Pattern formation at the bi-critical point of the Faraday instability

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We present measurements on parametrically driven surface waves (Faraday waves) performed in the vicinity of a bi-critical point in parameter space, where modes with harmonic and subharmonic time dependence interact. The primary patterns are squares in the subharmonic and hexagons in the harmonic regime. If the primary instability is harmonic we observe a hysteretic secondary transition from hexagons to squares without a perceptible variation of the fundamental wavelength. The transition is understood in terms of a set of coupled Landau equations and related to other canonical examples of phase transitions in nonlinear dissipative systems. Moreover, the subharmonic-harmonic mode competition gives rise to a variety of new superlattice states. These structures are interpreted as mediator modes involved in the transition between patterns of fourfold and sixfold rotational symmetry.

1. Introduction

The Faraday experiment has nowadays become a model system for pattern formation in hydrodynamic systems (Faraday (1831)). For a review see Miles (1990) and Müller, Friedrich & Papathanassiou (1998). Standing waves are generated on the liquid air interface in response to a time periodic gravity modulation. Under typical laboratory conditions and assuming that the excitation acceleration is sinusoidal with $g(t) = g_0 + a \sin \Omega t$ these surface waves oscillate with twice the period of the external drive (Benjamin & Ursell (1954)). This is a consequence of the parametric drive mechanism and denoted here as the subharmonic Faraday resonance. Surface waves synchronous (harmonic) with the drive can be generated, too. They have been observed first by adding a second frequency component to the excitation signal (Edwards & Fauve (1994)). Later on, following a suggestion of Kumar (1996), harmonic Faraday waves have also been excited with the usual single frequency drive (Müller et al. (1997)). This, however, requires rather extreme (parameter) conditions, namely thin fluid layers in combination with drive frequencies lower than some threshold $f_b$. Increasing $f = \Omega/(2\pi)$ beyond $f_b$ lets the Faraday waves resonate with their usual subharmonic time dependence (Wagner, Müller & Knorr (2000)). For operating frequencies $f$ close to the bi-critical value $f_b$ the harmonic and subharmonic modes compete. Owing to the dispersion of surface waves different frequencies imply different wavelengths. As a consequence non-linear pattern formation is

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Figure 1. Sketch of the experimental set-up. See text for further explanation.

affected in a significant manner: Subharmonic modes \((f < f_b)\) form square patterns, harmonic modes \((f > f_b)\) hexagons. The occurrence of a primary hexagonal surface tiling of the subharmonic regime is generically due to a three wave interaction. At elevated drive amplitude we observe a transition towards a square pattern. This is similar to the canonical hexagon-line transition in Rayleigh-Bénard convection, which can be observed if “non-Boussinesq” effects become significant (Walden & Ahlers (1981) and Ciliberto, Pampaloni & Pérez-Garzia (1988)). A transition from hexagons to squares has been found only recently in the Bénard Marangoni instability (Nitschke & Thess (1995), Bestehorn (1996) and Eckert, Bestehorn & Thess (1998)).

The measurements presented here give a comprehensive account of our investigations on Faraday pattern selection in the vicinity of the bi-critical point. Thereby the interaction between harmonic and subharmonic modes of different wavelengths gives rise to new resonant phenomena: superlattices with either fourfold or sixfold rotational invariance. Though superlattices are very common in solid state and surface physics, they have been found on macroscopic scales only recently (Pampaloni et al. (1997), Kidrolli, Pier & Gollub (1998), Arbell & Fineberg (1998), Wagner, Müller & Knorr (1999) or Wagner, Müller & Knorr (2000)).

Within a cascade of secondary phase transitions superlattices are found to mediate between the two incompatible symmetry classes, of squares and hexagons. For instance a primary subharmonic pattern with quadratic surface tiling experiences a crossover to a hexagonal superlattice via two quadratic superlattices (Wagner, Müller & Knorr (2000)) with a prominent displacive character in one or two lateral directions. After passing a phase with a hexagonal superlattice the transition process reaches a pure hexagonal pattern characterized by a single wavelength and oscillating in synchronous response to the external drive.

For several of the observed transitions we are able to provide explanations in terms of resonant amplitude equations for the governing spatial modes. The structure of these equations is simply based on symmetry and resonance arguments. In spite of their simplicity these equations provide an understanding of many remarkable features of the superlattices, in particular their displacive character. This phenomenological approach is certainly facilitated by the small number of experimental control parameters. This is unlike earlier experiments (Kidrolli, Pier & Gollub (1998) and Arbell & Fineberg (1998) or Wagner, Müller & Knorr (1999)), which use a more complicated multiple frequency drive or a viscoelastic fluid to drive the system into the bi-critical situation. Thereby different kinds of superlattices have been reported as well. But clearly, a larger number of control parameters renders a theoretical understanding more unwieldy and less intuitive. For the theoretical approach to superlattices see e.g. Dionne & Silber (1997) and Silber & Skeldon (1999).

2. Experimental set-up

2.1. Vibration system and sample fluid

Figure 2 shows a schematic diagram of the experimental setup. Its heart is a large displacement shaker unit (V617 Gearing & Watson Electronics Ltd.) connected to a 4kW power amplifier. The shaker supplies a maximum force of 4670N and a peak-to-peak elevation of \(s_{\text{max}} = 54\text{mm}\). Such a large displacement is necessary to obtain a sufficient acceleration \(a\) at lower driving frequencies. The drive signal for the power amplifier is
synthesized by means of a digital-analog card installed in a Pentium PC. The actual acceleration of the container is measured with a piezo-electric device, the amplified signal of which is routed to the PC for data acquisition. Since the characteristics of the shaker turned out to be rather non-linear at operation frequencies below $f = \Omega/(2\pi) < 10\text{Hz}$ a continuous control of the excitation signal was necessary. To guarantee a sinusoidal container acceleration $a \sin \Omega t$ the recorded accelerometer signal was decomposed into Fourier components. The parasitic higher harmonics of $\Omega$ were eliminated by admixing Fourier contributions with appropriate inverse phases to the excitation signal. Their amplitudes were determined by a proportional control loop. That way the power spectrum of the accelerometer signal is made monochromatic with a purity of 99%.

The cylindrical container for the sample liquid was machined out of aluminum and was anodized black. To avoid pollution and temperature drifts within the fluid, the container was sealed with a glass plate. The inner container diameter was $d = 290\text{mm}$, the depth $50\text{mm}$. Over a distance of $12\text{mm}$ from the edges of the container the depth continuously increases from zero to the bottom. This ”soft boundary condition” with an average angle of $30^\circ$ helps to minimize the generation of parasitic meniscus waves. A meniscus under vertical vibration always emits waves with the frequency $f$ of the external drive. Since these waves have non vanishing amplitudes even at subcritical drive amplitudes $a < a_c$ they blur the onset detection. The beach like boundary fulfill their purpose well, at least above $10\text{Hz}$.

The probe fluid was a low viscosity Silicon oil (Dow Corning 200) with the manufacturer specifications of kinematic viscosity $\nu = 5 \times 10^{-6}\text{m}^2/\text{s}$, surface tension $\sigma = 0.0194\text{N/m}$ and density $\rho = 920\text{kg/m}^3$ at our working temperature $T = 25^\circ\text{C}$. A heating foil was mounted on the outside of the container. By means of a temperature controller the temperature measured by a PT-100 resistor (embedded in the container body) was regulated by $\pm 0.1^\circ\text{C}$.

2.2. Visualization technique

To visualize the surface profile we used a full frame CCD camera (Hitachi KPF-1) situated above the fluid surface in the center of a ring consisting of 120 LEDs. The ring had a radius of $R = 0.3\text{m}$ and its distance from the fluid surface was $L = 1.50\text{m}$. The camera was synchronized to the excitation signal with an exposure time of $1/256$ of the drive period. It follows from geometrical optics that only surface elements with a certain steepness reflect light into the camera.

For an evaluation of the spatial symmetry of the surface deformation $\zeta(x, y)$ we relied on a Fourier technique. To that end the recorded light intensity $I(x, y)$ of a video image was convoluted with a Gaussian window function and processed by a FFT algorithm. This yields the two dimensional spatial power spectrum $P(k)$. To determine the wavelength of the pattern $P(k)$ is azimuthally averaged by integrating over circles with constant radius $|k| = k$. The primary peak in the resulting one-dimensional spectrum is fitted by a Gauss function the center of which determines the fundamental wave number. Clearly, the resolution of this procedure is limited by the number of wavelengths in the container. This is especially the case for subharmonic Faraday waves where the uncertainty of $\Delta k/k$ is about 10%.

Due to the nonlinear relationship between the surface elevation $\zeta(x, y)$ and the recorded light intensity $I(x, y)$, the power spectrum entails higher harmonics of the fundamental wave number, even if the surface profile $\zeta(x, y)$ does not. Thus the relation between $I(x, y)$ and $\eta(x, y)$ is generally too complicated to allow a reconstruction of the surface profile. Nevertheless for simple surface patterns (such as squares) we have solved this ”inverse problem” by the following method: Starting from an estimated surface profile composed
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Figure 2. Neutral stability curves \(a(k)\) computed for the parameters of the sample fluid at a drive frequency (a) \(f = 6.25\) Hz < \(f_b\) and (b) \(f = 7.25\) Hz > \(f_b\). In (a) the primary resonance is harmonic, in (b) it is subharmonic. Regions where the flat surface state is unstable are shaded. Horizontal lines denote the thresholds for secondary and higher order transitions (for details see text).

of a small number of spatial Fourier modes, the light distribution of the expected video image was computed by means of a ray tracing algorithm. Then we adapted the mode amplitudes and their relative phases such as to optimize the agreement between the calculated and recorded video picture.

A reconstruction of the full time dependence of an oscillating surface wave pattern was not possible with our equipment. Nevertheless, the electronic shutter of the camera provides an easy and very sensitive technique to discriminate subharmonic frequency components in an otherwise harmonic time signal. This is because a harmonic time dependence \(\zeta(t)\) is invariant under the symmetry operation \(t \rightarrow t + \frac{2\pi}{\Omega}\) implying a frequency spectrum of integer multiples of \(\Omega\) thus \(\zeta(t) = \sum_n \zeta_n e^{in\Omega t}\). In contrast the subharmonic time signal transforms after one drive period as \(\zeta(t) \rightarrow -\zeta(t)\) enforcing a Fourier representation in the form of \(\zeta(t) = \sum_n \zeta_n e^{i[(n+1)/2]2\pi t}\). Thus by triggering the camera shutter with the drive frequency \(\Omega\), video images with a harmonic time dependence appear stationary, while those with subharmonic frequency contributions flicker due to a slight optical asymmetry between heaps and hollow of the deformed surface. Note however, that this trigger technique does not allow to identify harmonic frequency components in an otherwise subharmonic spectrum.

3. The onset of the Faraday instability

It is well known that the stability problem of a free liquid surface under gravity modulation (Faraday instability) can be approximately mapped to that of a parametrically driven pendulum (Rayleigh (1883), Benjamin & Ursell (1954)). The primary resonance of which occurs at twice the period of the drive (subharmonic response). However, as first pointed out by Kumar (1996) the Faraday instability may also appear in synchronous resonance with the external drive, usually denoted as the harmonic response. The conditions under which the harmonic resonance preempts the subharmonic one have been worked out in detail by Cerda & Tirapegui (1997) and Müller et al. (1997) revealing that low filling levels in combination with small drive frequencies are necessary. In the present experiment we choose a fill height of \(h = 0.7\) mm, which is at the operation frequencies of 6 < \(f < 8\) Hz – comparable to the viscous penetration depth \(\xi = \sqrt{2\nu/\Omega} \approx 0.5\) mm.

For the fluid parameters at hand a linear stability analysis of the flat surface state (according to the method of Kumar & Tuckerman (1994), which assumes a laterally infinite system) reveals the location of the bi-critical point at a drive frequency of \(f_b = 6.3\) Hz. Figure 3 shows neutral stability diagrams (drive amplitude \(a\) vs. wave number \(k\)) for both situations \(f < f_b\) and \(f > f_b\). Experimentally the critical acceleration \(a_c\) (absolute minimum of the neutral stability diagram) has been determined by setting up the system at a constant frequency and ramping \(a\) quasi-statically in steps of 0.2% suspended by intervals of 240s. The onset amplitude \(a_c\) was defined when the camera detected the first light reflex (figure 3a). To enhance the detection sensitivity the surface was illuminated by a diffusive light source from the side rather than using the dark-field technique described above. This is because the latter method requires a minimum surface gradient...
Figure 3. (a) Critical amplitude $a_c$ and (b) critical wave number $k_c$ for the onset of the Faraday instability drawn as a function of the drive frequency $f$. The bi-critical point $f = f_b$ is located where the harmonic ($f < f_b$) and subharmonic ($f > f_b$) thresholds intersect. Symbols mark experimental data points, lines the theoretical results for a laterally infinite fluid layer. Circles and dotted lines refer to the harmonic response, squares and solid lines to the subharmonic one.

Figure 4. Phase diagram of the observed patterns obtained by quasi-statically ramping the driving force. The symbols mark the observed transition points between the different patterns, the lines are guides for the eyes. The spatial ordering of the patterns is indicated by Roman numerals, the superscript s and h denote the character of the time dependence being either purely subharmonic or harmonic. With $s+h$ we indicate patterns formed by an interaction of subharmonic and harmonic modes. The thick line separates harmonic from subharmonic and $s+h$ regions. I: flat surface; II: subharmonically oscillating squares (figure 11 a,b); IIIa: $\sqrt{2} \times \sqrt{2}$ superlattice p2mg (figure 11c,d); IIIb: $\sqrt{2} \times \sqrt{2}$ superlattice c2mm (figure 11f); within the subregion above the dotted line the pattern is time dependent and disordered (see figure 14); IV: $\sqrt{3} \times \sqrt{3}$ superlattice, (only for $f < 6.9$ Hz stationary, figure 13); V: harmonically oscillating hexagons (figure 11i); VI: harmonically oscillating squares (figure 5d); VII: $2 \times 2$ superlattice (figure 16); VIII: local instability and droplet ejection.

of $|\nabla \xi(x,y)| = \tan \alpha \approx 0.1$ for the onset detection. We estimate the accuracy of our threshold determination by 0.5%.

Once a standing wave pattern had covered the whole surface the fundamental frequency of the surface oscillation was determined with the help of the electronic shutter of the video camera. That way we located the transition point at a bi-critical frequency $f_b = 6.5 \pm 0.1$ Hz. After these preliminary measurements we switched back to the dark-field illumination to proceed with the spatial pattern analysis. The critical wave numbers $k^h_c$ and respectively $k^s_c$ were determined by Fourier transforming a surface image taken at a driving strength of $\varepsilon = (a - a_c/a_c) \approx 3\%$ (figure 1b). The operating prescription $k_c = k(\varepsilon \approx 3\%)$ for the determination of the critical wave number is motivated by the fact that we were unable to detect any change of the wave number $k$ by varying $\varepsilon$ (see also Wernet et al. (2001)). The experimental results for $a_c$ and $k_c$ as well the bi-critical frequency $f_b$ are (found to be) in good agreement with the theoretical predictions. For the critical acceleration the discrepancy is less than 2%. Here the uncertainty is mainly due to errors in the determination of the small fill height $h$. For larger values of $h$ the agreement improves up to 1%. For the critical wave number the discrepancy between theory and experiment is better than 4% in the harmonic case but it increases up to 10% on the subharmonic regime. This is due to the spatial resolution, which becomes worse at larger wavelength. Owing to the abrupt change of the response frequency at $f = f_b$ the wave number shows a discontinuous jump (see figure 3b). The empiric ratio of the wave numbers at $f = f_b$ is found to be

$$\left| \frac{k^h_c}{k^s_c} \right|_{exp} = 1.58 \pm 0.15,$$

in agreement with the prediction of the linear stability theory $\left| \frac{k^h_c}{k^s_c} \right|_{theo} = 1.59$.

4. Overview over the phase diagram

The phase diagram shown in figure 4 has been obtained at various constant driving frequencies $f = \Omega/(2\pi)$ while ramping the driving amplitude from $\varepsilon = -1\%$ up to 25%
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in steps of 0.2%. After each increment the ramp was suspended for 240 seconds to give the system time to relax. Then a photo or - in the case of time dependent patterns a video film - of the surface state was taken. At the point where a new spatial or temporal mode appeared or an existing one died out, the actual acceleration was defined as the transition boundary to a new ”phase”. At the maximum acceleration amplitude the ramp was reversed to check for an eventual hysteresis.

In the following chapter 5 we describe in detail the type of primary patterns, which appear near onset of the Faraday instability. Secondary and higher order transitions towards more complicated structures are dealt with in sections 6 and 7. Thereby two representative experimental runs will be described in detail, the first was taken at \( f = 6.25 \text{ Hz} < f_b \) and a second at \( f = 7.25 \text{ Hz} > f_b \).

In the former case the primary pattern exhibits a harmonic time dependence, which turns out to be quite robust as it persists over the whole investigated \( \varepsilon \)-ramp. The primary spatial surface wave structure starts with an ideal hexagonal symmetry (region V), which then transforms into a pattern of squares (region VI) as \( \varepsilon \) is raised. This transition is hysteretic, its global aspects can be understood in terms of a simple model of six coupled amplitude equations.

In the second run at \( f > f_b \) the primary surface pattern consists of subharmonically oscillating squares (region II). On increasing the drive strength \( \varepsilon \) the interaction with the neighboring harmonic Faraday instability leads to the appearance of a quadratic \( \sqrt{2} \times \sqrt{2} \) superlattice with a displacive character in one and/or two lateral directions (regions IIIa and IIIb, respectively). After crossing a phase region of non-stationary patterns with a slow time dependence the system enters a hexagonal \( \sqrt{3} \times \sqrt{3} \) superlattice (region IV). Mediated by a second local reconstruction process the final stationary surface pattern is a quadratic \( 2 \times 2 \) superlattice (region VII). Regardless of whether the response is s or h, the surface finally breaks up and droplets are ejected (region VIII, \( \varepsilon \simeq 15 - 25\% \)).

5. Pattern formation close above onset of the Faraday instability

Close to the harmonic onset of the instability \( (f < f_b) \) hexagons are the preferred primary surface pattern (5a, region V) for \( f > f_b \), however, squares are stable (figure 11a). Wagner, Müller & Knorr (2000) (see figure 3 in it) has shown that even for small \( \varepsilon \) the wave profile is rather anharmonic.

5.1. Theoretical model for the primary hexagonal pattern at \( f < f_b \)

The appearance of hexagons at the ”harmonic side” of the bi-critical point follows from a triple wave vector resonance: Since the spectrum of the harmonic Faraday mode \( \xi^h(t) \) consists of integral multiples of the drive frequency it allows for two critical standing wave modes \( k_1 \) and \( k_2 \) to resonate with a third one. The requirement \( |k_1| = |k_2| = |k_3| = k^h_c \) along with the resonance condition \( k_1 + k_2 + k_3 = 0 \) enforce a mutual angle of 120° between the wave vectors implying the hexagonal symmetry. The evolution equations for the respective mode amplitudes \( H_1, H_2, H_3 \) are of the following structure

\[
\partial_t H_1 = \varepsilon H_1 + \beta H_2^* H_3^* - \left[ |H_1|^2 + \Gamma(120°)(|H_2|^2 + |H_3|^2) \right] H_1.
\]

(5.1)

Thereby \( \beta \) is a second order coupling coefficient and \( \Gamma(\theta) \) is the cubic cross coupling coefficient, which depends on the angle between the interacting modes. Moreover the stars denote complex conjugation. The corresponding equations for \( H_1 \) and \( H_2 \) follow by permutation of the indices. The term of cubic order is crucial for saturation. A linear stability analysis of the finite amplitude stationary solution \( |H_1| = |H_2| = |H_3| \) given by Ciliberto, Pampaloni & Pérez-Garzia (1988) yields a backwards bifurcation out of
Figure 5. Photographs of the fluid surface as obtained by ramping the drive amplitude $\varepsilon$ at $f = 6.25\,\text{Hz}$. In the corners of the pictures the container boundary is visible. In (a) and (d) circles mark the nuclei at which the transition process is initiated. (e) is an enlarged sector of (b) with marked penta-lines and hepta-defects.

the trivial solution $H_i = 0$. This reflects a hysteretic transition from the undisturbed flat surface to a pattern of hexagons (compare figure 4). However, we were unable to resolve any hysteresis because of small amplitude (harmonically oscillating) meniscus waves emitted from the rim of the container.

5.2. Theoretical model for the primary square pattern at $f > f_b$

Understanding the pattern selection process at the subharmonic side of the bi-critical point is rather more complicated. Since the frequency spectrum of the subharmonic Faraday response consist of half integer multiples of $f$, any triple of linear unstable modes is prevented from resonating. Thus nonlinear pattern selection is dominated by a four-mode interaction. Unlike triad resonances, which operate exclusively at an interaction angle of $\theta = 120^\circ$, four-wave resonances are less selective as they work at arbitrary angles $\theta$. This fact is also reflected by the corresponding system of amplitude equations. Taking a set of $N$ standing waves with wave numbers $k_i$ at length $|k_i| = k_c^s$ but arbitrary relative orientation, the respective mode amplitudes $S_i$ are governed by the following evolution equations

$$\partial_t S_i = \varepsilon S_i - \sum_{j=1}^{N} \Gamma (\theta_{ij}) |S_j|^2 S_i,$$

with $\theta_{ij}$ being the angle between $k_i$, $k_j$. Usually the participating modes are taken to be equidistant on the circle $|k_i| = k_c^s$, thus $\theta_{i,i+1} = 2\pi/N$. In this case $N$ indicates the type of symmetry of the pattern, namely $N = 1$ lines, $N = 2$ squares, $N = 3$ triangles or hexagons, .... As outlined in Refs. Milner (1991), Müller (1994), Zhang & Vinals (1997) and Chen & Vinals (1997) the question of what is the most preferred symmetry is reduced to minimizing the "free energy"

$$F = -\varepsilon \sum_{i=1}^{N} |S_i| + \frac{1}{2} \sum_{i,j=1}^{N} \Gamma (\theta_{ij}) |S_i|^2 |S_j|^2$$

with respect to $N$ at given $\Gamma(\theta)$. For low viscosity fluid layers of infinite depth the coupling function $\Gamma(\theta)$ was first evaluated by Zhang & Vinals (1997) who found that a pattern of square symmetry is the most preferred one at drive frequencies beyond $f \approx 50\,\text{Hz}$. At lower frequencies patterns with a degree of rotational symmetry up $N = 7$ (quasi-periodic) are likely to occur These predictions were found to be in qualitative agreement with experiments Kudrolli & Gollub (1996), Binks & van de Water (1997). The latter authors also extended the above considerations to the case of finite fill heights and found square patterns to dominate also at lower drive frequencies in agreement with their own experiments and also with ours.

6. Secondary and higher order transitions at $f < f_b$

6.1. Hexagon-square transition

In this section we investigate the crossover from the primary hexagonal structure (region V in figure 4) to the square pattern of region VI of the harmonic regime. Throughout
the whole bifurcation cascade the time dependence is purely harmonic without perceptible subharmonic frequency contributions. The results described below were obtained by ramping the drive amplitude \( \varepsilon = -0.1\% \) up to \( \varepsilon = 16.5\% \) while keeping the frequency \( f = 6.25 \text{ Hz} \) fixed (see figure 4). The transition is connected with a strong hysteresis giving an overlap between the two ideal structures. Since the lateral aspect ratio (container size over wavelength) is not large the hexagonal pattern adapts to an azimuthal symmetry. This affects the defect dynamic of the pattern and thereby also the transition process. Starting the \( \varepsilon \)-ramp at the hexagonal structure, a rearrangement of the pattern sets in, initiated by six nuclei of local quadratic order as indicated by the circles in figure 5a. The size of these patches increases with \( \varepsilon \) (Fig. 5b). Conversely, starting at elevated \( \varepsilon \) with the perfect square pattern and ramping downwards generates four nuclei of local hexagonal order (Fig. 5d). Along the domain boundaries between the patches penta-hepta defects occur. This kind of defect is very common in 2-D hexagonal patterns. Experimental (Ciliberto, Pampaloni & Pérez-Garzia (1988) and Tan, Ohata & Wu (2000)) and theoretical (Prisman & Nepomnyashchy (1993) and Tsimring (1996)) investigations reveal that a penta hepta defect can be formed by a phase defect among two of the participating modes. Figure 5e presents an enlarged subrange of figure 5b depicting ”penta lines”. This is a row of penta defects (unit cells with five neighbors) ending in hepta defects (unit cells with seven neighbors). Strengthening the drive makes the domain walls invade the areas of hexagonal symmetry with new quadratic cells being generated along the penta lines.

To quantify the hysteresis of the hexagonal quadratic reconstruction we applied two different techniques, one in real space and the other in Fourier space. In the former case the number of unit cells with four neighbors \( A_4 \) and those with six neighbors \( A_6 \) was counted. The result of this procedure is shown in figure 6a. It reveals a hysteresis loop extending from \( \varepsilon = 5\% \pm 1\% \) up to \( \varepsilon = 15\% \pm 1\% \). The obvious staircase behavior reflects the discretization of the k-values due to the finite size of the container. For different runs the steps do not occur at the same \( \varepsilon \)-values.

The second method for evaluating the square-vs.-hexagon surface coverage we Fourier transformed the full surface picture and evaluated the power spectrum \( P(k) = P(k, \phi) \) at \(|k| = k_c\) as a function of the azimuthal angle \( \phi \). We then computed the azimuthal auto correlation \( C(\phi) \) as follows

\[
C(\phi) = \frac{\int_0^{2\pi} P(k_c, \phi)P(k_c, \phi + \phi')d\phi'}{\int_0^{2\pi}[P(k_c, \phi')]^2d\phi'}. \tag{6.1}
\]

That way a pattern with quadratic symmetry leads to a peak at \( \phi = 90^\circ \) and \( 180^\circ \), while a hexagonal pattern produces maxima at \( \phi = 60^\circ \), \( 120^\circ \) and \( 180^\circ \). Figure 6 illustrates how \( C(\phi) \) develops with the drive amplitude, both at increasing and decreasing \( \varepsilon \). In order to compare with the cell counting method (see figure 6a) the value \( C(\phi = 90^\circ) \) is...
Figure 8. Set of wave vectors necessary to build up patterns of rolls, hexagons or squares.

Figure 9. Sketch of the bifurcation diagram for the transition from a hexagonal to a quadratic pattern. Dotted lines indicate unstable, straight lines stable branches. The hysteresis observed in the experiment may be attributed to the bi-stable region, where squares and hexagons co-exist.

plotted versus $\varepsilon$ in figure 6b. Regarding the width of the hysteresis loop, the two methods agree within a few percent with each other.

An important feature of the above phase transition is the constancy of the wave number $k_h(\varepsilon) = k_{h_0}$: Within the experimental resolution of $\Delta k/k = \pm 1\%$ no dependency of the wave number on $\varepsilon$ could be detected throughout the whole investigated drive amplitude range. This is of particular significance as it allows for to describe the global aspects of the phase transition in terms of a simple model as given in the next section.

6.2. Comparison with theory

It was outlined in §5 that the hexagonal symmetry at small $\varepsilon$ is a consequence of a three wave resonance, reflected by the second order term in equation (5.1). However, upon increasing the control parameter $\varepsilon$ the term of cubic order becomes increasingly important thus finally enforcing the transition towards squares. A minimal model that allows stationary solutions in form of squares and hexagons (and also lines) along with a linear stability analysis of these solutions can be found in a recent publication of Regnier et al. (1997). This model is an extension of equation (5.1) as it relies on six independent modes $k_i,i=1,...,6$ rather than only three (see figure 8). Since $k_{i,i=1,2,3}$ enclose mutual angles of $120^\circ$ the remaining wave vectors $k_{i,i=4,5,6}$ describe a second pattern of hexagons, which is rotated by an angle of $30^\circ$ relative to the first. Depending on the number of saturated mode amplitudes $H_i$ the model allows stationary solutions in form of lines, squares, hexagons or even more complicated solutions without translational symmetries (quasi-periodic structures). We shall disregard these complications here and focus on the square-hexagon competition. Regnier et al. (1997) demonstrate that the hexagonal state (given by finite $|H_1| = |H_2| = |H_3|$) becomes unstable against shear distortions $\delta$ in form of rhombuses $(\delta H_2 \neq 0 \neq \delta H_3)$ at a drive amplitude $\varepsilon > \varepsilon_H$. By way of contrast, for a critical value of $\varepsilon < \varepsilon_S$ squares (given by finite $|H_1| = |H_4|$) become unstable against rhomboedric $(H_2, H_3)$ disturbances. These results are summarized in the bifurcation diagram of figure 9.

It is tempting to attribute the bi-stable square-hexagon region between $\varepsilon_S < \varepsilon < \varepsilon_H$ (see figure 9) to the hysteretic region depicted in figure 5b,c). Note however, that the observed transition runs through a reconstruction via penta-hepta defects, the complicated space dependence of which goes beyond the scope of the present model.

We mention that a discussion of a transition between hexagons and lines in terms of the above three mode model (5.1) has been given earlier in the context of Rayleigh-Benard convection by Walden & Ahlers (1981) and Ciliberto, Pampaloni & Pérez-Garzia (1988). Thereby the phase transition results from non-Boussinesq effects induced by a strong applied temperature gradient. Recently, several authors report Bénard-Marangoni experiments, which show a transition from hexagons to squares (Nitschke & Thess 1995, Bestehorn 1996 and Eckert, Bestehorn & Thess (1998)). A similar transition is found by Abou, Wesfreid & Roux (2000) on the surface inactivity of magnetic liquids (Rosensweig-Instability). However, unlike our measurements, all of the afore mentioned experiments with a hexagon to square transition exhibit a considerable nonlinear wave
Figure 10. Phase diagram of an amplitude scan at $f = 7.25$ Hz. Time dependent transient means an uncorrelated pattern (figure 14), quasistationary indicates a pattern of an almost perfect $\sqrt{3} \times \sqrt{3}$ superlattice (compare figure 15) with a slow defect dynamic.

Figure 11. A $10 \times 10$ cm sector of the photographs of the fluid surface and corresponding Fourier spectra at $f = 7.25$ Hz. a,b): $\varepsilon = 3\%$, region II, pattern of squares; c,d): $\varepsilon = 6.6\%$, region IIIa, $\sqrt{2} \times \sqrt{2}$ superlattice p2mg with a displacive character in $x$-direction; e,f): $\varepsilon = 14\%$, $\sqrt{2} \times \sqrt{2}$, region IIIb, superlattice c2mm with a displacive character in $x$- and $y$-direction. The dotted lines and arrows on the left pictures indicate the directions in which the rows and columns of elevation maxima are displaced.

Figure 12. Particle model of the a) $\sqrt{2} \times \sqrt{2}$ p2mg, b) $\sqrt{2} \times \sqrt{2}$ c2mm superlattice. Also marked are the mirror (m) and glide (g) planes.

number variation $k(\varepsilon)$ up to 10%, which rules out a description in terms of space independent amplitude equations of the type given above.

7. Secondary and higher transitions at $f > f_b$

In the same way as in the preceding paragraph we now turn to the bifurcation scenario at the opposite side of the bi-critical point at $f > f_b$. An amplitude ramp taken at $f = 7.25$ Hz serves as a representative example. §7.4 is an exception, where the focus is on the frequency regime $6.6 < f < 6.9$ Hz.

An overview over the transition behaviour at $f = 7.25$ Hz is given by figures 10 and 11. Starting from subharmonic oscillating squares (region II, figure 11a) a transition to a quadratic $\sqrt{2} \times \sqrt{2}$ superlattice with a displacement of neighbouring elevation maxima in the lateral $x$-direction (region IIIa, figure 11b) takes place. The next pattern is again a $\sqrt{2} \times \sqrt{2}$ superlattice of the original square lattice but this time it exhibits a displacement in both $x$ and $y$ direction. (region IIIb, figure 11c). After a time dependent transient (see figure 14 for a snapshot) this pattern transforms into a "quasi-stationary" hexagonal $\sqrt{3} \times \sqrt{3}$ superlattice (region IV, figure 15). With "quasi-stationary" we indicate that the pattern is slightly disturbed by defects, which induce a slow time dependence on the scale of minutes. Further raising the drive amplitude $\varepsilon$ at $f = 7.25$ Hz makes the quadratic symmetry reappear in form of a $2 \times 2$ superlattice (region VII, figure 16). Performing the same amplitude ramp at a lower frequency of $6.6 < f < 6.9$ Hz the $\sqrt{3} \times \sqrt{3}$ superlattice directly reduces to its underlying pure hexagonal tiling. This last transition will be discussed in §7.4.

7.1. The quadratic $\sqrt{2} \times \sqrt{2}$ superlattice

The bifurcation sequence starts at the primary ideal pattern of subharmonically oscillating squares (region II and figure 11a,b.) composed of the two fundamental wave vectors $k_{S1}$ and $k_{S2}$. Increasing the drive strength $\varepsilon$ beyond 5% displaces every other column of elevation maxima in the direction $\pm(k_{S1} + k_{S2})$. This is shown by the arrows in figure 11a. The resulting pattern (phase region IIIa) is depicted in 11b. The displacement is accompanied by the simultaneous appearance of the modes $k_{H1}$, $k_{H2}$, and $k_{D1}$ in the power spectrum (see figure 14). Due to the approximate equality $|k_{H1}| = |k_{H2}| \simeq k^h_c$ these modes are only slightly damped. In contrast, $|k_{D1}|$ is significantly smaller than $k^e_c$ and $k^h_c$. Therefore the mode $D_1$ is strongly damped. Its mission is to act as a mediator
By means of the ray tracing technique outlined in the previous section we can now simulate the video image associated with \( \eta \) and adapt it to the empiric result. Taking \( \phi_{S_1} = \phi_{S_2} = 0 \) (by a proper choice of the origin) our investigation reveals that the displacement visible in figure 11b can only be reproduced if the spatial phases \( \phi_{D_1}, \phi_{H_1}, \) and \( \phi_{H_2} \) adopt values close to \( \pi/2 \). The associated surface pattern exhibits a 180° rotational symmetry.

By increasing the drive further the \( \sqrt{2} \times \sqrt{2} \ p2mg \) superlattice undergoes a transition which restores the fourfold symmetry. Similar to the above described shift of the columns of elevation maxima, it is now additionally the rows, which experience a displacement in the direction \( \pm (k_{S_1} - k_{S_2}) \) (indicated by the arrows in figure 13). In figure 13 we measured the amount of symmetry restoration by comparing the spectral power associated with \( D_1 \) and \( D_2 \). Beyond \( \varepsilon \approx 9\% \) the transition is complete. The resulting so-called \( \sqrt{2} \times \sqrt{2} \ c2mm \) superlattice (see figure 12b) is depicted in figure 13, and associated with the phase space region IIIb. In Fourier space this transition is carried by the additional modes \( k_{H_2}, k_{H_2}, \) and \( k_{D_2} \) as shown in figure 11. Extending the surface representation 7.1 by these additional components and using it to re-perform the ray tracing image analysis yields the phase information \( \phi_{D_{1,2}}, i = 1, 2 = \phi_{H_{1,2}, i = 1, \ldots, 4} = \pi/2. \)

We mention that the \( \sqrt{2} \times \sqrt{2} \ c2mm \) superlattice state was not observed in an earlier measurement on a liquid of higher viscosity (Wagner, Müller & Knorr 2000). Thereby this structure is preempted by a transition to a hexagonal symmetry.

### 7.2. Theoretical model for the displacive phase

The experimental investigations outlined in the previous section reveal that the prominent displacive character of the \( \sqrt{2} \times \sqrt{2} \) superlattices is associated with the phase information carried by the spatial Fourier modes. In what follows, a minimal model is constructed, which is able to explain the experimentally measured phases. It is important to point out that the structure of these equations just relies on symmetry and resonance arguments (triad wave vector resonances). The numerical values of the appearing coefficients are not known; their evaluation would require a rather complicated nonlinear analysis. Furthermore, for the sake of simplicity, we limit our discussion to the \( \sqrt{2} \times \sqrt{2} \ p2mg \) superlattice. The generalization to the more symmetric \( \sqrt{2} \times \sqrt{2} \ c2mm \) pattern is straightforward.

Assuming that the amplitudes of the primary square pattern have settled at some finite value \( S_1 = S_2 \neq 0 \) the leading order behavior of the remaining modes is governed by the following set of equations

\[
\partial_t D_1 = \varepsilon_D D_1 + \mu_D (S_1^* H_1 + S_2^* H_2) + \chi_S S_1 S_2 D_1^\\star
\]
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Figure 13. Integrated intensity of the $D_1$-peak (circles) and the $D_2$-peak (squares) in the power spectrum as a function of the drive amplitude $\varepsilon$. The error bars mark the standard derivation resulting from 5 succeeding runs.

Figure 14. Snapshot of the fluid surface as obtained at $f = 7.25$ Hz and $\varepsilon = 17\%$. The pattern is strongly time dependent and spatially uncorrelated.

Figure 15. A $8 \times 8$ cm sector of the photographs of the fluid surface and corresponding Fourier spectra at $f = 6.65$ Hz and $\varepsilon = 5\%$. This photograph has been obtained by using a diffusive light source mounted off axis instead of the standard visualization technique. That way the character of the hexagonal $\sqrt{3} \times \sqrt{3}$ superlattice (region IV) appears to be more pronounced (compare figure 4e in (Wagner (2000))).

\[ \begin{align*}
\partial_t H_1 &= \varepsilon_H H_1 + \mu_H S_1 D_1 \\
\partial_t H_2 &= \varepsilon_H H_2 + \mu_H S_2 D_1.
\end{align*} \tag{7.2} \]

Thereby $\varepsilon_H < 0$ and $\varepsilon_D \ll 0$ are the coefficients of linear damping of respectively the $H$ and $D$ modes, while $\mu_{D,H}$ and $\chi_S$ are nonlinear coupling coefficients associated with the triad wave vector resonance.

Although the model equations (7.3) are linear in $D$ and $H$ and thus saturation is not implied, the appearing nonlinearities are phase selective. By writing the complex amplitudes in the form $A_i = |A_i| \exp (\phi_{A_i})$ with again $\phi_{S_1} = \phi_{S_2}$ taken to be zero (choice of space origin) the imaginary part of equation (7.3) yields the phase dynamics

\[ \begin{align*}
\partial_t \phi_{D_1} &= \mu_D |S||H|/|D|[\sin (\phi_{H_1} - \phi_{D_1}) + \sin (\phi_{H_2} - \phi_{D_1})] + \chi_S |S|^2 \sin (-2\phi_{D_1}) \\
\partial_t \phi_{H_1} &= \mu_H |D||S|/|H|[\sin (\phi_{D_1} - \phi_{H_1})] \\
\partial_t \phi_{H_2} &= \mu_H |D||S|/|H|[\sin (\phi_{D_1} - \phi_{H_2})].
\end{align*} \tag{7.3} \]

The fix points of these equations are

\[ \phi_{H_i} = \phi_{D_i} = m\pi/2 \tag{7.4} \]

with $m$ being an integer. By inspection one finds that the solution with even $m$ leads to an square pattern with amplitude modulation while odd $m$ gives rise to the observed displacement (phase modulation). Which one is stable, depends on the numeric values of the coefficients. From the experimental data we conclude that $\phi_{H_i} = \phi_{D_i} = \pi/2$ is the solution applicable to the present experiment.

7.3. The hexagonal $\sqrt{3} \times \sqrt{3}$ superlattice

Upon further increase of the driving force at $f = 7.25$ Hz the quadratic $\sqrt{2} \times \sqrt{2}$ c2mm superlattice transforms into a "quasi-stationary" hexagonal superlattice (region IV, compare figure 13) after passing a region of transient time dependent (figure 14) with squares, hexagons and disordered patterns appearing at the same value of $\varepsilon$. The term "quasi-stationary" is to indicate that the pattern is affected by defects on a slow time scale of minutes. The power spectrum shown in Figure 15b reveals that the structure is composed of a set of three harmonic modes $H_j$ and three subharmonic modes $S_j$. The angles between the wave vectors of the three $H$ modes is $120^\circ$ and so is the angle of the $S$-modes. The $H$-pattern is rotated by $30^\circ$ and locked in phase with respect to the $S$-pattern, the ratio $k_h/k_s$ is $1.73 \approx \sqrt{3}$. Therefore the pattern is denoted as a $\sqrt{3} \times \sqrt{3}$ superlattice. We
mention an earlier investigation on a more viscous fluid of $\nu = 10cS$ (Wagner, Müller & Knorr (2000)): Thereby the transition from the quadratic $\sqrt{2} \times \sqrt{2}$ p2mg to the hexagonal $\sqrt{3} \times \sqrt{3}$ superlattice took place in a more correlated manner via stacking faults while the $\sqrt{2} \times \sqrt{2}$ c2mm state could not be observed.

At higher $\varepsilon$-values the further development of the $\sqrt{3} \times \sqrt{3}$ superlattice depends on the drive frequency: At $6.6Hz < f < 6.9Hz$, where the superlattice is perfectly stationary and almost free of defects, the amplitude of the subharmonic components of the structure continuously becomes smaller until the hexagonal base pattern with a pure synchronous time dependence remains (region V, compare figure 13(a)). This transition is slightly hysteretic ($\Delta \varepsilon \simeq 1 - 2\%$) and can be localized accurately by means of the triggering technique described in §2. We come back to this crossover in §7.4, in order to present a theoretical model.

At drive frequencies larger than $\approx 6.9Hz$ (for concreteness let us return to our run at $f = 7.25Hz$) the hexagonal $\sqrt{3} \times \sqrt{3}$ superstructure transforms into a $2 \times 2$ superlattice (region IV → VII) thereby restoring the quadratic symmetry. The crossover takes place via a spatially weakly correlated transient, being subjected to a strong temporal dynamic. The corresponding pattern looks similar to the surface state shown in figure 14. In its final state the subharmonic and the harmonic wave vectors $k_{H,S}$ are aligned with each other (figure 16b). With $k^H \approx k^c$ but $k^S \approx 0.8k^c$ the resulting length ratio $k^H/k^S$ is about 2.

7.4. Theoretical model for the $\sqrt{3} \times \sqrt{3}$ superlattice-to-hexagon transition

The following section provides a theoretical model for the transition from the $\sqrt{3} \times \sqrt{3}$ superlattice (region IV) to the pure hexagon state in region V, taking place at $6.6Hz < f < 6.9Hz$. Following the lines of §7.2 spatial and temporal resonance arguments are used to construct the set of amplitude equations. We consider the underlying hexagonal base pattern, carried by the modes $H_i$ (c.f. equation (5.1)) as being saturated at some finite amplitude. Then the bifurcation of the subharmonic modes $S_1$, $S_2$, $S_3$ is governed by the following system of equations

$$\partial_t S_1 = \varepsilon S_1 + \beta_{HS} (H_1S_3 + H_2S_2) - \mu \sum_{j=1}^{3} |H_j|^2 S_1 + h.o.t.. \quad (7.5)$$

where the equations for $S_2$ and $S_3$ follow by cyclic permutation, and $\beta_{HS}$ and $\mu$ are nonlinear coupling coefficients. In this model the resonance condition between the $S$ and the $H$ modes enforces a wave number ratio $k^H/k^S|_{theo} = \sqrt{3} \approx 1.73$. Our measurements reveal $|k^c| = k^c| (\varepsilon = 4\%)$ and $|k^S| = 0.91k^c|$, giving $k^H/k^S|_{exp} \approx 1.73$. This justifies our assumption that harmonic modes dominate the pattern forming process, while the (slaved) subharmonic ones have to adapt their wavelength.

The occurrence of the $\sqrt{3} \times \sqrt{3}$ superlattice is caught by a linear stability analysis of equation (7.3). To that end it is convenient to take the amplitude $H = |H_1| = |H_2| = |H_3|$ as the control parameter. Then one obtains a positive growth rate (instability) for the $S_i$ if

$$\varepsilon + 2\beta_{HS}|H| - \mu|H|^2 > 0. \quad (7.6)$$

This condition determines the transition to the superlattice. Recall that (7.3) does not entail terms which are nonlinear in $S_i$ thus the model neither explains saturation of the $S_i$ nor the experimentally observed hysteresis in the transition.

Let us now focus on the spatial phase dynamic of the hexagonal superlattice. For the underlying hexagonal structure with amplitudes $H_i = |H_i| \exp (i\Phi_H)$ we have either $\Phi_H = \sum_j \phi_{H_j} = 0$ or $\pi$, corresponding respectively to "up" or "down" hexagons.
For Faraday waves this distinction is not significant because the surface oscillation periodically switches between these two possible states. Splitting the subharmonic mode amplitudes also into modulus and phase and assuming without loss of generality that \( \phi_H_1 = \phi_H_2 = 0 \), the imaginary part of equation (7.3) yields the identity \( \phi_S_1 = \phi_S_2 = \phi_S_3 = \phi_S_4 \).

In order to determine that value it is necessary to proceed with the amplitude expansion up to the quintic order. Barring all terms which are not phase selective the extension of equation (7.5) reads

\[
\partial_t S_1 = \varepsilon S_1 + \beta_H (H_1 S_3 + H_2 S_2) + \ldots + \chi S_1^* (S_2^*)^2 (S_3^*)^2 + \ldots,
\]

(7.7)

with \( \chi \) being a nonlinear coefficient. Introducing the parametrization \( \Phi_S = \sum_j \phi_S_j = 3\phi_S_1 \) equation (7.7) yields the stationary solution

\[
\Phi_S = n\pi/2.
\]

(7.8)

where \( n \) is an an integer. Following the usual nomenclature the pattern with odd \( n \) is a triangular superlattice, as its rotational symmetry is threefold. For even \( n \) the superlattice exhibits a sixfold symmetry (c.f. Müller (1993)). Since a sixfold symmetry center can be easily identified in figure 15, we conclude that \( n \) is even in our experiment. We point out that an example of a \( \sqrt{3} \times \sqrt{3} \) with third order rotational symmetry (\( n \) is odd) has recently been observed by Pi et al. (2000) (see also Wagner & Müller (2001)).

8. Conclusions

We have presented a comprehensive investigation on Faraday wave pattern formation in the vicinity of a bi-critical situation. Thereby modes with harmonic and subharmonic time dependence interact and lead to a variety of superlattice states. This kind of structure acts as a mediator during the transition process between two incompatible space groups (squares and hexagons). We developed several sets of amplitude equations to model the observed phase transitions. Special attention is devoted to the phase information carried by the participating modes, which is responsible for the remarkable displacive feature of one of the superlattices.

Superlattices are rather common in 2-D solid state physics, and a comparison is therefore instructive. The transition from a simple hexagonal lattice to a \( \sqrt{3} \times \sqrt{3} \) superstructure has been observed for instance in monolayers of \( C_2ClF_5 \) adsorbed on graphite (Fassbender et al. (1995)). The transition from the subharmonic quadratic base pattern to a \( \sqrt{2} \times \sqrt{2} \) superlattices with a displacive character in one direction (\( p2mg \), region IIIa) is analogous to the reconstruction of the clean (100) surface of W (=tungsten) crystals (Schmidt et al. (1992)). Here the surface atoms are displaced in exactly the same way as the elevation maxima of the surface profile in the present study. Most interestingly, the surface of W crystals obeys a transition to a \( \sqrt{2} \times \sqrt{2} \) superlattices with a displacive character in two directions (\( c2mm \), identical to our pattern in region IIIb) in the presence of Hydrogen atoms.

We were also able to provide for the first time a pattern forming system, which undergoes a hexagon-to-square transition without a simultaneous change of the fundamental wavelength. This is in contrast to earlier observations on the Bénard Marangoni system, which showed considerable wave number variations during the transition process. The accompanying description in terms of well defined spatial Fourier modes, makes the phase
transition very fundamental and identifies the underlying pattern selection mechanism to switch from a triad to a four-wave vector resonance.

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