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Competition of intrinsic and topographically imposed patterns in Bénard–Marangoni convection

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The structure of Bénard–Marangoni convection cells can be controlled by periodic topographic patterns on the heated surface that generates the convection. When the periodicity of the topographic pattern matches the intrinsic periodicity of the convection cells, a convective pattern is formed that is 1:1 commensurate with the topographic pattern. Arrays of hexagonal, square, and triangular convection cells were generated over the appropriately designed topographic patterns, and visualized by infrared imaging. For imposed patterns with periodicity in two dimensions, as the ratio of the intrinsic and perturbing length scales changes, the pattern of the convection cells shows sharp transitions between different patterns commensurate with the imposed pattern. For imposed patterns with periodicity in one dimension (i.e., lines) the convection cells use the unconstrained dimension to adapt continuously to the external perturbation. Topographically controlled convection cells can be used to assemble microscopic particles into externally switchable regular lattices. © *2001 American Institute of Physics.* [DOI: 10.1063/1.1384473]

Fascination with the spatiotemporal patterns that arise spontaneously in nonequilibrium systems has stimulated detailed experimental and theoretical investigations, and resulted in a sound understanding of many aspects of the formation of intrinsic patterns—those that arise under ideal, uniform conditions.[1,2] Real patterns—those that arise in the presence of defects and boundaries—are also important, but difficult to model. Here, we consider a subset of real patterns—those for which the extrinsic forcing is simple and periodic—and show both that these patterns can be understood within a simple conceptual framework, and that this understanding can be used to control them (e.g., switching between different convective patterns).

Many extensively studied equilibrium systems display competition between intrinsic and imposed periodicity and symmetry, e.g., in formation of self-assembled monolayers,[3] epitaxial growth of semiconductors on silicon crystals,[4] and adsorption of molecules on crystalline surfaces.[5] The general features of such systems can be understood within the framework of the Frenkel–Kontorowa model, which describes the free energy of the system as a function of the positions of the adsorbed atoms.[6] A few groups have studied nonequilibrium systems that show competition between intrinsic and imposed periodicity and symmetry.[7–11] These groups modulated the driving force of Rayleigh–Bénard convection and electroconvection of nematic liquid crystals, but in all cases, the ''intrinsic'' convection patterns were rolls and the imposed perturbations varied in only one direction, aligned with the orientation of rolls; the competition of length scales was thus limited to one dimension.

Here, we describe a well-controlled, effectively two-dimensional (2D) nonequilibrium system in which two-dimensional patterns can be controlled. This system is based on Bénard–Marangoni[12] convection; this type of convection forms 2D patterns of convection cells when unconstrained. This system displays two types of behavior when the driving force of the convection is modulated by topographic perturbations (arrays of raised posts and lines) on the heated surface: (i) When the modulation varies in two directions, the pattern of convection cells undergoes sharp transitions between different commensurate 2D patterns as the ratio of intrinsic size to imposed size varies; and (ii) when the modulation varies in only one direction, the pattern of convection cells continuously adapts to changes in the ratio of the intrinsic to the imposed length scale by changing size along the unconstrained direction.

The required patterned surfaces were fabricated by molding thermally conductive epoxy resin (Cast-Coat Inc.) on silicon wafers (50 mm diam) using soft lithography.[13,14] A thin layer (0.3–2 mm) of silicone oil (100 cSt) was heated over these patterned surfaces to 90–145 °C in ambient air (25 °C), and the resulting convective patterns were visualized with an infrared (IR) camera (model PM-290, FLIR Systems). Figures 1, 2, and 3 show IR images in which the

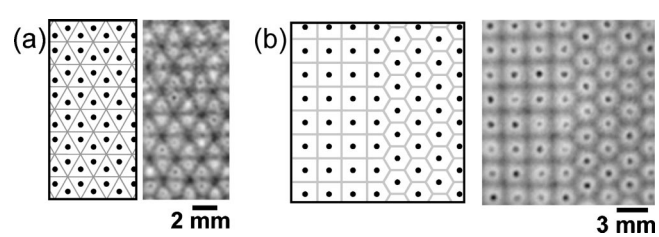

FIG. 1. IR images of convection cells formed by a thin layer of silicone oil [depth of oil $d = 0.39$ mm (a) and 0.79 mm (b)] heated in air to 145 °C (a) and 120 °C (b) over arrays of posts. The heights of the posts are 0.2 mm (a) and 0.25 mm (b); the diameters of the posts are 0.33 mm (a) and 0.50 mm (b); and the distances between adjacent posts are 1.25 mm (a) and 2.00 mm (b). There is a post (the dot in the schematic diagram) under each convection cell in these images.

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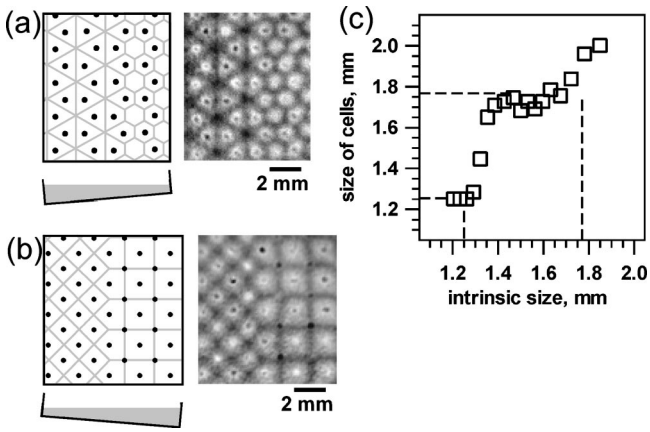

FIG. 2. (a) and (b) IR images of convection cells over patterned surfaces undergoing transitions in size to (a) smaller length scale and (b) larger length scale than the length scale of the imposed pattern. The transitions are due to a gradient in the intrinsic lenth scale induced by tilting the container by 0.1°–0.2° (a schematic representing the depth of oil is shown). The distance between the adjacent posts is 1.25 mm, the height of the posts is 200 μm, the diameter of the posts is 330 μm, and the temperature is 145 °C. (c) Dependence of the observed size of square cells shown in (b) on the intrinsic size of cells. Dashed lines correspond to the distance between the adjacent posts [1.25 mm, lattice parameter of the unswitched pattern in (b)] and to the distance between the diagonally adjacent posts [1.77 mm, the lattice parameter of the switched pattern in (b)].

light regions map the warm fluid rising in the centers of the cells, and the dark regions map the cooler fluid descending at the edges of the cells. The raised features are slightly reflective to ambient IR and, therefore, appear cold (black) when visible through the oil.

In the usual Bénard–Marangoni convective system, where the fluid layer is of uniform depth, the convection starts when the system reaches the critical value of a nondimensional driving force, the Marangoni number, $\mathrm{Ma} = (d\sigma/dT)(\Delta T d)/(\rho \eta k) \cong 80$.[12] Here, $d\sigma/dT$ [N/(mK)] is the coefficient that describes the decrease in surface tension with increasing temperature, ΔT (K) is the temperature difference between top and bottom surfaces, d (m) is the depth of the oil, ρ (kg/m^3) is the density of the oil, μ [kg/(ms)] is the viscosity of the oil, and κ (J^2/s) is the thermal diffusivity of the oil. The convection is driven by variations in surface tension of the fluid as a function of temperature: liquid is pulled from warmer regions of the liquid–air interface into cooler regions by the higher surface tension of the cooler regions. In our convective system, where the fluid layer is not of uniform depth, convection exists for all nonzero values of the Marangoni number (subcritial convection).

Near the critical value of the Marangoni number, the

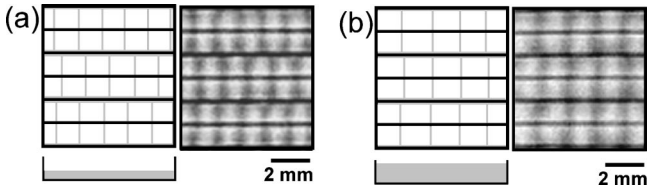

FIG. 3. IR images of convection cells formed over lines at small [1.73 mm, (a)] and large [1.90 mm, (b)] depths of oil. These cells can adapt to changes in intrinsic length scale by changing their horizontal size and translating in the unconstrained dimension, while their size and position remain constant in the orthogonal (constrained) dimension. The height and width of the lines is 0.63 mm, the spacing between the centers of the lines is 2.5 mm, and the temperature is 90 °C.

Bénard–Marangoni convection cells that form over an unpatterned, flat surface ("intrinsic" cells) have hexagonal symmetry, and their size [defined here by $(\mathrm{area})^{1/2}$] is directly proportional to the depth of oil.[12] When these cells form under the same conditions but over a periodically patterned surface, their shape and size depend on two factors: (i) the height of the topographic pattern relative to the depth of oil and (ii) the period and symmetry of the topographic pattern relative to the intrinsic period and symmetry. We controlled the strength of the interaction between the intrinsic and the imposed patterns by controlling the height of the raised features relative to the depth of the oil, and all results presented in this letter are in the limit when this interaction is dominant. We controlled the characteristics of intrinsic relative to imposed patterns by changing the depth of oil, and thus the intrinsic pattern.

When the intrinsic periodicity of the convection cells is close to the periodicity of the topographic pattern, both the dimensions and symmetry of the observed pattern are locked to those of the imposed pattern (a 1:1 commensurate structure, Fig. 1). Raised features modulate both the temperature gradient in the system and the shearing interaction of the fluid with the solid surface.[15] In 1:1 commensurate structures, the convection cells tend to form with the center of each cell—where fluid is ascending—above the raised surface features, because the fluid layer directly over a post is thinner than over the flat points of the surface, and the surface of the fluid film is correspondingly hotter; these hot spots become upwellings in the pattern of convection cells. We tiled the surface with convection cells of all three possible perfect polygonal shapes: Triangles (which have not previously been observed in Bénard–Marangoni convection), squares (previously observed only at higher Marangoni numbers[16]), and hexagons. In Fig. 1(b) the hexagonal and square cells are separated by a single row of pentagonal cells; this observation indicates that the perturbations imposed by the topographic pattern are local, in the sense that the shape of cells is determined by the interactions with adjacent posts only.

We found that the convection cells undergo a sharp transition as their intrinsic period is changed relative to the period of the topographic pattern. It was most convenient to visualize these transitions in a system designed to have a gradient in the intrinsic period of the cells. An appropriate gradient is easily generated in the depth of the oil, simply by tilting the container by 0.1°–0.2° from the horizontal (Fig. 2). As the dimensions of the intrinsic convection cells decreased to below the period of the lattice of posts, the cells underwent a transition from triangles to a lattice of hexagons with an area smaller than that of triangles [Fig. 2(a)]. This lattice required that some cells be generated between posts. Cells that were formed over a square lattice of posts formed a 1:1 commensurate lattice when the intrinsic size was close to the lattice spacing [Fig. 2(b)]. As the intrinsic length scale of convection cells increased to above the periodicity of the square lattice of posts, the cell structure underwent a sharp transition to a second square lattice of cells with twice the area. Figure 2(c) shows how the sizes of the cells over a square lattice of posts changed when the depth of oil was increased, and compares these sizes to the intrinsic size (the

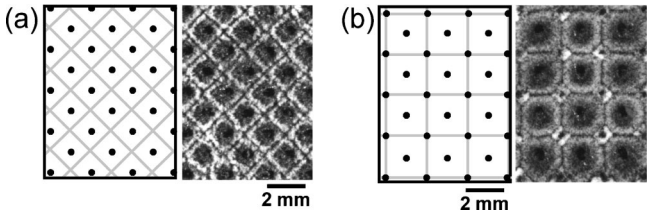

FIG. 4. Photographs of the switchable assembly of ragweed pollen grains (20 μm) generated with convection cells undergoing a transition shown in Fig. 2(b). This transition was triggered either by changing the level of oil d or changing the remperature gradient ΔT. Most of the pollen grains ($\rho \approx 0.2$ g/cm^3) are floating on the surface of the oil ($\rho \approx 0.9$ g/cm^3), although some are transported by the convective flow below the surface of the oil.

size of the intrinsic cells formed at the same ΔT and d).

In the examples shown in Figs. 1 and 2, the external 2D pattern completely controls the convection cells—both position and shape. By introducing one-dimensional constraints—arrays of raised lines instead of posts—we were able to observe the influence of perturbations that removed only one of the two horizontal degrees of freedom open to unconstrained cells (Fig. 3). Both the size and position of the cells formed over lines are constrained in the dimension normal to the lines; in the perpendicular horizontal direction, the dimension of the cells is determined by the height of the oil, and their position is determined by the packing of neighboring cells. In this system, when the depth of the oil was increased [Fig. 3(b)], the convection cells adapted by using the unconstrained dimension to change their size and position only in that dimension.

We believe that well-defined, nonlinear systems that show large responses to small external influences—such as the transitions between cell sizes observed with small changes in the depth of the oil ($<10\,\mu$m) in Fig. 2(b)—could be useful for sensing these influences, and for generating reconfigurable structures. For example, Fig. 4 shows 20 μm pollen particles floating at the surface of silicone oil patterned by Bénard–Marangoni convection using the same topography as in Fig. 2(b). The pattern of particles can be switched reversibly (a)$\leftrightarrow$(b) by any perturbation that affects the intrinsic size of the cells: for example, a change in the temperature gradient $\Delta T = 5\,°$C,[12,17] or a change in the thickness of the oil $d \cong 30\,\mu$m.

This system of convection cells allows the exploration of patterns in two-dimensional, nonequilibrium systems in which there is a competition between the imposed and intrinsic patterns. Both the experiments and the fabrication of topographically patterned surfaces are straightforward in this system. The IR imaging technique should allow direct measurements of the rate of energy dissipation (heat flux through the system) for comparison of constrained and unconstrained convection cells. One limitation of our system is that the intrinsic length scale and the relative strength of perturbation change simultaneously as the level of oil changes. A fully 2D theoretical model of competition between intrinsic and imposed convective patterns could allow one to tailor the pattern formation and the transitions for optimal sensitivity to external parameters. The richness of behavior of the convective system described in this letter would also allow for the fine tuning of the theoretical tools. We hope that this system will stimulate more detailed, quantitative theoretical investigation, and will contribute to understanding pattern formation under nonuniform conditions.

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