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Subharmonic phase clusters in the complex Ginzburg-Landau equation with nonlinear global coupling

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A wide variety of subharmonic n-phase cluster patterns was observed in experiments with spatially extended chemical and electrochemical oscillators. These patterns cannot be captured with a phase model. We demonstrate that the introduction of a nonlinear global coupling (NGC) in the complex Ginzburg-Landau equation has subharmonic cluster pattern solutions in wide parameter ranges. The NGC introduces a conservation law for the oscillatory state of the homogeneous mode, which describes the strong oscillations of the mean field in the experiments. We show that the NGC causes a pronounced 2:1 self-resonance on any spatial inhomogeneity, leading to two-phase subharmonic clustering, as well as additional higher resonances. Nonequilibrium Ising-Bloch transitions occur as the coupling strength is varied.

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Spatially extended oscillatory media are an important class of pattern forming systems [1–3]. The patterns are dictated by the mutual interaction between the individual oscillators, which may be purely diffusional. However, in many situations, the oscillatory field experiences, also a global coupling, giving rise to different modes of spatial organization. The dominant dynamics induced by global coupling are phase clusters, where the spatial domain organizes in a small number of synchronized regions with a different phase each. Typically, in an extended medium phase balance adjusts; i.e., the fraction of the system oscillating in each phase is the same. In the simplest case, the phase difference between the domains is $2\pi m/n$ (with m and n being small integers) [4]. These states, which were also termed type-I clusters [5], can be described by phase models, and then, obviously, each phase-balanced n-cluster state has a stationary mean field. In experiments, however, one often observes that the mean field exhibits a pronounced and simple periodic oscillation with the base frequency ν. This will, e.g., be the case if the global coupling term depends nonlinearly on the pattern forming variable or if the natural (uncoupled) oscillation exhibits some degree of anharmonicity. As a consequence, qualitatively different cluster patterns emerge, where the spatial structure oscillates with a subharmonic frequency ν/n and is superimposed by a uniform oscillation with frequency ν. These subharmonic cluster patterns were also coined type-II clusters [5]. The superposition of harmonic and subharmonic temporal modes causes the amplitude of the oscillation to change considerably, rendering thus a theoretical description based on phase equations [6] impossible. Subharmonic clusters were reported in two electrochemical experiments with global coupling, one employing the hydrogen oxidation on a Pt electrode [5] and the other employing the anodic oxidation of a silicon wafer [7]. Careful investigation of literature data suggests that they also formed in the oscillatory CO oxidation on Pt(110) with time delayed global feedback [8] and in the Belousov-Zhabotinsky reaction with global feedback [9]. Subharmonic cluster patterns emerge also when an oscillatory medium is parametrically forced [4], and in this context much insight into their dynamics could be obtained with the forced complex Ginzburg-Landau equation (CGLE). In terms of normal forms, there is, however, a significant difference between a parametric forcing close to a given resonance [4] and a global coupling [10–12]: the former breaks phase invariance and the second does not; hitherto the mechanism leading to the subharmonic cluster patterns in the experimental systems under global coupling without introducing a parametric forcing could not be explained. In this Rapid Communication, we demonstrate that the CGLE augmented by a nonlinear global coupling (NGC) term naturally leads to subharmonic cluster patterns. The nonlinearity in the global coupling term was chosen such that the oscillatory state of the homogeneous mode (HM) is conserved, mimicking therewith the experimentally observed oscillation of a global quantity. A NGC was also introduced in the Kuramoto-Daido phase model where the NGC described a phase shift of a sinusoidal coupling term [13], and it should not be confused with the one considered here.

We assume that we can describe the HM in the most basic way: a simple harmonic oscillator. Quite generally we consider a reaction-diffusion system governed by a modified complex Ginzburg-Landau equation (MCGLE) [7],

$$\partial_t W = W + (1 + ic_1)\partial_x^2 W - (1 + ic_2)|W|^2 W + B, \tag{1}$$

where

$$B = -(1 + i\nu)\langle W\rangle + (1 + ic_2)\langle |W|^2 W\rangle, \tag{2}$$

W is a complex amplitude and $\langle\cdots\rangle$ denotes spatial averages. The terms on the right-hand side of Eq. (2) describe the nonlinear effect of the average amplitude $\langle W\rangle$ on the dynamics and can be thought as an expansion of a negative global coupling to third order. Clearly Eq. (1) is invariant under phase transformation $W \to We^{i\gamma}$. The introduction of the cubic term on the spatial average of the amplitude is motivated by analogous studies on phase ordering under a global conservation law [14] although in [14] $\langle W\rangle$ is constant (the *real* Ginzburg-Landau equation is employed there) and in our case it is a harmonically oscillating function. This can be seen by taking spatial averages on both sides of Eq. (1): we obtain the differential equation $\partial_t\langle W\rangle = -i\nu\langle W\rangle$ which has the closed analytical oscillating solution,

$$\langle W \rangle \equiv W_0 = \eta e^{-i(\nu t+\phi_0)}, \tag{3}$$

which coincides with the HM. η is the (positive real valued) modulus of the average amplitude and ϕ_0 is an initial phase. The HM [Eq. (3)] is always a solution of the MCGLE [Eq. (1)], being *conserved* and unaffected by other active Fourier modes. In general, for periodic boundary conditions, we can write $W(x,t)=\Sigma_{n=-\infty}^{\infty} w_n(t)e^{i2\pi nx/L}$, where L is the length of the system. Although each Fourier mode $w_n(t)e^{i2\pi nx/L}$ is a solution of the MCGLE, it will be unstable since the general solution always contains the conserved HM as well. Thus, we can factorize out the HM from the general solution and write

$$W = W_0(1+w) = \eta e^{-(i\nu t+\phi_0)}\left[1+\sum_{n\neq 0} w_n(t)e^{i2\pi nx/L}\right]. \tag{4}$$

We now perform a linear stability analysis of the uniform oscillation [Eq. (3)] [2]. If we replace $W=W_0(1+w)$ in the MCGLE and keep only terms which are linear in w and w^* (the asterisk denotes complex conjugation), we can derive the stability conditions for the uniform oscillation,

$$\sqrt{(1-q^2)/2} < \eta, \tag{5}$$

$$1+\nu^2+(1+c_2^2)[3\eta^4+q^4]-4\eta^2(1+c_2\nu) + 2q^2[2(1+c_1c_2)-1-c_1\nu] > 0, \tag{6}$$

with $q=2\pi n/L$. There are four stability quadrants, depending on whether one relationship, none, or both are satisfied. Only in the latter case the uniform oscillation is stable. The first condition requires that $\eta > \eta_c \equiv 1/\sqrt{2}=0.7071$ and it is independent of other parameters. For either η or ν sufficiently high the positive terms going as η^4 and ν^2 in Eq. (6) dominate so that it is fulfilled. For $\eta\to\infty$ the uniform oscillation is everywhere stable. Finally, for finite η and ν and $1+c_1c_2 \leqslant 0$ (which corresponds to the Benjamin-Feir unstable regime of the unmodified CGLE) it is possible to violate Eq. (6).

Let us consider first the case where Eq. (5) is satisfied but Eq. (6) does not hold. In most of the patterns found in this regime, the collective oscillations to the base frequency are always discernible even when locally the behavior of phase and amplitude can be chaotic. They have spatially chaotic subharmonic oscillations resembling the spatiotemporal patterns found at the vicinity of multiple bifurcation points [15]. A wide variety of standing waves is also found, as is the case with linear global coupling [11] to which the MCGLE reduces when the nonlinearity of the coupling does not strongly rule the local dynamics. By increasing either ν or η the chaotic behavior can be entirely suppressed, the uniform oscillation [Eq. (3)] being stabilized.

Qualitatively different dynamics is obtained when Eq. (5) is violated and Eq. (6) holds. In this regime, cluster patterns are observed. We have carried out simulations with Eq. (1) by using a pseudospectral method with 1024 Fourier modes, periodic boundary conditions, and uniform noise superimposed to a localized pulse as initial condition and normalized so that $\int_{-L/2}^{L/2} W(x,0)dx/L=\eta$ (the initial phase ϕ_0 is taken to be zero). The integration is done with an exponential time stepping algorithm [16]. In Figs. 1(a) and 1(b) a value $\eta = 0.66 < \eta_c$ is considered so that now Eq. (5) does not hold but Eq. (6) does. The spatiotemporal evolutions of the modulus of the complex amplitude $|W|$ and the phase ϕ are plotted. A spatial pattern where the $L/2$ translation symmetry is broken is observed in $|W|$ in Fig. 1(a): the amplitude shifts $L/2$ spatially at the base frequency so that the same oscillatory state for each point in space is attained at a period that is twice the one of the base frequency ν. An inspection of the phase, plotted in Fig. 1(b), shows indeed two different spatial domains that oscillate with different phases. We can now perform a Fourier analysis of the complex amplitude in time, $W(x,t)=\int_{-\infty}^{\infty} a_\omega(x)e^{-i\omega t}d\omega$, at each position in space. In Fig. 1(c) the cumulative power spectrum of the time series $\langle |a_\omega|^2\rangle = \int_{-L/2}^{L/2}|a_\omega(x)|^2 dx$ is shown. Two peaks arise, one at the main frequency, ν, and the other at the subharmonic one, $\nu/2$. They correspond to the main active modes in the pattern. In Figs. 1(d) and 1(e) we plot, respectively, the arrangement of the phase in the complex plane corresponding to the modes at frequencies ν and $\nu/2$. Full synchronization of all oscillators is observed in the main mode, while in the subharmonic mode, a two-phase cluster exhibiting an Ising wall is observed. The same conclusions on the clustering to the main and the subharmonic mode were reached in [5] by means of a Karhunen-Loeve decomposition on the experimental data. It is interesting that these type-II clusters appear at a higher value of the coupling strength compared to the type-I clusters [5] which, in fact, do not require a NGC and it was found already with a nonlocal CGLE with *linear* global coupling [12]. The type-II clusters were also found in experiments with catalytic CO oxidation on PT (110) under peri-

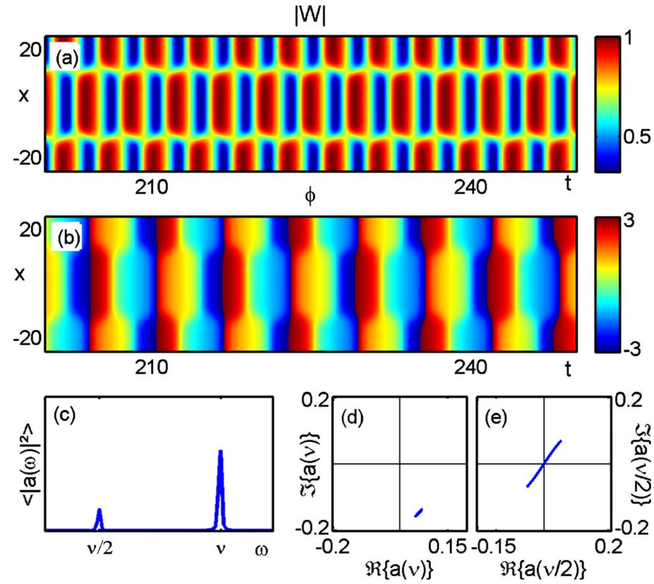

FIG. 1. (Color online) Spatiotemporal evolution of (a) the modulus $|W|$ and (b) the phase of the complex amplitude W obtained from Eq. (1) for $c_1=0.2$, $c_2=-0.58$, $\nu=1$, $L=50$, and $\eta=0.66$. (c) Space integrated power spectrum $\langle |a(\omega)|^2\rangle$ of the time series of the amplitude $W(x,t)$. (d) Projection on the complex plane of the Fourier coefficient $a(\nu)$ corresponding to the main frequency of the power spectrum at $\omega=\nu$ and (e) of the subharmonic coefficient at half the frequency $a(\nu/2)$.

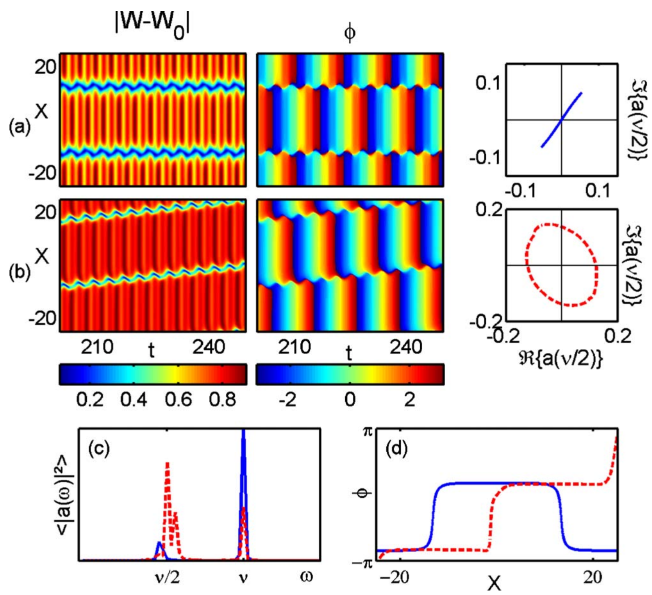

FIG. 2. (Color online) Spatiotemporal evolution of the modulus of the amplitude $|W-W_0|$ (left) and phase (right) after subtracting the HM and of the projection of the subharmonic $a(\nu/2)$ Fourier coefficient on the complex plane. (a) $\eta=0.66$; (b) $\eta=0.42$. (c) Space integrated power spectrum $\langle|a_\omega|^2\rangle$ of the time series of the amplitude and (d) spatial arrangement of the phase for the patterns in (a) (continuous line) and (b) (dashed line). Other parameter values as in Fig. 1.

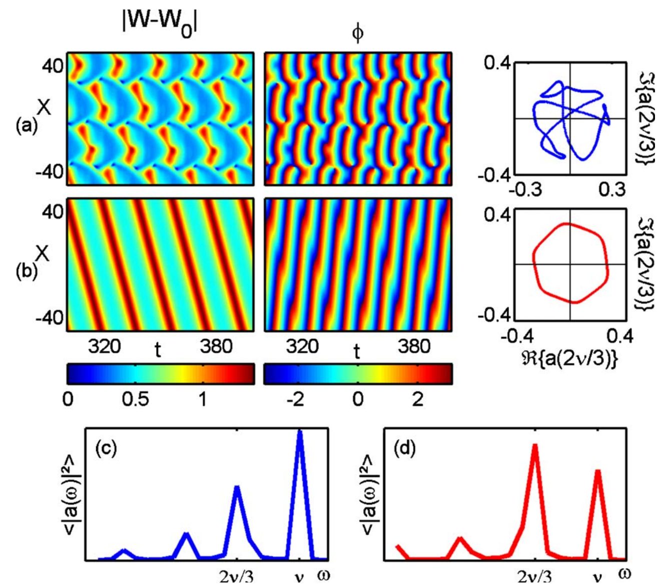

FIG. 3. (Color online) Spatiotemporal evolution of the modulus of the amplitude $|W-W_0|$ (left) and phase (right) after subtracting the HM and of the projection of the subharmonic $a(2\nu/3)$ Fourier coefficient on the complex plane. (a) $\eta=0.6$; (b) $\eta=0.5$. (c) and (d) Space integrated power spectrum $\langle|a_\omega|^2\rangle$ of the time series of the amplitude for patterns (a) and (b), respectively. Other parameter values are $c_1=-0.5$, $c_2=0.5$, $\nu=1$, and $L=100$.

odic uniform forcing [8] although no attempt was made to connect them to a normal form. In Fig. 2(a) the same pattern as in Fig. 1 is plotted after subtracting the HM. We observe clearly the Ising walls separating the two-phase clusters, as indicated by two dips in the amplitude $|W-W_0|$ in the spatiotemporal evolution where it goes to zero. With the subtraction of W_0 it is also apparent that the two domains are in antiphase. If we lower the coupling strength, which as we prove below is directly related to η^2, we find a nonequilibrium Ising-Bloch transition. The Ising wall loses stability to a Bloch wall as shown in Fig. 2(b). The walls separating the two-phase clusters in antiphase travel now with constant velocity and the amplitude in the center of the walls is no longer zero. In Fig. 2(c), we plot $|a_\omega|^2$ for the patterns in Figs. 2(a) and 2(b) (continuous and dashed curves, respectively) and observe that during the transition, the subharmonic peak at $\nu/2$ becomes higher than the one at ν. Finally, in Fig. 2(d) we show the spatial arrangement of the phase for the patterns in Fig. 2(a) (continuum curve) and Fig. 2(b) (dashed curve).

To clarify all these results, we can replace $W=W_0(1+w)$ [Eq. (4)] in the MCGLE so that we obtain a description of the spatiotemporal evolution of a general inhomogeneity w. By using also Eq. (3) and the fact that $\langle w\rangle$ and $\langle w^*\rangle$ both vanish we obtain the following expression:

$$\partial_t w = (\mu + i\beta)w + (1 + ic_1)\eta^2\partial_x^2 w - (1 + ic_2)\eta^2(|w|^2 w + w^*) + C, \tag{7}$$

where

$$C = (1 + ic_2)\eta^2[\langle 2|w|^2 + w^2\rangle - 2|w|^2 - w^2], \tag{8}$$

$\mu=1-2\eta^2$, and $\beta=\nu-2c_2\eta^2$. Equation (7) is, again, a CGLE modified by a term which is proportional to $\eta^2 w^*$. *This term introduces a 2:1 resonance* as in the well-studied parametric CGLE [4,17,18]. There is, however, a further modification in our case through the term C that adds a wide variety of interesting behavior and further resonances as shown below. Let us look for fixed points corresponding to two-phase clusters by replacing $w=Re^{i\chi}$ in Eq. (7). The term C vanishes in such a situation and we are left with the equation $0=(\mu+i\beta)-(1+ic_2)\eta^2(R^2+e^{-i2\chi})$, which is invariant upon a change $\chi\rightarrow\chi+\pi$, so that if χ is a solution, so is $\chi+\pi$. Multiplying this equation by its complex conjugate and solving for R and χ we obtain a pair of solutions $w_\pm=Re^{i\chi_\pm}$, where $R^2=[\mu+c_2\beta+\sqrt{(1+c_2^2)^2\eta^4-(\beta-c_2\mu)^2}]/(1+c_2^2)\eta^2$, $\chi_-=\frac{1}{2}\arcsin[(c_2\mu-\beta)/(1+c_2^2)\eta^2]$, and $\chi_+=\chi_-+\pi$. These solutions correspond to two-phase clusters which are contained for a given ν in $\eta_b<\eta<\eta_c$. They arise through a saddle-node bifurcation at $\eta_b^2=|c_2\mp\nu|/\sqrt{1+c_2^2}$, where the borders of the 2:1 resonance tongues are located [17]. Equation (7) contains, indeed, more fixed points than the ones considered above for the 2:1 resonance. For fixed points $Re^{\chi\pm}$ corresponding to two-phase clusters, C in Eq. (7) vanishes. However, this is *a particular case*. There can exist fixed points as well for which, for example, globally, $\langle|w|^2\rangle-|w|^2$ and $\langle w^2\rangle$ cancel out each other and then $C=-(1+ic_2)\eta^2 w^2$. These other fixed points *coexist* with the ones related to two-phase clusters, and there exist higher order resonances as well because of the *NGC in the inhomogeneities* present in C. Indeed, in the same stability regime of the MCGLE as before, we can find parameter values, where an Ising-Bloch transition in six-phase clusters is observed at a subharmonic peak at two third of the base frequency. This is shown in Fig. 3 where we observe again that the Ising-Bloch transition occurs when the subharmonic peak at $2\nu/3$ becomes higher than the main peak at ν by lowering the coupling strength η. This is a remarkable aspect of the NGC present in the MCGLE. While with the parametrically forced CGLE higher resonances are required to reproduce such behavior [18], in the MCGLE they arise naturally from the NGC, which, in turn, is simply related to the conservation of the oscillatory state of the HM. Indeed, in experiments with electrochemical systems under global coupling a wide variety of clustering behavior was observed, including five-phase clusters *without breaking phase invariance*. In this Rapid Communication, we have provided a simple and general explanation for this possibility, going well beyond electrochemical systems, and we have shown how the conservation of the oscillatory state of the HM allows the phase invariance to be preserved while allowing the excitation of phase-balanced subharmonic clustering states. Since the 2:1 resonance is the main one arising from the NGC, it is now also evident why it is prevalent in parameter space.

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