs. (2.3) and (2.5) become

$$\alpha_B D_A > \alpha_A D_B \tag{2.6}$$

Fingers can then appear when the reactant diffuses more rapidly than the product. In this network, the upgoing fingers lose reactant to the downward-moving fingers, making the former less dense. Indeed, very little product (B) is transferred between the fingers, because of the lower diffusivity of B. It is this density difference that triggers the motion even though the mean density gradient over the fingers is hydrostatically stable, as is the case when the expansion coefficient of the reagent is larger than that of the product: $\Delta\rho = \Delta(\alpha_A - \alpha_B) > 0$. This situation seems to occur in the case of the photoreduction of ferric ion (irradiation from above) by oxalic acid followed by complexation with potassium ferricyanide to produce the soluble blue dye known as Turnbull's blue. Avnir and Kagan⁽²⁹⁾ have shown that spectacular patterns can form in a dish covered with a plate to avoid evaporation. In that case the diffusion coefficient of Turnbull's blue (the product), $D_B = 2.5 \times 10^{-7}$ cm²/sec, is much smaller than that of the reactant, $D_A = 10^{-5}$ cm²/sec, and the expansion coefficient of the blue dye is also lower than that of the starting species ($\alpha_B \lesssim \alpha_A$); therefore, the conditions (2.5) and (2.6) can easily be satisfied in this experiment.

The presence of a stabilizing temperature gradient resulting, for instance, from the absorption of the radiation by the product (a process that fixes the initial position of the absorbing layer) does not modify these conclusions. Indeed, since the thermal diffusion coefficient is larger than the solutal diffusion constant, this gradient reinforces the destabilizing mechanism leading to the formation of the fingers.(26)

2.2.2. Oscillatory Convection. In the case of the idealized conditions described above, the marginal stability conditions for the onset of oscillatory convection is

$$\frac{R_B S_A^2}{(S_A + S_B)(1 + S_A)} + \frac{R_A S_B^2}{(S_A + S_B)(1 + S_B)} = -R_c \tag{2.7}$$

In most liquids the Schmidt numbers $S_i = \nu/D_i \gg 1$ are much larger than 1 and the condition (2.7) then becomes

$$\frac{1}{(D_A + D_B)}(D_A R_A + D_B R_B) = -R_c \tag{2.8}$$

When the system is irradiated from below one has $\Delta x_A < 0$ ($R_A < 0$); $\Delta x_B > 0$ ($R_B > 0$), and since these gradients are generated by the photochemical reaction, mass conservation implies that $\Delta x_B \approx -\Delta x_A = \Delta$.

The condition (2.8) therefore reduces to

$$\alpha_A - \alpha_B > 0 \qquad \text{or} \qquad v_B - v_A > 0 \tag{2.9}$$

As a result, oscillatory convection is possible in this configuration when the volume increases during the reaction. Again these conditions are satisfied in the case of the photoreduction of ferric ion by oxalic acid when the Petri dish is illuminated from below.(29)

The nature of the periodic structures that appear beyond the overstability threshold has been the subject of many experimental and theoretical investigations.(30) In a laterally infinite system standing waves are unstable to traveling waves and states consisting of convective rolls that move laterally in the plane have indeed been observed. The effects of finite geometry play an important role in this competition between traveling and standing waves and they can lead to interesting spatial structures, including multistability of confined traveling waves.(31)

In the photochemical experiments, time-dependent convection has not yet been observed. For some range of the parameters the instability corresponding to the threshold (2.7) can also lead to finite-amplitude, nearly steady-state convection.(30) Oscillatory convection can thus easily be missed by stepping too quickly through the threshold. It would be interesting to perform further experimental work on photochemical systems satisfying the condition (2.9) because they provide one of the few systems where dissipative waves can emerge at a primary bifurcation point.

3. CHEMICAL INSTABILITIES AND CONVECTION

We consider a physicochemical system confined between two plates separated by $2l$ and discuss the effect of the following simple flows on the Turing instability:

1. Shear flow:

$$v_x = \alpha z$$

2. Poiseuille flow:

$$v_x = v_m[1 - (z/l)^2]$$

3. Time-independant periodic flows:

$$v_x = -v_m q_c^{-1} \frac{\pi}{2l} \sin(q_c x) \cos\left(\frac{\pi z}{2l}\right)$$

$$v_z = v_m \cos(q_c x) \sin\left(\frac{\pi z}{2l}\right)$$

where v_m is the maximum velocity.

These convective motions greatly enhance the mass transfer in the direction of the flow. Since the work of Taylor,(32) it is well known that this enhancement can often be described by an effective diffusion coefficient K which takes the general form

$$K = D + C/D \tag{3.1}$$

where $C = 2\alpha^2 l^4/15$ for the shear flow, $C = (16/945)\, l^2 v_m^2$ for the Poiseuille flow, and $C = v_m^2 (2l)^2/3\pi^2$ for the periodic flow.(33)

This last expression is valid at low Peclet numbers $\mathrm{Pe} = v_m l/D < L/l$, where L is the longitudinal characteristic length. Because ordered flows can be well controlled only in cells of small aspect ratio, it is experimentally difficult to reach values of Pe less than 10 in the case of periodic flows. For higher values of Pe the effective diffusion coefficient in this case takes the form(34)

$$K = \gamma (D v_m l)^{1/2} \tag{3.2}$$

where γ is a numerical constant. This expression is related to the flux across the narrow boundary layers that develop between adjacent convective rolls.

When chemical reactions are taken into account, the parameters become numerous and a complete analysis has not been done even for idealized models. In the vicinity of a chemical instability one generally distinguishes the long relaxation time of the marginal mode t_c and the characteristic times t_r associated with the other rapid relaxation processes. When the following conditions are satisfied

$$t_c \gg t_r \gg l^2/D \tag{3.3}$$

the concentrations of the species are essentially constant over a section of the channel on the chemical time scales. In this regime and in a coordinate system moving with the mean velocity, the system is described by a reaction-diffusion model including the anisotropy resulting from the renormalization of the diffusion coefficients in the direction of the flow [cf. Eqs. (3.1) and (3.2)].

For instance, the kinetic equations of the Brusselator model(8) become, in the regime defined by the conditions (3.3),

$$\begin{aligned}
\frac{\partial X}{\partial t} &= A - (B+1)X + X^2 Y + D_X \nabla_\perp^2 X + D_X^{\mathrm{a}} \frac{\partial^2 X}{\partial x^2} \\
\frac{\partial Y}{\partial t} &= BX - X^2 Y + D_Y \nabla_\perp^2 Y + D_Y^{\mathrm{a}} \frac{\partial^2 Y}{\partial x^2}
\end{aligned} \tag{3.4}$$

where $\nabla_\perp^2 = \partial^2/\partial x^2 + \partial^2/\partial y^2$ and A and the control parameter B are kept constant. $D_i^a = K_i - D_i$ is a measure of the anisotropy induced by the flow. As in the isotropic case, this model displays a Turing instability.(35) Indeed, the uniform solution $X_0 = A$, $Y_0 = B/A$ becomes unstable for

$$B > B_c = [1 + A\eta(\phi)]^2 < 1 + A^2 \tag{3.5}$$

where

$$\eta(\phi) = \left(\frac{D_X + D_X^a \cos^2 \phi}{D_Y + D_Y^a \cos^2 \phi}\right)^{1/2} \tag{3.6}$$

for fluctuations of wavenumber

$$q_c^2(\phi) = A[(D_X + D_X^a \cos^2 \phi)(D_Y + D_Y^a \cos^2 \phi)]^{-1/2} \tag{3.7}$$

and making an angle ϕ with the flow. The uniaxial anisotropy introduced by the flow thus selects a preferred orientation for the critical wavevector.

If the system can already present a Turing instability when $v_x = 0$ (i.e., $D_X < D_Y$), then the flow will align the axis of the concentration pattern rolls in a direction parallel to the velocity, but the threshold and wavevector

$$B_c = \left[1 + A\left(\frac{D_X}{D_Y}\right)^{1/2}\right]^2, \qquad q_c^2 = A(D_X D_Y)^{-1/2}$$

are not modified by the convective motion.

On the other hand, when the molecular diffusion coefficient of the inhibitor (Y) is smaller than that of the activator (X), the Turing instability is impossible for $v_x = 0$ as a Hopf bifurcation already occurs at $B_c = 1 + A^2$. In that case flows at small Pe numbers (3.1) can induce a Turing instability leading to a concentration pattern, the axis of which is perpendicular to the flow. The corresponding threshold becomes

$$B_c = \left[1 + A\left(\frac{D_Y}{D_X}\right)^{1/2}\right]^2$$

and the wavenumber depends on the velocity according to

$$q_c^2 = (A/C)(D_X D_Y)^{1/2}$$

An experiment devised to illustrate this latter effect would take place in a flow through a channel connected with a well-mixed flow reactor. In the reactor the system is in a steady state (in this zone the mass transfer coefficients are all equal to the turbulent diffusion coefficient). When flowing through the channel the mass transfer coefficients are renormalized differently [see Eq. (3.1)] and could then induce a Turing instability. The corresponding chemical pattern would move with the mean velocity of the fluid. By magnifying the differences between the diffusion coefficients, laminar flows could provide favorable conditions for the onset of spatial chemical structures through the Taylor diffusion phenomenon. However, if the system brings more then two determining species into play, the instability could lead immediately to traveling waves. Experiments of this type, exhibiting spatial structure formation, have been reported by Marek and Svobodova.(36)

When condition (3.3) is replaced by

$$t_c \gg l^2/D \gtrsim t_r \tag{3.8}$$

the renormalized diffusion coefficient then contains mixed chemodiffusive contributions, as shown for a shear flow.(37) In this case the flow is also able, in particular circumstances, to produce a diffusive instability leading to a chemical pattern, even when no such pattern is obtainable in the absence of convection.

4. CONCLUSION

It thus seems that in fluid phases, because of the possible occurrence of uncontrolled convective currents, Turing structures would be difficult to produce. Other structures, such as the precipitation patterns(38) (Liesegang rings), are also easier to produce in a medium such as a gel where convection is absent. Such a medium or one with controlled convection (as in Section 3) has not been thoroughly investigated with respect to such chemical structures.

However, various structures that appear in quite different systems may have some relation with the problem we discuss here.

Indeed, crystalline solid solution and metals under particle irradiation may respectively lead to solute clustering(39) or void-lattice formation.(40) Also, mixed conducting–superconducting(41) and metal–insulator(42) phases under nonequilibrium conditions have been reported. In these four cases, the creation, annihilation, and interactions of interstitials, vacancies, or elementary excitations may all be described by phenomenological laws mimicking nonlinear chemical kinetics. Dislocation patterning in fatigued metals may also be described in terms of dynamical instabilities arising in pseudochemical kinetic schemes involving populations of defects.(43) Furthermore, a nonlinear, passive optical system driven by a coherent plane-wave stationary beam may support structures in the transverse profile of the transmitted beam through a diffraction-driven instability.(44)

As a last example, we cite the segregation under steady source conditions that occurs when the very simple, diffusion-limited reaction(45) $A + B \rightarrow 0$ takes place on a fractal surface (here 0 means that the product of the reaction desorbs instantaneously).

In conclusion, the unifying view of the concept of dissipative structures introduced by Prigogine has brought many gratifying successes in the understanding of the fundamental aspects of organization far from equilibrium. These ideas are now making their way in more applied situations.

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