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LETTER TO THE EDITOR

Turbulent crystals in macroscopic systems

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Abstract. We demonstrate circumstances in which gradient systems can minimize their free energies with a spatially turbulent planform.

When subject to external stresses, spatially extended, continuous, dissipative systems far from equilibrium can undergo a series of symmetry breaking phase transitions in which certain planforms or patterns are preferentially amplified. Near the transition, the nonlinear competition between degenerate states, each of which is linearly amplified at the same rate, can lead to stationary periodic patterns such as rolls [1], squares, rhombi [2], hexagons [3] which tile the plane if the system is two-dimensional or to equivalent periodic crystal structures in three dimensions. Such patterns are observed in convecting fluids, liquid crystals, counterpropagating light beams, Raman lasers, and in a whole variety of natural phenomena. Very recently, standing wave quasiperiodic patterns, analogous to a two-dimensional dodecahedral quasicrystal have been observed by Edwards and Fauve [4] in a modification of the Faraday experiment. Again, the pattern is dynamically generated and sidewalls play no role.

In each of these examples, the degeneracy is infinite and is simply a reflection of the continuous rotational symmetry associated with an infinite planar geometry that is a good approximation when the horizontal dimension of the experiment is much larger than the pattern wavelength. The horizontal structure of all modes which are equally amplified is $\exp i\boldsymbol{k}\cdot\boldsymbol{x}$ where each $\boldsymbol{k}$ lies on a circle $|\boldsymbol{k}|=k_c$. The roll, square and hexagon patterns correspond to final states in which only a finite number of modes (respectively one, two, three) participate. The purpose of this letter is to explore circumstances in which a very large (and potentially infinite) number of modes, each of whose wavevectors lie on the critical circle, participate. From the physical point of view, the key ingredient in producing such a state is to have the nonlinear coupling between modes much weaker than the self-coupling feedback. In this way, each mode can draw more or less independently on the source of potential energy and thereby maximize (minimize) the transport properties (the free energy) of the system. In these cases, the asymptotic planforms will be spatially turbulent and spatial correlations of the fields will decay. At the end of this letter, we briefly mention two areas where a manifestation of spatially turbulent planforms might be seen.

Near onset, the fields giving rise to the pattern can be divided into two components, active and passive. The active modes, the number of which depends on the degeneracy of the system at the phase transition, draw directly on the source of external stress. Their amplitudes $\{A_j\}$, $j=1,\ldots,N$, are called order parameters. The passive modes on the other hand may be regenerated through nonlinear interactions by the active

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modes but their amplitudes $\{B_k\}$, $k=1,\ldots,M$, are algebraically slaved to those of the active modes through a graph, $B_k=B_k(A_j)$, called the centre manifold. After a short relaxation time, the dynamics of the system takes place on this manifold $B_k(\{A_j\})$. Ignoring quadratic interactions, namely cubic terms in the free energy which bias the outcome in favour of a hexagonal lattice, the dynamics is governed by the set of Landau equations

$$\frac{\mathrm{d}A_j}{\mathrm{d}t}=\mu A_j-\sum_{l=1}^{N}\beta_{jl}|A_l|^2A_j \qquad j=1,\ldots,N. \tag{1}$$

The properties of the matrix $\beta=\{\beta_{jl}\}$ determine the asymptotic state of the system. In a two-dimensional system with rotational and translational symmetry, the active set is spanned by Fourier modes $\{\exp \mathrm{i}\boldsymbol{k}_j\cdot\boldsymbol{x}\}$ where each $\boldsymbol{k}_j$ lies on a critical circle $|\boldsymbol{k}_j|=k_c$ reflecting the fact that the resulting pattern will have a preferred wavelength $\lambda=2\pi k_c^{-1}$ but no preferred direction. We represent the field $w(x,y,t)$ as 2 Real $(\Sigma_{j=1}^{N} A_j \exp \mathrm{i}\boldsymbol{k}_j\cdot\boldsymbol{x})$ where N is potentially very large. The order parameter equations (1) are invariant under arbitrary phase twists, $A_j\to A_j\exp \mathrm{i}\phi_j$, reflecting the continuous translational symmetry of the original system and the state whose stability is lost at $\mu=0$. If the matrix β is real and symmetric, (1) is gradient with free energy

$$\bar{F}=-\mu\sum_{j=1}^{N}|A_j|^2+\frac{1}{2}\sum_{j,l=1}^{N}\beta_{jl}|A_j|^2|A_l|^2. \tag{2}$$

The key idea of this letter is that, if the state that minimizes $\bar{F}$ is the one in which all modes share the resources equally so that $|A_j|^2=A^2$, $j=1,\ldots,N$, then the attractor will be a time independent but spatially random field because the phases ϕ_j are arbitrary. We call this spin-glass-like state a turbulent crystal, although we admit the name is a misnomer.

We want to emphasize again that the fact that the phases ϕ_j are arbitrary is simply a reflection of the property of translational invariance (in the horizontal directions) of the original system under study so that, if $\exp \mathrm{i}\boldsymbol{k}_j\cdot\boldsymbol{x}$ is an active mode, then so is $\exp \mathrm{i}\boldsymbol{k}_j\cdot(\boldsymbol{x}-\boldsymbol{x}_j)$ for each j. The Landau equations (1) for the order parameters are a valid approximation near onset, that is, in the small μ limit. All higher interactions are of higher order in μ. The only other terms possible at this order (or lower orders) of approximation are quadratic terms. If present, they would have the effect of biasing the outcome towards hexagonal planforms although their presence does not guarantee that outcome for all values of (small) μ and β. However, they are often absent because a large class of systems have an additional symmetry which inhibits the broken parity (high intensity in the middle, low intensity on the outside or vice versa) associated with hexagonal planforms. For example, in horizontal convection layers with symmetry about the mid-plane, the quadratic terms in (1) have zero coefficients. We stress, therefore, that the order parameter equations (1) hold for a wide class of systems near onset. The property that they are invariant under the transformation $A_j\to A_j\exp \mathrm{i}\phi_j$ is purely a reflection of the translational invariance of the solutions whose stability has been lost at $\mu=0$. The stationary planform realized for positive values of μ depends entirely on the properties of β, the matrix of coupling coefficients.

The main idea of this letter draws on several previous works. Mermin and Troian [5] and, more recently, Malomed, Nepomayaschii and Tribel'skii [6] have used similar approaches to argue how competition between the various allowable configurations leads naturally to the appearance of quasicrystalline order and other exotic planforms. Because of a rather special choice of model and a somewhat loose conversion of the free energy F into an equivalent expansion $\bar{F}$ in terms of the order parameters, the former authors conclude that planforms with a small number of wavevectors would always be favoured. The latter set of authors did not relate the choice of matrix elements β_{jl} to an underlying field theory but worked directly with (1). This was not crucial as they dealt with wavevectors with finite angular separations. However, they missed a general property of the matrix elements obvious from combinatorial arguments, namely that $\beta_{jl} \to 2\beta_{jj}$ as $\boldsymbol{k}_l \to \boldsymbol{k}_j$, which is important as the number of wavevectors increases.

The observation that random phases leads to a turbulent crystal draws on the works of Voros [7], Berry [8], McDonald and Kaufman [9] and Gutzwiller [10] who point out that a similar property obtains for high energy wavefunctions of Schrödinger's equation.

To see the circumstances under which the asymptotic states of (1) can give rise to a turbulent crystal, we consider the behaviour of the system governed by the free energy

$$F = \iint \mathrm{d}x\,\mathrm{d}y \left\{ \tfrac{1}{2}((\nabla^2+1)w)^2 - \tfrac{1}{2}\mu w^2 + \alpha_1(\nabla^2 w)^4 + \alpha_2(\nabla w)^4 + \sum_{m=1}^{n} \tfrac{1}{2}((\nabla^2+q_m^2)u_m)^2 + \tfrac{1}{2}\varepsilon^2 u_m^2 + \varepsilon r_m u_m w^2 \right\} \tag{3}$$

in the neighbourhood $\mu > 0$ of the phase transition at $\mu = 0$ at which the solution $w = u_m = 0$ loses its stability. Writing $w = \Sigma_{j=1}^{N}(A_j \exp \mathrm{i}\boldsymbol{k}_j\cdot\boldsymbol{x} + (*)) + O(\mu^{3/2})$, where A_j is of order $\mu^{1/2}$ and $u_m = \mu u_{m1} + \ldots$, it is easy to see that, for $\mu \ll \varepsilon^2$, we obtain (1) with $\beta_{jl} = \beta(\theta)$, $\theta = \pi|j-l|/N$, $\beta_{jj} = \beta_0 = \beta(0)/2$ where

$$\beta(\theta) = 24\alpha_1 + (8+16\cos^2\theta)\alpha_2 - \sum_{m=1}^{n} 8r_m^2 \times \left(\frac{\varepsilon^2}{\varepsilon^2 + (q_m^2 - 2 - 2\cos\theta)^2} + \frac{\varepsilon^2}{\varepsilon^2 + (q_m^2 - 2 + 2\cos\theta)^2} + \frac{\varepsilon^2}{\varepsilon + q_m^4} \right). \tag{4}$$

We remark on several important properties of the coupling matrix β. First, it is important to realize that the off-diagonal matrix element β_{jl} will be exactly twice the on-diagonal self-interaction element β_{jj} as $\boldsymbol{k}_l \to \boldsymbol{k}_j$. This is a general property and depends on the simple combinatorial observation that the mutual interaction $\boldsymbol{k}_j + \boldsymbol{k}_l - \boldsymbol{k}_l = \boldsymbol{k}_j$ can be realized twice as often as the self interaction. This means that rhombi with narrow angles are unlikely because if the off-diagonal elements of β dominate, rolls are the planform which minimizes the free energy. Second, the subsidiary fields u_m always lead to negative values of the coupling function $\beta(0)$. For a supercritical bifurcation (second-order phase transition) therefore, we require the presence of higher order terms in the free energy of the primary field. Nevertheless, the subsidiary fields a play a crucially important role in designing the shape of $\beta(\theta)$ necessary for turbulent crystals. As noted by Mermin and Troian [4], their contribution is largest when there is a strong triad interaction between two wavevectors $\boldsymbol{k}_j$ and $\boldsymbol{k}_l$ of the primary field w and the wavevector $\boldsymbol{q}_m$, with $|\boldsymbol{k}_j \pm \boldsymbol{k}_l| = |\boldsymbol{q}_m|$, of the mth subsidiary field. Third, contrary to the conclusion of Mermin and Troian who do not take advantage of the shape of $\beta(\theta)$ (their $\beta(\theta)$ is essentially constant), we shall see that if $\beta(\theta) < \frac{1}{2}\beta(0)$ (a situation most easily realized when the number of subsidiary fields n is large) over most of the interval $-\pi < \theta < \pi$, planforms with many wavevectors are indeed preferred. Fourth, the coupling function $\beta(\theta) = \beta(\pi - \theta) = \beta(-\theta)$ is even and symmetric about $\theta = \pi/2$, a manifestation of an underlying reflection symmetry in the original free energy. Fifth, the matrix β has circulant form with first row $(\beta_0, \beta_1, \ldots, \beta_{N-1})$ where $\beta_r = \beta(r\pi/N) = \beta((N-r)\pi/N) = \beta_{N-r}$ and each new row is obtained by cycling the previous one so that the second row is $(\beta_{N-1} = \beta_1, \beta_0, \beta_1, \ldots, \beta_{N-2})$ and so on.

Roll solutions of (1) are given by $A_1 = \mu\beta_0^{-1}$, $A_j = 0$, $j \neq 1$. Rhombi are given by $A_1 = A_r = \mu(\beta_0 + \beta_{r-1})^{-1}$, $A_j = 0, j \neq 1, r$. Squares are a special case. The turbulent crystal solution is $A_1 = A_2 = \ldots = A_N = \mu(\beta_0 + \beta_1 + \ldots + \beta_{N-1})^{-1}$. The free energies $\bar{F}$ of these states are $-\mu^2(2\beta_0)^{-1}$, $-2\mu^2(2(\beta_0 + \beta_{r-1}))^{-1}$ and $-N\mu^2(2(\beta_0 + \ldots + \beta_{N-1}))^{-1}$ respectively. More complicated solutions for which a certain subset of the N modes have each zero amplitude, and the remaining members, which are no longer distributed at equal distances along the circle, have non-zero and in general non-equal amplitudes $|A_j|$, are not easy to write down. Moreover, as far as we know, there is no known necessary and sufficient condition on the elements of the matrix β which makes the corresponding free energy $\bar{F}$ of any of the solutions mentioned above an absolute minimum†. Here we offer a sufficient condition, a recipe for constructing a coupling function $\beta(\theta)$, for which the turbulent crystal gives the lowest free energy.

The key idea is to recognize that if the matrix β is strongly diagonally dominant, $\beta_0 > \beta_r = \bar{\beta}$ say for all $r \neq 0$, then the free energy of the turbulent crystal is $(-\mu^2/2)(\bar{\beta} + (\beta_0 - \bar{\beta})/N)^{-1}$ which decreases as N increases. It is intuitively reasonable to conclude that if the intermode coupling is weak, then the solution which takes advantage of as many modes as possible will minimize the free energy because minimizing free energy is often associated with maximizing some transport property (e.g. heat) and this is best done when each mode draws on the source equally and (almost) independently. However, it is important to realize that as $N \to \infty$, modes become denser on the unit circle and since $\lim_{\theta\to 0} \beta(\theta) = 2\beta_0$ is a general property of the coupling function, some interactions will in fact be locally stronger than the self interaction, so that β in general will not be diagonally dominant. Nevertheless, the basic idea still holds and by judicious choices of ε^2, q_m^2, r_m^2 in (4) we can construct a coupling function $\beta(\theta)$ which leads to a turbulent crystal. In fact, for large n, the sum in (4) can be approximated by an integral and one can (by contour integration) find the measure (the continuum limit of r_m^2) such that the resulting function $\beta(\theta)$ has any prescribed shape. In particular, the distance over which $\beta(\theta)$ is not $\bar{\beta}$ can be made arbitrarily small. For example, we may choose the squares of the preferred wavenumbers q_m^2 for the subsidiary fields u_m to lie between 0 and 4 so that the zeros of $q_m^2 - 2 \mp 2\cos\theta$ are densely distributed along $0 < \theta < \pi$. Then the weights r_m^2 can be chosen so that the resulting measure obtained by converting the sum in (4) to an integral gives rise to a top hat shape. When this is subtracted from $24\alpha_1$ (we can take $\alpha_2 = 0$), we obtain a graph of $\beta(\theta)$, $0 \leqslant \theta \leqslant \pi/2$, which takes the value $2\beta_0$ for $0 < \theta < \theta_0$, θ_0 arbitrary, and the positive value $\bar{\beta} \ll \beta_0$ for $\theta_0 < \theta < \pi/2$. The turbulent crystal occurs when θ_0 is small. In order to make the transition at θ_0 sharp, however, ε^2 must be very small and since the analysis requires $\mu \ll \varepsilon^2$, this means the range of μ over which the turbulent crystal solutions are realized is also small. In general, the function $\beta(\theta)$ will have a much smoother shape and this coupled with the fact that $\lim_{\theta\to 0} \beta(\theta) = 2\beta_0$ means that turbulent crystals may be relatively rare. Continuing work is aimed at estimating how rare. Clearly there is much non-trivial physics involved, first in identifying the critical ingredients which lead to

† One might think that one can use the convolution form of β_{jl} (i.e. $= \beta(|j-l|(\pi/N))$) to diagonalize the double sum in (2) via a Fourier transform. It will, but the Fourier coefficients of the positive quantities $|A_j|^2$ and $|A_l|^2$ are not independent and are related in a non-trivial way, making it difficult to draw concrete conclusions.

a realization of turbulent crystals and second in obtaining a measure of how common or how rare these states may be.

Given that the turbulent crystal solution obtains, we can calculate the statistical properties of the asymptotic field

$$w_\infty(x, y) = w(x, y, t=\infty) = \sqrt{\frac{\mu}{\langle\beta\rangle}}\frac{1}{\sqrt{N}}\sum_1^N \cos(\boldsymbol{k}_j\cdot\boldsymbol{x}+\phi_j) \tag{5}$$

where $\langle\beta\rangle = (\beta_0+\ldots+\beta_{N-1})/N$. Since each ϕ_j is randomly distributed over $(-\pi, \pi)$, then, for a fixed $\boldsymbol{x}$, w has a Gaussian distribution with zero mean. Of more interest, however, is the two point spatial correlation $\langle w_\infty(x, y)w_\infty(x', y')\rangle$ which, because of rotational invariance, we can calculate by setting $y = y' = 0$. We find

$$\begin{aligned}
&\langle w_\infty(x, 0)\, w_\infty(x', 0)\rangle \\
&\quad = \left\langle \frac{\mu}{\langle\beta\rangle}\frac{1}{N}\sum_{j,l}\cos(x\cos\theta_j+\phi_j)\cos(x'\cos\theta_l+\phi_l)\right\rangle \\
&\quad = \frac{\mu}{2\langle\beta\rangle}\frac{1}{N}\sum_j\cos((x-x')\cos\theta_j)\xrightarrow{N\to\infty}\frac{\mu}{2\langle\beta\rangle}\frac{1}{\pi}\int_0^\pi\cos((x-x')\cos\theta)\,\mathrm{d}\theta \\
&\quad = \frac{\mu}{2\langle\beta\rangle}J_0(|x-x'|).
\end{aligned} \tag{6}$$

The decay for large separation distances is weak (algebraic) and oscillatory and the distance over which the correlation is successively zero is the pattern wavelength.

We end this letter with two examples of rotationally symmetric systems with infinite degeneracy and briefly discuss modifications which may lead to turbulent crystals. The grandaddy of canonical examples of pattern forming systems with infinite degeneracy is Rayleigh–Benard convection in ordinary fluids, binary mixtures and liquid crystals in large aspect ratio boxes. In single layers, complicated time dependence is often seen but complicated spatial dependence can also occur when the dynamics is non-gradient because of modulational instabilities or defect formation. However, it may also be possible to construct multilayer systems with the properties of the adjacent layers (with fields u_m) chosen appropriately to produce a coupling coefficient so that the overall dynamics is relaxational and the main layer (with field w) exhibits the kinds of behaviour suggested here. The necessary coupling to adjacent fields may be easier to realize in optical contexts. Because of an increased interest in the nature of optical turbulence, there has been much activity in recent months in wide gainband lasers in which a large number of transverse modes are excited [11]. In particular in a two-level laser (e.g. CO_2 laser) with almost planar mirrors, all transverse modes can be excited and, in order to offset the positive detuning Ω between the cavity and two-level atom frequencies, the system will elect to lase by picking a wavevector (k_x, k_y, k_z) where $\alpha(k_x^2+k_y^2) = \gamma\Omega$ (α measures diffraction, γ is the homogeneous broadening associated with the medium polarization). Again, a circle of modes destabilizes at the same value of the laser pump. Already in a single CO_2 laser, time independent and time dependent patterns (of an approximately square shape) have been observed by Glorieux *et al* [12]. It should not be too difficult to find ways to couple such systems in a way that leads to all modes on the critical circle participating in the final state.

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