## Pattern Formation and Localized Structures in Degenerate Optical Parametric Mixing

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We report the pattern formation of degenerate optical parametric mixing—a system with the realvalued order parameter. The phenomena have similarities with and extend those of chemical and hydrodynamic systems described by real order parameter equations, and are different from laser-type pattern formation, where the order parameter is complex. The structure formation is based on a phase bistability. Structures are dominated by fronts separating domains of opposite phase. The domains expand or shrink depending on resonator detuning. Small circular domains, however, are stable and constitute the localized structures of the system. The phenomena are demonstrated experimentally using degenerate four-wave mixing as a prototype process in a resonator of a large Fresnel number. [S0031-9007(98)07079-3]

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The light generated by nonlinear optical systems can have a free phase as in lasers, or there are preferred phase values as in degenerate wave mixing. The first class of nonlinear optical systems is described by complex order parameter equations, while for the second class the order parameter is real valued. The pattern formation in the first class of systems has been studied in detail recently theoretically [1,2] and experimentally [3,4], so that a reasonably coherent picture of its structure formation exists. The second class has so far been investigated only theoretically. Stripe patterns (Turing structures) were predicted for degenerate optical parametric oscillators [5], along with the quantum properties of the generated fields [6]. Recently, the existence of black fronts separating phase domain has been discussed [7] and analyzed in detail [8]. It was shown in [8] that the dynamics of the fronts depends on the detuning. In [8,9] it was shown that small circular domains are stable for a certain detuning range, thus constituting (bistable) localized structures ("Ising" structures [10]) of the system.

In this Letter we prove experimentally the structure formation, the dynamics of the phase domains, and the existence of the localized structures (spatial solitons), as theoretically predicted in [8].

A large class of pattern forming systems in physics is described by real order parameter equations, e.g., the periodically forced Belousov-Zhabotinsky reaction [11], driven space charge wave fields [12], and Rayleigh-Benard convection [13]. For these Turing patterns (rolls, hexagons) [14,15] are characteristic. The results reported here are therefore characteristic not only for optics, but for a large class of systems in physics and nature. We note that the mentioned systems [11–13] represent particular cases for the detuning parameter. As opposed, optical systems constitute the general case as they permit us to work at arbitrary detuning values.

The spatiotemporal dynamics in these systems can be described by the real Swift-Hohenberg equation

(SHE) [16]:

$$\partial_t A = A - A^3 - (\nabla^2 + \Delta)^2 A \tag{1}$$

with the real-valued order parameter  $A(\vec{r})$ , defined in 2D space  $\vec{r} = (x, y)$  and evolving in time *t*.

Turing patterns occur for large positive detuning  $\Delta$  of (1), e.g., in the form of stripes  $A(\vec{r}) \approx \sqrt{4/3} \cos(\vec{k} \cdot \vec{r})$  with a resonant wave number  $|\vec{k}| = \sqrt{\Delta}$ . This limit of large detuning is characteristic, e.g., for Rayleigh-Benard convection, where convective rolls have a defined size. The zero detuning limit of SHE (1) describes parametrically excited systems [11,12], for which solutions of homogeneous amplitude and phase of  $\varphi_0 = 0$  or  $\pi$  are characteristic. Optical parametric oscillators allow us to realize pattern formation for the whole detuning range. The patterns are contracting or expanding domains, and the spatial localized structures [8,9].

Familiar degenerate parametric systems in nonlinear optics are the degenerate four-wave-mixing oscillator (DFWMO) and the degenerate optical parametric oscillator, both described by Eq. (1) [16]. For the experiments a DFWMO with a slow material (photorefractive BaTiO<sub>3</sub>) was chosen. The characteristic time scale at the light intensities used is 1 s, permitting recording with ordinary video equipment. The experimental scheme is given in Fig. 1: Two counterpropagating pump beams (single frequency Ar<sup>+</sup> laser at 514.5 nm, beam width of 20 mm, typical intensities of 100 mW/cm<sup>2</sup>) illuminate the photorefractive BaTiO<sub>3</sub> crystal (of  $4.3 \times 4.3 \times 4.6$  mm dimensions) mounted inside a near-self-imaging resonator. In the limit of precise self-imaging such a resonator has an infinite Fresnel number (within the limits of the paraxial aproximation), and all transverse modes are exactly degenerate, allowing the resonance of arbitrary images [4]. Increasing the resonator length with respect to the self-imaging length by l makes the resonator equivalent to a plane-plane resonator of length l. In the experiment lwas 30 mm, which corresponds to a characteristic spatial scale  $\Delta x_0 = 100 \ \mu m \ (\Delta x_0 \approx \sqrt{\lambda l} \ [4])$ . The degeneracy



FIG. 1. Near self-imaging resonator used for experiments. M: mirrors, f: focal lengths of lenses, l: deviation from self-imaging length, D: diaphragm, filtering high transverse modes.

of generated waves is achieved by propagation in the same resonator. To control the resonator tuning the optical length of the resonator was actively stabilized to the pump frequency in a manner described in [17].

The variation of the resonator length on an optical wavelength scale allows us to choose the detuning parameter. The pump intensity was fixed to approximately 70% above generation threshold. Only the detuning was varied. Typically we observe emission, as predicted, in domains separated by black lines of irregular shape [Fig. 2(a)]. The domain boundaries can have quite a complicated structure, including self-crossings, and move in general. In Fig. 2(b) a part of near field distribution is shown, together with an interferogram [Fig. 2(c)] showing the phase difference between domains. The phase changes by  $\pi$  across the domain boundaries, which shows the real-valued nature of the field.

We use the results reported in [8] as a guide for the experiments, where domain dynamics was shown to depend



FIG. 2. Domains of complicated shape as obtained experimentally: (a) The near field of the whole beam, (b) a small section from the whole beam, and (c) interferogram, showing that the adjacent domains have opposite phase.



FIG. 3. Evolution of domains, depending on the detuning as obtained numerically by solving SHE (1) with zero boundary conditions. The initial distribution for calculations with different detunings (different columns) was the same (top plot). The rows correspond to times t = 15, and t = 150. The size of the integration region is  $\Delta x = \Delta y = 45$ . This corresponds to a Fresnel number  $F = 2 \times 10^3$ . Details for computation can be found in [8].

on the detuning. In Fig. 3 we summarize qualitatively the domain dynamics.

For small detuning (first column in Fig. 3), where the homogeneous states are strongly preferred, the domains shrink, and disappear eventually. The domain boundaries behave like elastic ribbons. Corresponding experimental recordings near zero detuning are given in Fig. 4, showing the shrinking of a domain boundary. The domain boundaries disappear finally, and the homogeneous field is the final state (experimentally as well as numerically).

At large detunings the domains grow, and a labyrinth structure develops, as shown in the third column of Fig. 3. The dark lines behave like the antielastic ribbons (negative elasticity coefficient). Reconnection of the black lines is not observed in this region. Domains neither disappear, nor do new domains nucleate; thus the topology of the initial structure is preserved. Such a topology-preserving expansion of domain boundaries, as recorded experimentally, is shown in Fig. 5. The domains expand until the space is filled, and the final state, the "labyrinth" is reached [Fig. 5(d)].

The labyrinth structure is a Turing structure with a characteristic spatial scale determined by the detuning.



FIG. 4. A shrinking domain boundary; experimental; detuning near zero. Time between successive snapshots is 2 s.



0.5 mm

FIG. 5. Expanding domain boundaries ending in a labyrinth structure. Experiment. Detuning is around 0.7 (normalized to the width of the resonator mode). Time between snapshots a, b, c is 2 s. Time between snapshots c and d is 10 s.

The labyrinth can also be considered as a stripe pattern with defects. Such kinds of Turing structures were extensively investigated outside optics [15]. One example of such a structure in optics was shown in [18].

For larger values of detuning the homogeneous solution is modulationally unstable. Therefore domains not only grow, but nucleation of new domains, and also reconnection of the black lines is possible, as shown in the fourth column in Fig. 3. Such cases are shown in Figs. 6 and 7 experimentally. In Fig. 6 the appearance of new domains is clearly seen. Figure 7 shows reconnection of domain boundaries. The pattern is roughly a stripe pattern here with adjacent bright stripes having the opposite phase. From 7(a) to 7(d) the stripe in the middle of the picture splits into two separate domains, while the domains to the left and to the right of the domain in the middle become connected.

For intermediate values of detuning, where the elasticity of the domain boundaries is close to zero, the shrinking/expansion of the domains can stop at a particular (minimum) domain size so that a localized structure is formed [8,9]. One can interpret the phenomenon as a balance between the surface tension (elasticity of the domain boundary) and the internal pressure of a "bubble." The balance can also be understood considering that every segment of a dark line interacts with the segment of the dark line on the opposite side of the localized structure. Formation of these localized structures is illustrated by the



FIG. 7. Breaking and new connection of domains. Experiment. Detuning is the same as that of Fig. 6. Time between snapshots is 2 s.

second column in Fig. 3 as obtained numerically, and by Fig. 8 as recorded experimentally. Figure 8(a) is taken in the transient phase in which the domains shrink and Fig. 8(b) when all domains have shrunk to their minimum diameter. We observe experimentally that the domains remain stable as long as the detuning is kept in a finite detuning parameter range. Consequently we conclude that the circular domains shown in Fig. 8(b) are the supercritically bistable localized structures predicted in [8,9]

In conclusion we have observed experimentally the structure formation of a four-wave-mixing process prototypical for optical parametric degenerate mixing, in qualitative agreement with the predictions of the Swift-Hohenberg equation describing parametric processes in general. We show the change of domain structure with detuning as predicted in [8,9], and we observe the predicted bistable localized structures. The observed localized structures are based on a supercritical bistability (phase bistability), as opposed to the common subcritical bistability. The relative simplicity of the field structure with the order parameter being a real function, of this class of optical systems, allows us to obtain a relatively complete understanding of its structure formation.

All phenomena predicted by SHE (1) (which is strictly valid near generation threshold) were observed in the experiment, where the gain was moderately above threshold. This does of course not exclude the existence of phenomena or structures additional to the ones of (1) under these experimental conditions.



0.5 mm

FIG. 6. Nucleation of new domains. Experiment. Detuning is slightly larger than for Fig. 5. Time between snapshots is 2 s.



FIG. 8. Localized structures based on supercritical bistability: (a) Transient shrinking of the domains and formation of localized structures; (b) stable localized structures. Experiment. Detuning is in between that of Figs. 4 and 5. Time between snapshots is 10 s.

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