PHYSICAL REVIEW E VOLUME 59, NUMBER 1 JANUARY 1999

Quasipatterns in second-harmonic generation

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(Received 3 September 1998)

A cascade of two-dimensional quasipatterns is predicted in intracavity optical frequency doubling due to a polarization instability at the fundamental frequency. Amplitude equations derived from a microscopic model of the frequency conversion process show that quasiperiodic structures of arbitrary orientational order can be spontaneously selected due to bulk nonlinear effects when detuned operation for both fundamental and second-harmonic fields occurs. Multistability and coexistence of patterns and quasipatterns is also discussed. [S1063-651X(99)50401-7]

PACS number(s): 05.45.−a, 47.54.+r

The study of two-dimensional spatial structures with quasicrystalline order in spatially extended pattern forming systems [1,2] has received an increasing interest in recent years, especially since the first experimental observations of quasiperiodic patterns in hydrodynamic [3] and nonlinear optical [4] systems. Quasipatterns of different orientational orders have been observed in Faraday wave experiments [3,5] and corroborated by theoretical models [2,6]. In the optical context, quasipatterns have been predicted and experimentally observed in $\chi^{(3)}$ (cubic) nonlinear systems, including liquid-crystal light valve devices with imposed feedback rotation [4] and single-mirror feedback devices with continuous rotational symmetry [7–9]. Regardless of the specific physical system, it was pointed out [2,6,8,9] that stability of quasipatterns in isotropic systems with a continuous rotational symmetry requires the existence of a complicated nonlinear mode coupling acting on a single band of active modes or, differently, the existence of triadic resonances among modes belonging to two different bands of active modes [2]. In most boundary-free hydrodynamic and optical systems, the bulk nonlinearity is usually rather simple and interband resonances are essential for the observation of quasipatterns [2,9]. Although in real systems boundary effects can influence the bulk nonlinearity, making possible the existence of quasiperiodic structures involving a single band of active modes [2,5,6], so far no examples of complex pattern formation, which do not require interband resonances nor boundary constraints, have been reported. In a recent work [8], Leduc *et al.* showed that, in a single-feedback-mirror optical device, the bulk nonlinearity can support quasipatterns of eightfold orientational order, but the relatively simple mode coupling function does not allow for the appearance of quasipatterns with higher orientational orders.

The aim of this Rapid Communication is to show that spontaneous formation of quasipatterns of *arbitrary* orientational orders is possible in a nonlinear optical process involving a *quadratic* nonlinearity, namely nondegenerate second-harmonic generation (SHG) in an optical cavity. Furthermore, it is shown that multistability and coexistence of patterns and quasipatterns of different symmetries is possible in this nonlinear optical system.

The starting point of the analysis is provided by the mean-field model for type-II second-harmonic generation in a plane-plane optical cavity containing a nonlinear $\chi^{(2)}$ crystal, externally driven by a linearly polarized plane-wave fundamental field [10,11]. For a symmetric pumping configuration [10], the dynamical equations for the normalized second-harmonic field envelope A_0 at frequency 2ω and for the two orthogonally polarized fundamental-frequency field envelopes A_1, A_2 at frequency ω read [10]

$$\partial_t A_0 = \gamma_0[-(1+i\Delta_0)A_0 + ia_0\nabla^2 A_0] - \gamma_0 A_1 A_2,$$
$$\partial_t A_1 = \gamma[-(1+i\Delta)A_1 + ia\nabla^2 A_1] + \gamma A_0 A_2^* + \gamma E, \quad (1)$$
$$\partial_t A_2 = \gamma[-(1+i\Delta)A_2 + ia\nabla^2 A_2] + \gamma A_0 A_1^* + \gamma E,$$

where E is proportional to the amplitude of the incident plane-wave pump field at frequency ω, linearly polarized at 45° with respect to the crystal axes, γ_0 and γ are the cavity decay rates for the second-harmonic and the fundamental frequency fields, respectively, a_0, a, and Δ_0, Δ are the diffraction ($\gamma a \sim 2\gamma_0 a_0$) and the cavity detuning parameters for the two fields, respectively, and $\nabla^2 = \partial_x^2 + \partial_y^2$ is the transverse Laplacian. In writing Eqs. (1), we assumed same cavity detuning parameters and same cavity losses for the two orthogonally polarized fundamental fields. Equations (1) have the homogeneous symmetric solution given by $A_1 = A_2 = \sqrt{X}\exp(i\phi)$, $A_0 = -X\exp(2i\phi)/(1+i\Delta_0)$, where $X>0$ is a solution of the cubic equation $[(1+X-\Delta_0\Delta)^2+(\Delta_0+\Delta)^2]X=(1+\Delta_0^2)|E|^2$. This solution corresponds to an intracavity fundamental-frequency field that has the same polarization state as that of the external driving field. As the amplitude E of the external driving field is increased, lose of stability occurs due to the appearance of a symmetry-breaking *polarization* instability at the fundamental-frequency [10–12]. For a negative detuning Δ of the fundamental field, the wave numbers $\mathbf{k}$ of the most unstable perturbations lie on the critical circle of radius $k_c = \sqrt{-\Delta/a}$, and the instability threshold is reached for $|E| = E_c$, where $E_c^2 = (2+\Delta^2)(1+\Delta_0^2)^{1/2} + 2(1-\Delta_0\Delta)$. Although in the linear stage of the instability all the neutral modes are equally amplified, due to their nonlinear competition few of them typically survive and saturate, leading to the formation of regular patterns. The mode competition can be studied by the derivation of amplitude equations in a multiple-scale asymptotic analysis of the field equations (1) near the instability point [13]. This is done by looking for a solution of Eqs. (1) as an asymptotic expansion in a smallness parameter ϵ, $\mathbf{v}=\mathbf{v}^{(0)}+\epsilon\mathbf{v}^{(1)}+\epsilon^2\mathbf{v}^{(2)}+\cdots$, where $\mathbf{v}=(A_0,A_1,A_2)^T$ contains the field variables, $\mathbf{v}^{(0)}$ is the homogeneous symmetric solution at $|E|=E_c$, $\epsilon\mathbf{v}^{(1)}$ is the bifurcating solution in the linearized problem, and $\epsilon=(|E|-E_c)^{1/2}$ provides a measure of the distance far from the instability point. The solution at $O(\epsilon)$ can be taken as $\epsilon\mathbf{v}^{(1)}=(0,\psi,-\psi)^T$, where ψ is a real function given by an arbitrary superposition of neutral modes on the critical circle

$$\psi=\sum_{n=1}^{2M} F_n\exp(i\mathbf{k}_n\cdot\mathbf{x}). \tag{2}$$

In Eq. (2), F_n is the complex amplitude of the nth mode, $|\mathbf{k}_n|=k_c$, $\mathbf{k}_{n+M}=-\mathbf{k}_n$, $F_{n+M}=F_n^*$, and M is the number of competing modes. The time evolution of the amplitudes F_n, as obtained from solvability conditions at $O(\epsilon^3)$ in the asymptotic expansion, read explicitly [13]

$$\frac{1}{\gamma}\partial_t F_n=\mu F_n-\left(\sum_{m=1}^{M}\beta(\vartheta_{mn})|F_m|^2\right)F_n \tag{3}$$

$(n=1,2,3,...,M)$. In Eq. (3),

$$\mu=2E_c[(4+\Delta^2)\sqrt{1+\Delta_0^2}+4(1-\Delta_0\Delta)]^{-1}(|E|-E_c),$$

ϑ_{mn} is the angle between the wave vectors $\mathbf{k}_m$ and $\mathbf{k}_n$, and the coupling function $\beta(\vartheta)$ is given by

$$\beta(\vartheta)=\begin{cases}2g(0)+g(\pi) & \text{if } \vartheta=0\\ 2[g(0)+g(\vartheta)+g(\pi+\vartheta)] & \text{otherwise}\end{cases} \tag{4}$$

where

$$g(\vartheta)=\frac{\delta^2+2(1+\sqrt{1+\Delta_0^2})}{\delta^2+4\sqrt{1+\Delta_0^2}+(\delta_0\delta-2\sqrt{1+\Delta_0^2})^2}, \tag{5}$$

and

$$\delta(\vartheta)=-ak_c^2\left[1-4\sin^2\frac{\vartheta}{2}\right]=\Delta\left[1-4\sin^2\frac{\vartheta}{2}\right],$$

$$\delta_0(\vartheta)=\Delta_0+4a_0k_c^2\sin^2\frac{\vartheta}{2}=\Delta_0-2\Delta\frac{\gamma}{\gamma_0}\sin^2\frac{\vartheta}{2}.$$

After introduction of the polar decomposition $F_n=R_n\exp(i\phi_n)$, from Eqs. (3) it turns out that the phases ϕ_n are constants (i.e., $\partial_t\phi_n=0$), whereas the dynamics for the real amplitudes R_n can be represented in the gradient form $\partial_t R_n=-\delta L/\delta R_n$, where the Lyapunov function (or free-energy) $L=L(R_n)$ is given by [1,2]

$$L=-\frac{1}{2}\mu\sum_{n=1}^{M}R_n^2+\frac{1}{4}\sum_{m,n=1}^{M}\beta(\vartheta_{mn})R_n^2R_m^2. \tag{6}$$

The amplitude equations (3) have a class of solutions, involving N pairs of oppositely oriented wave vectors $\pm\mathbf{k}_1,\pm\mathbf{k}_2,\ldots,\pm\mathbf{k}_N$ equally spaced on the critical circle and with equal amplitudes $R_n=\mu^{1/2}(\Sigma_{m=0}^{N-1}\beta(m\pi/N))^{-1/2}$ $(n=1,2,3,...,N)$, which correspond to periodic $(N\leqslant 3)$ or quasiperiodic $(N>3)$ patterns with $2N$-fold orientational order

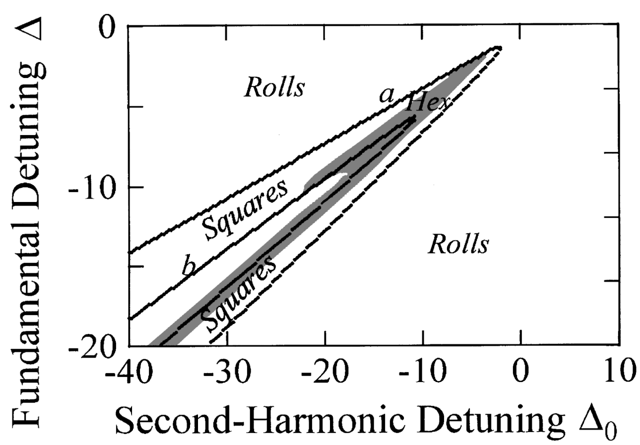

FIG. 1. Stability domains of roll, square, and hexagonal patterns in the (Δ_0,Δ) plane for $\gamma_0=\gamma$. Curve a marks the stability boundary of rolls, whereas curve b delimits the stability boundary of squares with respect to external perturbations. Squares are linearly stable in the domain contained between curves a and b. The dashed area corresponds to the stability domain of hexagons (Hex). Note that there exists bistability between square and hexagonal patterns, and that in a region inside the domain delimited by curve b all periodic patterns are unstable.

[1,2]. A linear stability analysis indicates that the $2N$-folded pattern is locally stable provided that [1,13]

$$\sum_{n=0}^{N-1}\beta(n\pi/N)\cos\left(\frac{2\pi nl}{N}\right)\geqslant 0 \tag{7}$$

for each $l=1,2,...,N-1$, and

$$\sum_{n=0}^{N-1}\beta\left(\vartheta+\frac{n\pi}{N}\right)\geqslant\sum_{n=0}^{N-1}\beta\left(\frac{n\pi}{N}\right) \tag{8}$$

for any $\vartheta\in(0,\pi/2N]$. Equation (7) represents the stability criteria with respect to *internal* perturbations (i.e., perturbations of modes belonging to the pattern), whereas Eq. (8) arises from the stability requirement against *external* perturbations [1]. The above conditions, together with Eqs. (4) and (5), can be used to determine the *local* stability domains in the parameter space of the various patterns, and the domains of multistability [14]. In case of multistability, the *global* stability can be investigated by comparing the values of the Lyapunov function in correspondence of the different values of N, $L_N=-\mu^2N/[4\Sigma_{n=0}^{N-1}\beta(n\pi/N)]$, the relaxational dynamics of the system favoring the regular pattern which minimizes the free-energy.

In Fig. 1 the domains of local stability for rolls, squares, and hexagons as derived from Eqs. (7) and (8) are shown in the (Δ_0,Δ) plane for $\gamma=\gamma_0$. As can be seen, the stability domain of hexagonal patterns (shaded area) shares a common region with the stability domain of square patterns (lying between curves a and b), indicating the possibility of coexistence between square and hexagonal patterns. A comparison of the free-energy for these two patterns in the bistability region indicates that squares correspond to the absolute minimum and hexagons to a local minimum of the free-energy. Most interesting, in a wide region inside curve a, roll, square, and hexagonal patterns are unstable, which is a

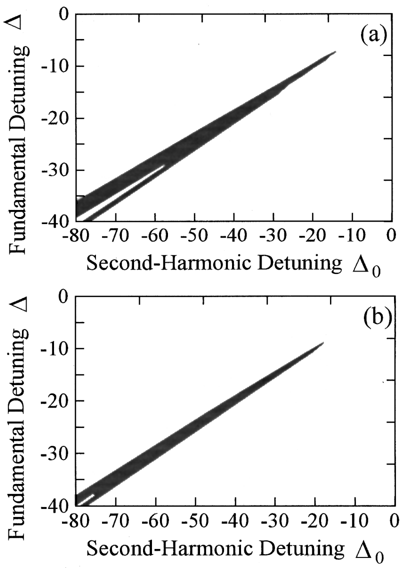

FIG. 2. Stability domains (dashed areas) of (a) decagonal, and (b) dodecagonal quasipatterns in the (Δ_0,Δ) plane for $\gamma_0=\gamma$.

signature of the existence of quasipatterns. Indeed, from Eqs. (7) and (8) we found that quasipatterns with an arbitrary orientational order are stable in certain domains of the parameter space. As an example, Fig. 2 shows the local stability domains of decagonal ($N=5$) and dodecagonal ($N=6$) quasipatterns in the (Δ_0,Δ) plane. The figure also shows the possibility of coexistence of these two quasipatterns. In general, multistability involving more than two patterns or quasipatterns can be found [14]; as an example, for $\Delta=-15$, $\Delta_0=-30$ and $\gamma=\gamma_0$, it turns out that octagonal, decagonal, and dodecagonal quasipatterns are linearly stable, whereas for $\Delta=-16$, $\Delta_0=-30$, and $\gamma=\gamma_0$ multistability among hexagons, octagons, and decagons is possible. The stability properties of patterns and quasipatterns are determined basically by the shape of the coupling function $\beta(\vartheta)$, which rules the nonlinear interaction among the neutral modes on the critical circle [2]. In particular, a necessary condition for the stability of quasipatterns is that $\beta(\vartheta)$ be smaller than $\beta(0)$ in some domain of the interval $[0,\pi]$. For the coupling function given by Eqs. (4) and (5), it turns out that, for a positive value of the second-harmonic detuning Δ_0 or for low values of the detuning parameters Δ and Δ_0, $\beta(\vartheta)$ reaches its minimum value at $\vartheta=0,\pi$, so that the patterns of lowest free-energy are rolls, i.e., $L_1\leqslant L_N$ for all N (see also Fig. 1). However, when the second-harmonic detuning is negative, the condition $\beta(0)>\beta(\vartheta)$ can be satisfied in some domain and higher-order regular patterns can be selected. In particular, an inspection of Eqs. (4) and (5) reveals that the condition $\beta(0)>\beta(\vartheta)$ can be satisfied in a wide domain of the interval $[0,\pi]$ whenever $\Delta_0\sim-4ak_c^2=2\gamma\Delta/\gamma_0$ and $|\Delta_0|$ is large. As an example, Fig. 3 shows the shape of the normalized function $\beta(\vartheta)/\beta(0)$ when $\Delta_0=2\gamma\Delta/\gamma_0$ for some values of the fundamental-frequency detuning Δ. As $|\Delta|$ is increased (with $\Delta_0=2\Delta\gamma/\gamma_0$), the region where $\beta(\vartheta)<\beta(0)$ becomes wider and, as a consequence, the minimum of the free-energy, L_N, is expected to occur at increasing values of N. This is shown in Fig. 4, where the order N of the regular pattern which minimizes the free-energy is plotted as a function of the fundamental-frequency detuning Δ for $\gamma=\gamma_0$

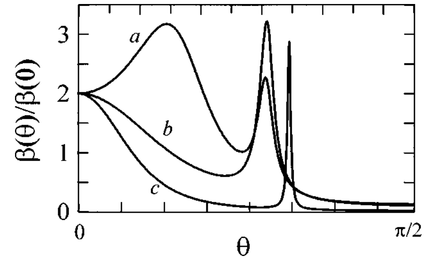

FIG. 3. Behavior of the normalized coupling function $\beta(\vartheta)/\beta(0)$ in the interval $[0,\pi/2]$ for $\gamma=\gamma_0$, $\Delta_0=2\Delta$ and for different values of the detuning Δ: *a*: $\Delta=-4$; *b*: $\Delta=-16$; *c*: $\Delta=-25$. Note that $\beta(\vartheta)$ is discontinuous at $\vartheta=0$ with $\beta(\vartheta\to 0)=2\beta(0)$.

[Fig. 4(a)] and for $\gamma=10\gamma_0$ [Fig. 4(b)]. As it can be seen, a cascading of patterns with increasing rotational symmetry is predicted for increasing values of detuning parameters. Notice that patterns corresponding to some orientational orders are missed in Fig. 4, although they have been found to be locally stable and, hence, observable. As an example, in Fig. 4(a) quasipatterns corresponding to the orders $N=4,6,9,...$ are not selected as $|\Delta|$ is increased, although in a wide interval of the curve $\Delta_0=2\Delta$ they satisfy the local stability conditions [Eqs. (7) and (8)]. This is due to the presence, in the shape of $\beta(\vartheta)$, of two peaked maxima located at $\vartheta=\vartheta_m$, $\pi-\vartheta_m$ [where ϑ_m is determined by imposing $(\partial\beta/\vartheta)_{\vartheta_m}=0$; see Fig. 3], which do not favor selection of wave vectors oriented along such directions.

It is worth discussing, as a conclusive point, the physical mechanism underlying the spontaneous appearance of quasipatterns in the intracavity frequency conversion process when a detuned configuration is used. The polarization instability at the fundamental frequency is responsible for the growth of a set of off-axis modes $\{\mathbf{k}_l\}$ on a critical circle at the expense of the second-harmonic field already established in the cavity. This instability can be viewed, in fact, as a parametric oscillation process for a polarization component at the fundamental frequency which draws its energy from the second-harmonic field [10]. The linearly growing modes can be frequency up-converted again in the nonlinear crystal, and saturation of the linear growth occurs. In particular, the frequency up-conversion process will generate all possible spatial components $\mathbf{s}=\mathbf{k}_l+\mathbf{k}_n$ at frequency 2ω. As this process tends to limit the growth of linearly unstable modes, the instability will favor the growth of mode combinations for which the up-conversion process is less efficient. The up-conversion efficiency is strongly dependent on the cavity

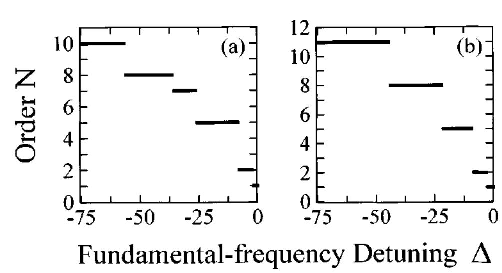

FIG. 4. Order N of the regular pattern which minimizes the free-energy as a function of the detuning parameter Δ for (a) $\gamma=\gamma_0$ and (b) $\gamma=10\gamma_0$. The detuning for the second-harmonic field is $\Delta_0=2\Delta$ in (a) and $\Delta_0=5\Delta$ in (b).

phase matching term $\Delta\phi=\Delta_0+a_0|\mathbf{s}|^2$, the conversion efficiency being maximized at perfect phase-matching ($\Delta\phi=0$). If we now consider the case of a large (and negative) second-harmonic cavity detuning Δ_0, the up-conversion process is generally less efficient; however, if the fundamental-frequency cavity detuning Δ is large too and close to $\Delta\sim\gamma_0\Delta_0/2\gamma$, the phase matching $\Delta\phi$ vanishes when $|\mathbf{s}|\sim 2k_c$, i.e., the up-conversion process becomes very efficient for self-interaction terms (i.e., when $\mathbf{k}_n=\mathbf{k}_l$). This phenomenon is reflected in the shape of the nonlinear coupling function $\beta(\vartheta)$, which is large near $\vartheta\sim 0$ (see Fig. 3), and explains the tendency of the polarization instability to favor structures with an increasing rotational symmetry. It should be pointed out that the values of the detunings needed for the observation of quasipatterns of high orientational orders are quite large, and in this case a more complete model, which takes into account longitudinal field variations, should be used. For the same reason, numerical simulations aimed to observe quasipatterns are not trivial in such regions of the parameter space.

In conclusion, a theoretical analysis of complex pattern formation for a mean-field model of intracavity optical frequency doubling has been presented. This analysis provides a remarkable example in nonlinear optics of spontaneous formation of quasipatterns with an arbitrary orientational order and of coexistence of patterns with different symmetries.

The author wishes to thank Dr. D. Michaelis for sharing Refs. [11] prior to their publication.

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[12] We consider here parameter values where the polarization instability prevails over a competing instability characteristic of the scalar SHG (see [10]). For values of detuning parameters here considered where bistability occurs, it can be shown that the polarization instability always arises below the lower bistability turning point [13].

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[14] Precise limitations to the existence of pattern multistability are set by Eqs. (7) and (8). It can be shown [13] that regular patterns of orders N and N' can not coexist whenever N'/N is an integer. In particular, rolls ($N=1$) cannot coexist with all other patterns.