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Parametrically excited quasicrystalline surface waves

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In an experiment on the parametric excitation of capillary waves (the Faraday instability) we have observed a stable standing-wave pattern with twelvefold orientational order. This "quasipattern" is analogous to a two-dimensional quasicrystal, but it occurs in a macroscopic nonequilibrium fluid-dynamical system. It is observed in containers whose side walls are of various shapes including an irregular shape, and thus it cannot be ascribed to side-wall boundary effects.

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Recently it has been proposed [1] that two-dimensional quasicrystalline patterns might arise naturally in spatially extended dissipative pattern-forming systems. This is an interesting idea because it suggests that the orientational order observed in quasicrystals is not restricted to the microscopic world but could also occur in a macroscopic, continuum-mechanical context. We report in this Rapid Communication the observation of a twelvefold "quasipattern," analogous to a two-dimensional quasicrystal, in an experiment on the parametric excitation of capillary waves. The observed quasipattern does not depend on the shape of the side walls of the container. We note that an eightfold pattern was recently reported in Ref. [2].

As discovered by Faraday [3], when an open container of fluid is forced to oscillate vertically, a pattern of standing waves can be excited on the free surface. The wave number k depends on the forcing frequency [4]. If the container is of sufficiently large horizontal dimensions, it is reasonable to pose the linear and weakly nonlinear stability problems on the infinite plane [5] and to ignore side-wall boundary effects. The linear problem has all of the symmetries of the plane and thus a circle of critical wave vectors $|\mathbf{k}| = k$ simultaneously becomes unstable as the amplitude a of the forcing oscillation passes a threshold value a_c . The pattern that develops just above the threshold is determined only by nonlinear interactions among wave vectors on or very near this circle.

The experiment described herein differs in two important ways from recent "large-aspect-ratio" Faraday experiments [6,2,7,8]. The fluid is viscous, with about 85 times the kinematic viscosity of water [9]. And the vertical oscillation is a superposition of two frequencies. We have shown recently [10] that two-frequency forcing has important consequences for the symmetries of the nonlinear stability problem.

The fluid is a mixture of 88% (by weight) glycerol and 12% water, with kinematic viscosity $v=0.85\pm0.05$ cm²/s and density $\rho=1.22$ g/cm³. The container is housed in an airtight glass and Plexiglass cover to prevent the evaporation of water. The temperature is controlled at 23±0.1 °C by a water bath and infrared lamps.

All data reported in this Rapid Communication, except as otherwise noted, are obtained with a superposition of the two frequencies 4ω and 5ω where $\omega/2\pi = 14.60$ Hz. The vertical acceleration f(t) is given by

$$f(t) = a \left[\cos(\theta) \cos(4\omega t) + \sin(\theta) \cos(5\omega t + \phi) \right], \quad (1)$$

where the phase of 4ω is zero by choice of time origin and where the angle θ , with $0^{\circ} \le \theta \le 90^{\circ}$, serves to mix the two amplitudes.

The container used for precise measurements is a cylinder of 12 cm diameter and 0.29 cm depth with a black Formica bottom and aluminum side walls. We use the brimful technique [11,8] to reduce meniscus effects [13]. Other containers of square, hexagonal, and octagonal form, and an irregularly shaped container, are used to check that the observed patterns are independent of sidewall shape. The container is attached rigidly to an electromagnetic vibration exciter.

The signal that controls the vibration exciter is gen-



FIG. 1. Pattern arising near the primary instability. For each point in this plane, i.e., for fixed ϕ and θ of the vertical acceleration $f(t) = a [\cos(\theta)\cos(4\omega t) + \sin(\theta)\cos(5\omega t + \phi)]$, instability of the flat surface is obtained by slowly increasing the amplitude a. This figure shows which pattern is observed just above the primary transition from the flat surface: L1, lines with $k \approx 8.8$ cm⁻¹; Q, twelvefold quasipattern; H, hexagons; L2, lines with $k \approx 7.5$ cm⁻¹; D, dynamic states including breaking of the surface. The quasipattern is found only very near the bicriticality (the horizontal line) for ϕ near 75°. The figure is compiled from measurements taken on a 16×17 grid of (ϕ , θ) values. Thick gray lines indicate regions where two patterns are in competition or are simultaneously present near the primary transition.

erated by a computer and digital-to-analog converter. To correct for amplitude and phase errors in the electronics, the vertical acceleration f(t) is *measured* by a piezoelectric accelerometer and a calibrated charge amplifier, and the amplitudes and phases are determined by a twofrequency lock-in technique. Values reported for a, θ , and ϕ are those measured from the accelerometer signal.

In the three-parameter space (a, θ, ϕ) instability is obtained by fixing θ and ϕ and slowly increasing *a* through the critical value a_c at which the flat surface loses stability. Note that ϕ may be chosen within $0^{\circ} \le \phi < 90^{\circ}$, since Eq. (1) has the symmetry $\phi \rightarrow \phi + \pi/2$, $t - \pi/2\omega$.

Figure 1 shows, in the (ϕ, θ) plane, the patterns obtained just above $a = a_c(\theta, \phi)$. The twelvefold quasipattern is observed for $\phi \approx 75^\circ$ near the bicriticality of two wave numbers, each corresponding approximately to the critical wave number of one of the two superposed fre-



FIG. 2. Stability boundaries for $\phi = 75^{\circ}$ in the (a, θ) plane. (a) shows the primary transition (solid line) from the flat surface F, hysteresis (dashed line), and the pattern which develops just above $a = a_c(\theta)$; instability is produced by fixing θ and increasing a as shown by the arrow. (b) shows in detail the transitions among stable states in the neighborhood of the bicriticality. H/F, Q/F, and Q/L1 denote regions of hysteresis. The thick gray line separating H and Q indicates a regime where the twelvefold quasipattern competes with hexagons, including oscillations from one to the other. The thick gray line separating Q and D indicates a "melting" regime where the quasipattern order breaks down via localized defects, which become more frequent with increasing a. Irregularities in the boundaries are due mainly to temperature fluctuations.

quencies.

Figure 2(a) shows the primary stability boundary $a = a_c(\theta)$ for $\phi = 75^\circ$. We note several features of this diagram.

Two neutral curves depart from the horizontal and vertical axes and meet at a bicritical point at $\theta = 65.5^{\circ}\pm0.5^{\circ}$. The case $\theta = 0^{\circ}$ (the horizontal axis) is the single frequency $4\omega/2\pi = 58.4$ Hz. Here the pattern observed is parallel lines of wave number $k = 7.5\pm0.3$ cm⁻¹. This differs from the low-viscosity case where



FIG. 3. Photographs of the twelvefold quasipattern. (a) Circular container, 12 cm in diameter and 0.29 cm deep. The fluid is 88% glycerol, $v=0.85\pm0.5$ cm²/s at 23 ± 0.1 °C; forcing parameters for Eq. (1) are a = 142 m/s², $\theta = 66.1^{\circ}$, $\omega/2\pi = 14.6$ Hz, $\phi = 75^{\circ}$. (b) Irregularly shaped container, 0.3 cm deep, with a flat bottom and with side walls in the shape of France; the distance Bordeaux-Geneva is 5 cm. The fluid is 82% glycerol, $v\approx0.60$ cm²/s, at ambient temperature; forcing parameters are $\theta = 65^{\circ}$, $\omega/2\pi = 31$ Hz, $\phi = 66^{\circ}$; a was adjusted to be slightly above a_c (not measured).

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theory [5] and experiment [3,6] agree that the pattern is squares [12].

For small but nonzero θ , that is, when the single frequency 4ω is slightly perturbed by 5ω , the observed pattern is still lines. For θ greater than approximately 20°, the pattern changes to hexagons, although there is no sharp changeover and hexagons, lines, and disordered patterns are observed together over a small range of θ . For θ above 30° a perfect hexagonal pattern is observed [14]. The transition to hexagons becomes increasingly hysteretic as θ is increased further.

The case $\theta = 90^{\circ}$ (the vertical axis) is also a single frequency, $5\omega/2\pi = 73.0$ Hz. Here the pattern is lines of wave number $k = 8.8 \pm 0.2$ cm⁻¹. For $90^{\circ} \ge \theta > 65.5^{\circ}$ the pattern of lines is essentially unchanged from the pure single frequency. There is no evidence of hexagons or hysteresis for this upper branch of the neutral stability boundary.

The twelvefold quasipattern is found near the bicritical point. Figure 2(b) shows in detail the transitions occurring near the bicriticality.

Figure 3 exhibits photographs of the quasipattern in two containers. We invite the reader to view these at glancing angles to observe that the quasipattern is orientationally ordered and that this ordering extends across almost the full width of each container. The pattern is visualized axisymmetrically by reflected light from 30 small incandescent lamps arranged on a circle of radius 14.5 cm. To eliminate the thirtyfold symmetry this light is passed through an axisymmetric translucent plastic diffuser in the form of an annulus of inner radius 14 cm and outer radius 18 cm. The lamps and annular diffuser surround the camera, which is 128 cm from the surface. The exposure time is 1 s, which is much longer than the standing-wave period.

Figure 4 is a computer-generated image of the twelvefold surface height function

$$\zeta(\mathbf{x}) \equiv \zeta_0 \sum_{j=1}^{12} \exp(i\mathbf{k}_j \cdot \mathbf{x})$$

for \mathbf{k}_j symmetrically arranged on a circle. The effect of the experimental visualization has been approximated by calculating $\nabla \zeta$ at each point and determining if this gradient would permit light from the annular diffuser to be reflected into the camera. The surface is assumed to move simusoidally in time, and reflected light is averaged over one period of the oscillation to yield gray levels; ζ_0 and contrast are chosen to match the photographs. Many details in the experimental photographs are also apparent in this image [15].



FIG. 4. Computer-generated image of the twelvefold surface height function $\zeta(\mathbf{x}) \equiv \zeta_0 \sum_{j=1}^{j} \exp(i\mathbf{k}_j \cdot \mathbf{x})$ with an approximation of the effect of the experimental visualization.

Stroboscopic methods were used to determine the response frequency of the patterns. The vertical acceleration f(t) of Eq. (1) is periodic with period $2\pi/\omega$. The patterns L2, H, and Q all appear stationary when the strobe period is $2\pi/\omega$, while the pattern L1 appears stationary only at period $4\pi/\omega$ [16].

We speculate that a weakly nonlinear theory of quasipatterns similar to the N = 6 case considered in Ref. [1] may be appropriate to describe our results. The hexagons are weakly subcritical (less than 5% hysteresis) and their overall behavior is similar to that of hexagons in non-Boussinesq convection for which the theory is well established [17]. A sufficient change in the cubic-order coefficients [$\beta(\theta)$ in the notation of [1]], perhaps due to proximity to the wave-number bicriticality, would suffice to destabilize the hexagons in favor of the twelvefold quasipattern. We note that most analyses of patternforming systems consider only regular patterns (lines, squares, hexagons). But because hexagons arise in a variety of such systems, the transition to a twelvefold quasipattern may be more common than previously supposed.

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- [9] The viscosity serves to widen the band of unstable wave numbers and thus reduce quantization effects in a finite container. For kinematic viscosity v assumed small and for a sinusoidal forcing $f(t)=a\cos(2\omega t)$, the width is $\Delta k = (8\sqrt{2}/3)vk^3\omega^{-1}\sqrt{\mu}$ where $\mu \equiv (a-a_c)/a_c$. This should be compared to π/L , where L is the size of the container. The vanishing width as $v \rightarrow 0$ results in severe quantization for fluids such as water, mercury, butanol, or ethanol.
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- [11] T. B. Benjamin and C. Scott, J. Fluid Mech. 92, 241 (1979). The container is filled to the brim, which is a precisely machined corner. The surface becomes pinned at the corner and the volume of fluid is adjusted so that the surface is flat everywhere and thus has no meniscus.
- [12] We have found that mixtures of less than about 85% glycerol, with viscosity less than about 0.70 cm²/s, also yield squares when forced with a single frequency.
- [13] Low-amplitude waves of circular form coming from the boundaries are observed but are ignored in the determina-

tion of the pattern plan forms reported in the figures. The surface is considered "flat" when these waves occur. No recognizable circular waves are observed simultaneously with any other pattern.

- [14] A symmetry argument used by Milner [5] and others explains why hexagons do not usually arise in Faraday experiments. Since the critical modes respond subharmonically, the standing-wave amplitude equations must be sign invariant and thus quadratic terms do not appear. This argument does not necessarily apply to the case of two-frequency forcing.
- [15] Fourier transforms of the images show peaks with twelvefold symmetry, but no useful information about spatial harmonics of the surface deformation survives the very strongly nonlinear experimental visualization. We find no evidence for tenfold or eightfold orientational order.
- [16] In the single-frequency cases 4ω and 5ω , the lines are found to have the usual subharmonic responses 2ω and 2.5ω , respectively.
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FIG. 3. Photographs of the twelvefold quasipattern. (a) Circular container, 12 cm in diameter and 0.29 cm deep. The fluid is 88% glycerol, $v=0.85\pm0.5$ cm²/s at 23 ± 0.1 °C; forcing parameters for Eq. (1) are a=142 m/s², $\theta=66.1^{\circ}$, $\omega/2\pi=14.6$ Hz, $\phi=75^{\circ}$. (b) Irregularly shaped container, 0.3 cm deep, with a flat bottom and with side walls in the shape of France; the distance Bordeaux-Geneva is 5 cm. The fluid is 82% glycerol, $v\approx0.60$ cm²/s, at ambient temperature; forcing parameters are $\theta=65^{\circ}$, $\omega/2\pi=31$ Hz, $\phi=66^{\circ}$; a was adjusted to be slightly above a_c (not measured).



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