Quasi-Patterns in a Model of Multi-Resonantly Forced Chemical Oscillations

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Multi-frequency forcing of systems undergoing a Hopf bifurcation to spatially homogeneous oscillations is investigated using a complex Ginzburg-Landau equation that systematically captures weak forcing functions that simultaneously hit the 1:1-, the 1:2-, and the 1:3-resonance. Weakly nonlinear analysis shows that generically the forcing function can be tuned such that resonant triad interactions with weakly damped modes stabilize subharmonic quasipatterns with 4-fold and 5-fold rotational symmetry. In simulations starting from random initial conditions domains of these quasi-patterns compete and yield complex, slowly ordering patterns.

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In recent years complex, but ordered spatio-temporal patterns have been found experimentally. In particular on the surface of vertically vibrated fluid layers (Faraday system) various kinds of fascinating periodic superlattice patterns and quasi-patterns have been found experimentally [1]. Subsequently, such patterns have also been observed in optical systems [2], in vertically vibrated fluid convection [3], and on the surface of ferrofluids driven by time-periodic magnetic fields [4]. Here we show that superlattices and quasipatterns should be accessible quite generally in a different class of systems: resonantly forced systems undergoing a Hopf bifurcation to spatially homogeneous oscillations. Paradigmatic for such systems are chemical oscillations [5]. In chemical systems patterns with multiple length scales have so far been obtained only by imposing an external length through spatially periodic illumination [6].

The stability of multi-mode patterns depends on the interaction between their constitutive Fourier modes. For small angles \( \theta \) between the modes the cross-coupling coefficient \( b(\theta) \) is twice as large as the self-coupling coefficient \( b_0 \). Nearly parallel modes therefore suppress each other and unless the cross-coupling coefficient decreases substantially with increasing \( \theta \) only stripe-like patterns are stable. Strong angle dependence can arise if the basic modes couple to weakly damped, resonating modes [7, 8]. Complex patterns with different symmetries can then become stable [9].

The resonances stabilizing complex patterns have been studied in great detail in the Faraday system. Their spatio-temporal nature [10] allows a very controled tuning through the frequency content of the driving [11]. Broadly speaking, there are two mechanisms by which complex patterns can be stabilized: either by enhancing the self-damping \( b_0 \) [12, 13] or by reducing the cross-coupling coefficient \( b(\theta) \) [11, 13].

In this paper we exploit spatio-temporal resonances to induce complex spatial patterns in two-dimensional systems undergoing a Hopf bifurcation to spatially homogeneous oscillations. To this end we apply spatially homogeneous, resonant multi-frequency forcing. By including in the forcing spectrum a frequency component close to twice the Hopf frequency (1:2-resonance) we excite standing waves with a wavenumber determined by the detuning between the forcing and the Hopf frequency [14]. A second frequency near three times the Hopf frequency (1:3-resonance) induces a quadratic interaction term, which otherwise is not allowed in the normal form for Hopf bifurcations. To avoid transcritical bifurcations off the basic state to hexagonal patterns we further include a second forcing frequency close to the 1:2-resonance with a slightly different detuning. Within the weakly nonlinear regime we show that quite independent of the two parameters characterizing the unforced Hopf bifurcation the forcing function can be tuned such that instead of the usual stripe, spiral, and labyrinthine patterns [13] one obtains superlattices and quasipatterns.

The systematic, weakly nonlinear description of a weakly forced super-critical Hopf bifurcation is given by the complex Ginzburg-Landau equation for the complex oscillation amplitude \( A \), which is extended to include near-resonant components of the forcing function [16],

\[
\frac{\partial A}{\partial t} = (1 + i\beta)\nabla^2 A + (\mu + i\sigma - (1 + i\alpha)|A|^2) A + \gamma (\cos \chi + \sin \chi e^{i\nu t}) A^* + \rho e^{i\Phi} A^2. \tag{1}
\]

Here \( \chi \) measures the relative contributions from the two forcing components that are close to the 1:2-resonance, which differ in their frequencies by \( \nu \). The detuning between the Hopf frequency and half the frequency corresponding to the forcing \( \gamma \cos \chi \) is given by \( \sigma \). The strength and phase of the 1:3-forcing is given by \( \rho \) and \( \Phi \), respectively. Non-
linear interactions of the 1:2-resonant forcing and the 1:3-resonant forcing introduce an additional forcing near the Hopf frequency itself. To cancel the resulting additional, inhomogeneous term in \( \Omega \) we assume a further explicit forcing component near the 1:1-resonance. It is straightforward to derive \( \Omega \) from Oregonator-type models for the photosensitive Belousov-Zhabotinsky reaction [17].

The slight detuning between the two 1:2-forcing components introduces the explicit, periodic time dependence of the coefficients in \( \Omega \). Using Floquet theory we determine the instability of the basic state \( A = 0 \) with respect to time-periodic solutions that are phase-locked to the forcing [19]. Due to the dispersion \( \beta \) the detuning \( \sigma \) induces phase-locked modes with a non-zero wave number [14]. Depending on the forcing parameter \( \gamma \) and the detuning \( \nu \) the mode that destabilizes the basic state first is either harmonic or subharmonic relative to the period \( 2\pi/\nu \).

A typical set of neutral curves \( \gamma^{(H,SH)}(k) \) for the harmonic and subharmonic mode is shown in Fig. [1]. We focus here on the subharmonic case for which the time-shift symmetry \( t \to t + 2\pi/\nu \) suppresses any transcritical bifurcations off \( A = 0 \) to hexagons. Weakly damped harmonic modes excited at quadratic order modify the competition between subharmonic modes of different orientation [10, 12]. Their effect is strongest if the forcing \( \gamma \) is only slightly below the critical forcing strength \( \gamma_c^{(H)} \) of the harmonic modes (inset of Fig. [1]). For this reason we tune the forcing parameter \( \gamma \) and the detuning \( \nu \) so that \( \gamma_c^{(H)} \) is only slightly above the critical forcing strength \( \gamma_c^{(SH)} \) of the subharmonic modes. In this paper we focus on the enhancement of the self-damping \( b_0 \) of the subharmonic modes and choose the forcing function such that the critical wavenumber \( k_c^{(H)} \) of the harmonic mode is close to twice that of the subharmonic mode, \( K \equiv k_c^{(H)}/k_c^{(SH)} \approx 2 \). To reduce the competition between modes subtending a specific angle \( \theta \) a wave-number ratio \( K < 2 \) would be chosen [17].

To compute the interaction between modes of different orientation within weakly nonlinear analysis we expand the oscillation amplitude as \( \epsilon \ll 1 \)

\[
A = \epsilon (A_1 e^{ik_1 t} + A_2 e^{ik_2 t}) F(t) + h.o.t. \tag{2}
\]

For subharmonic patterns \( F(t) \) has periodicity \( 4\pi/\nu \) and the amplitude equations for \( A_{1,2} \) do not contain any quadratic terms,

\[
\frac{dA_1}{dt} = \lambda A_1 - b_0 A_1 |A_1|^2 - b(\theta) A_1 |A_2|^2, \tag{3}
\]

with a similar equation for \( A_2 \).

Relevant for the pattern selection is the ratio \( b(\theta)/b_0 \). It is strongly affected by spatiotemporally resonant triads, which are induced by the 1:3-forcing \( \rho e^{i\Phi} \). The resulting \( \rho \)-dependence of \( b(\theta)/b_0 \) is shown in Fig. [2]. The stability conditions for rectangular patterns (corresponding to a rhombic arrangement of the wave vectors) are \( b_0 > 0 \) and \( |b(\theta)/b_0| < 1 \). Thus, with increasing 1:3-forcing \( \rho \) a large range of angles arises for which rectangular patterns are stable, whereas without that forcing only stripe patterns would be obtained.

Given \( b(\theta) \), the linear stability of various types of periodic super-lattice patterns comprised of three or more modes on a fixed periodic Fourier lattice can be determined systematically [18]. The competition between different planforms involves, however, often a collection of modes that cannot be represented on a single Fourier lattice. We have determined the relative stability of such planforms approximately by keeping all modes on the critical circle that are involved in the pattern competition and find bistability between a number of different complex patterns [17]. This approach ignores possible side-band instabilities [19] and higher-order
resonances \[20\] and does not account for possible convergence problems due to small divisors \[21\].

To address the competition between different, simultaneously stable planforms we exploit the variational character of \(\mathcal{A}_j\), \(\frac{\partial \mathcal{A}_j}{\partial t} = -\frac{\partial F_N}{\partial A_j}\) for \(j = 1, \ldots, N\). Fig. \[3\] shows the energies \(F_N\) of patterns comprised of \(N\) modes that are equally spaced on the critical circle as a function of the 1:3-forcing strength \(\rho\). When starting from random initial conditions planforms with lower energy are expected to invade those with higher energy. Thus, for \(\rho \lesssim 0.82\) the final state is expected to consist of stripes, whereas for \(\rho \gtrsim 1.07\) patterns with four or more modes should dominate.

To test the predictions of our weakly nonlinear analysis we have performed direct simulations of the complex Ginzburg-Landau equation \[1\]. For small sizes they confirm the linear stability of periodic patterns comprised of four modes. To investigate the dependence of the pattern selection on the 1:3-forcing strength \(\rho\) we performed simulations in a large square system of linear size \(L \approx 473.39\), which is equivalent to forty wavelengths, for increasing forcing strengths \(\rho\), starting from identical random initial conditions. We have chosen the system size such that the modes making up hexagons and super-squares have equal growth rates to bring out clearly how an increase in \(\rho\) alone tips the balance from hexagons to 4-fold patterns. For \(\rho = 0.9\) a pattern with hexagonal planform rather than a stripe structure arises. Because of the reflection symmetry \(A \rightarrow -A\) induced by the time-shift symmetry \(t \rightarrow t + 2\pi/\nu\) domains with up- and down-hexagons coexist separated by walls containing narrow layers of triangle patterns \[17\].

Increasing \(\rho\) decreases the \(\theta\)-range over which modes suppress each other (cf. Fig \[2\]) and more modes persist, as shown in Fig \[4\] for \(\rho = 1.2\). The pattern exhibits domains of periodic super-lattice patterns with four-fold rotational symmetry ('super-squares' and 'anti-squares' \[18\], marked by dashed-dotted and dashed circles) as well as elements with eight-fold rotational symmetry (solid circle). Increasing the 1:3-forcing to \(\rho = 3\) increases the number of persisting modes further and introduces numerous elements with five- and with ten-fold rotational symmetry (dashed and solid circles) in Fig \[5\].

The patterns shown in Figs \[4,5\] are still evolving, albeit very slowly. Nevertheless, it is clear that for \(\rho \gtrsim 1.1\) they will not evolve to simple hexagon states. While in our simulations for all values of \(\rho\) domains of hexagons appeared for early times, they were replaced for \(\rho \geq 1.1\) by domains of patterns comprised of four or more modes, which have lower energy. A condensed view of the temporal evolution of the patterns for different values of the forcing \(\rho\) is given in Fig \[6\]. It depicts the evolution of the relevant number of Fourier modes \(e^{\xi},\) estimated by the spectral pattern entropy \(S \equiv -\sum_{ij} p_{ij} \ln p_{ij}\). Here \(p_{ij}\) denotes the normalized power spectrum. Clearly the number of significant modes increases with \(\rho\), albeit not monotonically at all times.

In conclusion, we have shown that in systems undergoing a Hopf bifurcation to spatially homogeneous oscillations multi-frequency forcing can sub-
stantially reduce the competition between modes of different orientation leading to complex multimode patterns. By an appropriate choice of the amplitudes and phases of the forcing function, which constitute external control parameters, this regime should be accessible generically, essentially independent of the specifics of the unforced system. Our results should therefore apply to realistic chemical oscillators. From a practical point of view it should be mentioned, however, that the complex patterns possibly arise only very close to onset. This may require systems with relatively large aspect ratios and a very precise tuning of the forcing parameters.

Using direct simulations of the complex Ginzburg-Landau equation we confirmed that these complex patterns arise from general random initial conditions. The appropriate, quantitative characterization of the transients, in which multi-mode structures like super-squares and anti-squares compete with each other, is still an open problem (cf. [22]). Another interesting question is the long-time scaling of the ordering of such complex structures.

Compared to the Faraday system, the forced Hopf bifurcation considered here allows an additional level of complexity by going slightly above the Hopf bifurcation. There complex patterns would compete with spatially homogeneous oscillations. For single-frequency forcing labyrinthine stripe patterns arise from the oscillations through front instabilities and stripe nucleation [22]. It is not known what happens to this scenario when the stripes are unstable to the more complex patterns discussed here.

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[1] B. Christiansen, P. Alstrom, and M. T. Levinsen, Phys. Rev. Lett. 68, 2157 (1992); W. S. Edwards and S. Fauve, Phys. Rev. E 47, R788 (1993); A. Kudrolli, B. Pier, and J. Gollub, Physica D 123, 99 (1998); H. Arbaj and J. Fineberg, Phys. Rev. Lett. 81, 4384 (1998); M.-T. Westra, D. J. Binks, and W. van de Water, J. Fluid Mech. 496, 1 (2003); Y. Ding and P. Umbanhowar, Phys. Rev. E 73, 046305 (2006).
[2] R. Herrero et al., Phys. Rev. Lett. 82, 4627 (1999).
[3] J.L. Rogers, M.F. Schatz, O. Brausch, and W. Pesch, Phys. Rev. Lett. 85, 4281 (2000).
[4] H.-K. Ko, J. Lee, and K. J. Lee, Phys. Rev. E 76, 056222 (2002).
[5] V. Petrov, Q. Ouyang, and H. L. Swinney, Nature 388, 655 (1997); M. Bertram, C. Beta, H. H. Rotermund, and G. Ertl, J. Phys. Chem. B 107, 9610 (2003); M. Orban, K. Kurin-Csorgei, A. M. Zhabotinsky, and I. R. Epstein, Faraday Discuss. 120, 11 (2001).
[6] I. Berenstein et al., Phys. Rev. Lett. 91, 058302 (2003).
[7] N. D. Mermin and S. M. Troian, Phys. Rev. Lett. 54, 1524 (1985).
[8] A. C. Newell and Y. Pomeau, J. Phys. A 26, L429 (1993).
[9] B. Malomed, A. A. Nepomnyashchy, and M. I. Tribelsky, Sov. Phys. JETP 69, 388 (1989).
[10] M. Silber, C. M. Topaz, and A. C. Skeldon, Physica D 143, 205 (2000).
[11] J. Porter, C. M. Topaz, and M. Silber, Phys. Rev. Lett. 93, 034502 (2004).
[12] W. Zhang and J. Vinals, Phys. Rev. E 53, R4283 (1996).
[13] A. M. Rucklidge and M. Silber, Phys. Rev. E (accepted).
[14] P. Coullet, T. Frisch, and G. Sonnino, Phys. Rev. E 49, 2087 (1994).
[15] A. Lin, A.L Hagberg, E. Meron, and H.L. Swinney, Phys. Rev. E 69, 066217 (2004).
[16] P. Coullet and K. Emilsson, Physica D 61, 119 (1992).
[17] J. Conway and H. Riecke (in preparation).
[18] B. Dionne, M. Silber, and A. C. Skeldon, Nonlinearity 10, 321 (1997).
[19] B. Echebarria and H. Riecke, Physica D 158, 45 (2001).
[20] M. Higuera, H. Riecke, and M. Silber, SIAM J. Appl. Dyn. Syst. 3, 463 (2004).
[21] A. M. Rucklidge and W. J. Rucklidge, Physica D 178, 62 (2003).
[22] A. Yochelis et al., SIAM J. Appl. Dyn. Systems 1, 236 (2002).
[23] H. Riecke and S. Madruga, Chaos 16, 013125
(2006).