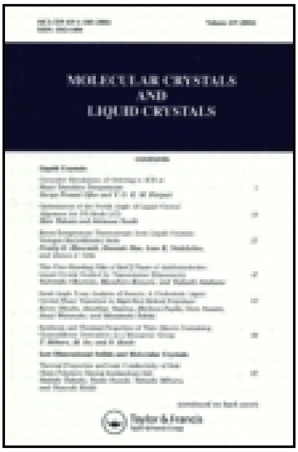

Molecular Crystals and Liquid Crystals Science and Technology. Section A. Molecular Crystals and Liquid Crystals

Publication details, including instructions for authors and subscription information:
http://www.tandfonline.com/loi/gmcl19

Spontaneous Pattern Formation in Nonlinear Optical Systems

William J. Firth [a]

[a] Department of Physics and Applied Physics, University of Strathclyde, Glasgow, G4 0NG, Scotland.

Version of record first published: 24 Sep 2006.

To cite this article: William J. Firth (1994): Spontaneous Pattern Formation in Nonlinear Optical Systems, Molecular Crystals and Liquid Crystals Science and Technology. Section A. Molecular Crystals and Liquid Crystals, 251:1, 111-125

To link to this article: http://dx.doi.org/10.1080/10587259408027197

Mol. Cryst. Liq. Cryst. 1994, Vol. 251, pp. 111–125
Reprints available directly from the publisher
Photocopying permitted by license only

Printed in the United States of America

SPONTANEOUS PATTERN FORMATION IN NONLINEAR OPTICAL SYSTEMS

WILLIAM J. FIRTH
Department of Physics and Applied Physics, University of Strathclyde, Glasgow G4 0NG, Scotland.

Abstract Some of the pattern formation and related processes which occur in nonlinear optical systems are described. Among such systems liquid crystal devices are prominent, because they can exhibit very large optical nonlinearity, making it relatively easy to achieve significant nonlinear effects over large areas. A Kerr slice with single feedback mirror forms hexagonal patterns for plane waves, but its pattern-forming and dynamic properties change significantly for gaussian beam input. Liquid crystal light valve feedback loops are seen to be closely related to the single-mirror system, and are used in some striking recent experiments. Other pattern-forming systems, such as lasers, are briefly reviewed.

INTRODUCTION

This article reviews some of the pattern formation and related processes recently described in nonlinear optical systems. Among such systems liquid crystal devices are prominent, because they can exhibit very large optical nonlinearity. While this is usually accompanied by a response time in the millisecond range, this is actually an advantage for demonstration systems, since pattern formation and evolution can be viewed directly by eye and/or recorded on standard video equipment. Liquid crystal films are usually used in *passive* systems, which means that the excitation is by an external driving field - smooth and constant in the ideal case - rather than through population inversion. As well as regular patterns, we will consider some of the *defect* structures and *turbulent* behaviour observed under strong excitation. These higher-excitation phenomena may well hold the key to the practical application of these effects in information processing.

Much of the discussion will turn on analysis and simulations, because experimental confirmation of these phenomena is still somewhat limited. Nevertheless, it is encouraging that most of the models discussed have some positive experimental evidence in their support, and that that body of evidence is growing at quite a rapid rate.

Most of the systems examined employ a third-order optical nonlinearity, of which the optical Kerr effect is the prototype. In a Kerr medium the refractive index is intensity dependent, increasing (or decreasing) in proportion to the local optical energy density. The wave equation in a Kerr medium is thus nonlinear - of cubic order - in the field amplitude. In principle all materials exhibit a Kerr effect, though it is generally too weak to be of practical use. Many materials exhibit a Kerr-like effect mediated by an electronic or molecular excitation: if the excitation is long-lived, a relatively weak light field can generate a large excitation, and thus induce a substantial index change. As a consequence, such systems exhibit a large effective nonlinearity, and have been extensively investigated for a number of effects, including pattern formation. Among these materials are semiconductors and photorefractive media as well as liquid crystals.

In this article the Kerr approximation will be assumed adequate to describe the main features of spontaneous optical patterns in liquid crystal systems. Detailed comparison of theory with experiment naturally requires a more precise model of the nonlinearity and its dynamics: other articles in this volume successfully accomplish such comparisons.

An alternative and very effective approach is to *synthesise* a Kerr-like nonlinearity by a transducer which measures the optical intensity and induces a proportional refractive index change. This is strikingly successful in the case of *liquid crystal light valves* (LCLV). This concept was pioneered in Moscow by Akhmanov and Vorontsov[1] and has recently been used to demonstrate several important phenomena.

We will concentrate on systems where the Kerr medium "sees itself" in a single mirror[2]. This system has recently been used for a number of successful experiments, and turns out to be functionally closely analogous to the LCLV configuration just mentioned.

Other systems and configurations are of interest also in this field of spontaneous optical patterns, and a brief review and reference list is given. A survey of work through 1989 describing transverse effects in lasers and nonlinear optics has been published as foreword to a collection of papers in the field by Abraham and Firth[3], while articles by Lugiato[4] and Weiss[5] review more recent work and provide further references. Several articles in this volume provide evidence of the exciting progress in observations of spontaneous optical patterns in liquid crystal systems.

All these phenomena are counterparts of similar behaviour previously known in nonlinear systems in other branches of science. This introduces the concept of *universality* in the behaviour of nonlinear systems. Universality is not only interesting, but fundamental in nonlinear science where exact models and understanding are all but unknown. It is therefore a great help when ostensibly very different objects give rise to the same categories of behaviour. Thus in the case of spontaneous optical patterns hexagon formation shares many characteristics in optics and thermal convection.

INDUCED AND SPONTANEOUS PATTERNS

How is an optical pattern created? A *kaleidoscope* is an example of a *linear* pattern generator, the apparent complexity of its output images being simply a projection of the arrangement of its parts and the external illumination thereof. The much richer alternative, in optics as in biology, is to endow a simple system with an appropriate combination of free energy and nonlinearity, and allow it to *organise itself* into a complex structure. This requires a *nonlinear* interaction between the light and the matter. As we will see, the emergence of complex patterns and dynamics does not demand that either the properties or the driving of the system be complicated.

Patterns can emerge *spontaneously* only in nonlinear systems: such patterns involve *symmetry breaking* - for example, a regular lattice of bright spots has only discrete translational symmetry, so its emergence from a homogeneous state involves a breaking of that state's continuous translational symmetry.

Each symmetry breaking involves a choice among two or more equivalent broken-symmetry states: the *set* of such states retains the underlying symmetry, but the system chooses only one. Coexistence of two (or many) states consistent with the *same* input environment is the key distinction between linear and nonlinear optical patterns. It also offers a possible route to applications, since coexistent states can form the basis of all-optical memory devices.

KERR SLICE WITH FEEDBACK MIRROR

This section reports very complex space-time behaviour in a rather simple optical system, as described in three recent papers[6,7,8] following a modulational stability analysis by the author[2]. Hexagon formation is prevalent close to threshold (Fig. 1). Several recent experiments on this kind of system have revealed pattern formation, often with hexagonal structures. In addition, the system is functionally closely related to a feedback system in which a liquid crystal light valve is used to synthesise a Kerr nonlinearity[1].

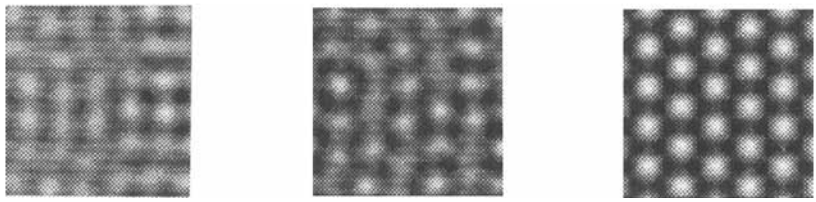

FIGURE 1 Spontaneous formation of a regular hexagon pattern in a defocusing Kerr slice with plane feedback mirror[7]. Time increases from left to right, and the feedback field intensity is plotted as a positive image.

The basic model, as stated above, is very simple, namely a thin slice of Kerr medium irradiated from one side by a spatially smooth beam - indeed a plane wave for the moment - with a mirror behind to generate a counterpropagating feed-back beam in the Kerr slice (Fig. 2).

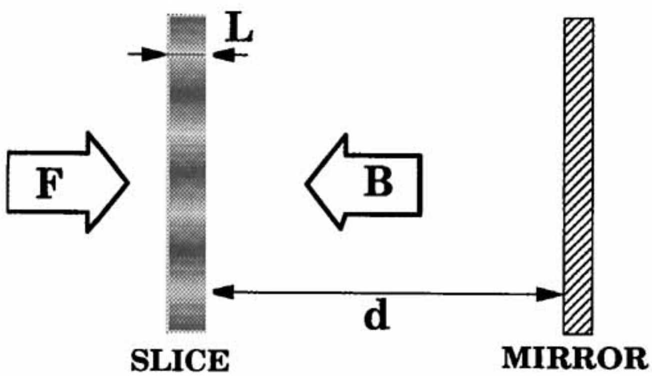

FIGURE 2 Schematic set-up of Kerr slice with feedback mirror. The forward amplitude *F* is smooth on input, but in pattern formation acquires a phase modulation through interaction in the Kerr slice with the feedback field *B*. *B* is formed from *F* by diffractive propagation to the mirror and back[7]. See also Eq. (1,2).

We describe the system as comprising a thin slice of Kerr-like medium in which the equation obeyed by the refractive index change Δn is:

$$(-l_D^2\nabla^2 + \tau\partial_t + 1)\,\Delta n(\mathbf{r},t) = |F|^2 + |B|^2 \tag{1}$$

Here *F* and *B* are respectively the amplitudes of the forward and backward travelling fields in the slice, whose intensities act as a source for the index change Δn. We neglect diffraction within the slice, so that these fields are simply phase modulated in traversing the slice, by an amount proportional to Δn. The Kerr excitation itself is assumed to relax with time constant τ and to diffuse transversely with diffusion length l_D. The Laplacian is transverse to the optical propagation.

In free space, the amplitude E of a field propagating in the +z direction obeys:

$$\frac{\partial E}{\partial z} = \frac{i}{2k}\nabla^2 E \tag{2}$$

Integrating this equation from slice to mirror and back determines *B* given *F* on exit from the slice. The latter is the input amplitude F_{in} phase modulated in proportion to Δn.

Eq. (1,2) thus define the system of Figure 2, and are the basis of both analysis and simulations[2,6-8]. Note that the nonlinearity and diffraction are separated between these two equations, which is what makes them particularly simple to analyse and simulate.

Obviously these equations have a trivial steady-state plane wave solution. Modulational stability analysis shows that this solution becomes unstable for nonlinear phase shifts of around half a radian, for a feedback mirror of reasonably high relectivity[2]. In particular, integrating this system for a defocusing medium gives rise to the hexagon formation shown in Fig. 1. Detailed analysis and extensive simulations of this system were reported by D'Alessandro and Firth[7].

Spontaneous spatial structures do not usually arise in defocusing media. They do arise here, not because of the nonlinear medium itself but due to the effects of free-space propagation, particularly clearly illustrated in this feedback mirror model.

What happens is that a smooth beam traversing a slice of Kerr medium gets no spatial structure of any kind from self-action, but can be *phase* modulated by a counterpropagating beam if that beam is *amplitude* modulated. Indeed only then, because the Kerr effect is insensitive to the phases of the fields involved. A closed positive feedback loop thus requires that the phase modulated input field returns from the mirror as an amplitude modulated beam. This interconversion of phase and amplitude modulation is precisely what diffractive propagation does, as has been known for well over a century since Talbot showed that a spatially modulated beam will self-image due to diffraction, after a distance now called the *Talbot length*. This length T is equal to $2\Lambda^2/\lambda$ where Λ is the pattern period and λ the optical wavelength.

More relevant here is that a beam which is amplitude modulated at Λ becomes phase modulated after a distance $T/4$ and vice versa, i.e after a relative phase shift of $\pi/2$ as compared to the 2π of the full Talbot length. Accordingly in the feedback mirror system one could expect the wave vector of the spontaneous pattern to be that which makes the mirror distance just $T/8$. Indeed this is the case, both in the theory and the recent experiments by Tamburrini et al[9] employing a liquid crystal as Kerr-like medium.

The critical roundtrip distance is $T/4$ only where the Kerr effect is focusing. For a defocusing medium, the phase modulation is reversed, and thus one might expect that three-quarters of the Talbot length would be the optimum roundtrip. Indeed this is the case in both theory and experiment. The Rome/Naples group[9] were able to access both signs of nonlinearity by putting the mirror at a *negative* distance. This was achieved by interposing a lens between slice and mirror, which could be translated so as to project the image on either side of the slice. For a collimated input beam it is only the product of Kerr phase shift and diffraction distance which matters, and so a positive nonlinearity at a negative distance behaves just like a negative nonlinearity at positive distance.

Using perturbation techniques from applied mathematics[10], nonlinear analysis of the Kerr slice with feedback mirror yields predictions, including hexagon formation, in reasonable ageement with simulations in the near-threshold regime[7]. In particular, hexagons are favoured in the presence of a *quadratic* nonlinear coupling between unstable wave vectors. Perturbation techniques cannot be relied upon for strong excitation, however, and both quantitative and qualitative deviations from the theory develop. The hexagons break up and what appears to be optical turbulence ensues[7].

As well as the experiments already mentioned, Macdonald and Eichler demonstrated spot patterns using a liquid crystal cell with feedback mirror[11]. These patterns showed the Talbot scaling described above, but no unambiguous evidence for hexagonal structures. Honda reported hexagonal patterns in the far field of a photorefractive crystal with reflective feedback[12]. Grynberg (private communication) has observed hexagonal and other structures in a rubidium vapour cell with feedback mirror: one extra complication is that the vector nature of the field seems to play a strong role, perhaps not surprisingly in view of the multilevel nature of alkali vapours.

GAUSSIAN BEAM EXCITATION

A further complication, shared by most experiments, is that the input field had a gaussian-beam, rather than a plane-wave, distribution. This makes comparisons with theory and simulations less direct than one would like, and so it is desirable to try to analyse gaussian beam problems.

Finite beam effects are serious for the analyst, because there is no longer an exact solution whose modulational stability can be tested. It is for just this reason that most analysis concentrates on plane wave excitation, trusting that real beams, if broad enough, will behave similarly.

One way forward is to concentrate on features of the beam which are qualitatively distinct from the plane wave, such as its *symmetry*. In particular, while the extensive suite of translational and rotational symmetries possessed by the field equations are preserved under plane wave excitation, the *translational* symmetry is lost for gaussian beam input.

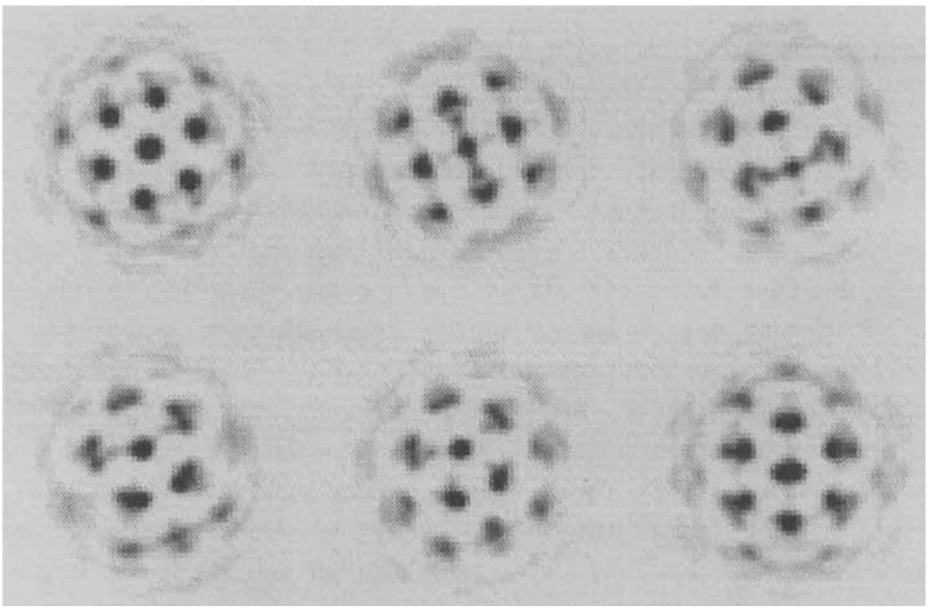

FIGURE 3 A set of negative images showing chaotic itinerancy in the Kerr slice with feedback mirror for the case of gaussian beam input. The six frames (read from left to right) show an approximate sixfold symmetry evolving through stages of near fourfold, twofold, onefold and threefold symmetries before returning close to, but not quite, its original state. *(Courtesy F Papoff).*

Applying techniques of symmetry analysis, Papoff *et al* were able to categorise the possible bifurcations through which the surviving rotational symmetry could be broken[8]. Not only were polygonal patterns predicted, including some - such as pentagons - which are forbidden in infinitely extended systems, but statements could be made about the *secondary bifurcations* through which such polygons lose stability. These include the onset of bodily rotation of polygons (observed in the experiments of Giusfredi *et al*[13]), as well as chaotic itinerancy, in which there is an erratic succession of imperfect polygons. These analytic results were in good accord with the simulations reported in the same work[8], of which Figure 3 is a sample.

LIQUID CRYSTAL LIGHT VALVE SYSTEMS

In the Kerr slice system of Figure 2, it will be observed that the only effect of the feedback beam is to modulate the phase of the input beam - there is no energy exchange between the input and feedback beams. It follows that one could instead *synthesise* a Kerr effect through any kind of transducer which detects the intensity of the feedback beam and encodes the phase of the input beam in proportion. Spatial phase modulators in general, and liquid crystal light valves in particular, perform just this function. There is thus a close functional similarity between LCLV and Kerr slice systems. In particular one should expect hexagonal patterns for the case where the long range coupling in the feedback loop is entirely due to diffraction. A recent experiment of this kind by the Darmstadt group[14] shows a very large area pattern with a very strong hexagonal coordination over a substantial central domain.

The LCLV system also points the way to a rich generalisation of pattern formation, since diffraction is by no means the only means of inducing long-range transverse coupling in the feedback loop. Aperturing and spatial filtering are others, but the most dramatic effects are achieved with a twist of the feedback through a fixed angle. The Moscow group have observed strikingly complex and beautiful patterns, with domain structures, spiral waves, and even apparently turbulent character[1,15]. A Florence-Moscow collaboration has used a 180° twist to demonstrate competition between hexagons and stripes in the LCLV system[16].

Linking the "all-optical" Kerr slice systems to LCLV experiments is important not only in stressing the universality of many pattern-forming processes, but also suggestive of routes to application. The Moscow group have already demonstrated optical logic and adaptive optics in the LCLV system, and there are clear links also to neural networks[17]. Some of these ideas may be workable also in faster optical materials such as semiconductors.

TRANSVERSE PATTERNS IN OTHER SYSTEMS

For a saturable atomic medium enclosed in a cavity and irradiated by a gaussian beam early simulations suggested that the cylindrical symmetry spontaneously breaks with the formation of bright "spots"[18]. More recently, studies of a simplified "mean field limit" of a nonlinear cavity system with transverse effects[19] showed qualitatively similar behaviour, in that the homogeneous solution may be modulationally unstable against pattern formation. In two transverse dimensions, however, analysis and simulations indicate[20] that *hexagonal* patterns are preferred.

Grynberg and co-workers made seminal experimental observations of hexagonal patterns in the far field of laser beams counterpropagating in a sodium vapour cell with no mirror or other external feedback[21]. Hexagons have been observed in the near field in more recent experiments by the Grynberg group using rubidium vapour[22]. This system used *cw* excitation, and was thus able to categorise the pattern formation as a function of optical tuning around the resonance lines of rubidium.

The dominant theme of this article so far has been *passive* nonlinear optical systems, systems which are externally driven by a coherent optical field (usually from a laser), and which exploit passive nonlinearities such as the Kerr effect. Many of the methods and techniques can, however, be carried over to *active* media such as lasers, and the review by Weiss[5] can be consulted for that purpose.

It is useful to make a few remarks on the similarities and differences between active and passive systems. Perhaps the key difference is that hexagon formation is generally absent in lasers. This can be traced to the fact that a free-running laser has an arbitrary phase. This symmetry disallows the quadratic coupling responsible for hexagons, and means that laser patterns are dominated by the cubic couplings.

The arbitrariness in the phase of the laser field implies the possible existence of *topological defects* in the field pattern. The phase may have a singularity, increasing or decreasing by a multiple of 2π around a point. Such points are termed *optical vortices* by Coullet *et al*[23].

An idealised broad-area laser exhibits, on the positive detuning side, *exact* solutions in the form of transverse travelling waves[24]. These can be viewed as a tilting of the optical wave vector to fit the cavity. Indeed the lowest threshold mode occurs exactly at the wavelength of peak gain.

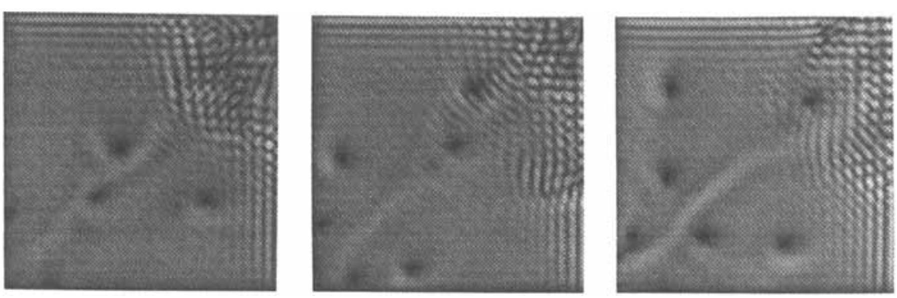

FIGURE 4 Transverse travelling wave structures in the output intensity of a wide-aperture laser. Vortices are created at the bottom left corner and travel diagonally with the underlying wave. Localised standing wave patterns are formed where the wave meets the (reflecting) boundary. *(Courtesy G K Harkness)*.

The pure travelling wave solutions are of uniform amplitude. They thus give rise to patterns in the intensity only where they become unstable, or where they have defects, such as must occur at a boundary. Figure 4 shows a simulation for the case of reflecting boundary conditions on a square section. Since all real lasers have finite transverse extent, the effect of boundaries on these modes, and by extension the manner in which they connect with the conventioNal transverse modes of curved-mirror resonators, is an interesting and important question. The development of broad area and surface-emitting semiconductor diode lasers in search of high power makes these studies technologically important.

CONCLUSION

In this paper I have tried to show that spatial patterns are interesting, complex and widespread in optics, and that liquid crystals have been prominent in developing our experience and understanding of these patterns.

On the experimental side the main problem is the need for adequate power, preferably continuous-wave, over relatively large areas. This indicates a use of strong, and therefore usually slow, nonlinearities like those in liquid crystals. On the theoretical side, the problem is one of a lack of analytical tools, leading inevitably to numerical simulations, which in this field are extremely resource-hungry.

These problems also represent opportunities. At the fundamental level experiments and theory on spatial patterns in optics may lead to new insights into the difficult and topical problems of morphogenesis, spatio-temporal chaos and turbulence. At the device level, the trend to increasingly complex optical systems such as diode laser arrays leads to problems of instability and spatial complexity which demand, and will respond to, the same kind of techniques as those fundamental investigations. Lastly, spatial complexity is a necessary feature of information processing systems, especially in configurations such as neural networks which require a certain plasticity of design or operation. Studies of spontaneous pattern formation in optics thus seem capable of leading to significant and important applications.

ACKNOWLEDGMENTS

The support and hospitality of the organisers of the conference were greatly appreciated. Technical assistance from G D'Alessandro, G K Harkness, G-L Oppo and F Papoff is gratefully acknowledged. This work is supported in part by SERC grants GR/F 49811, GR/F 76502 and GR/G 12665, and in part through ESPRIT BRA 7118 (TONICS).

REFERENCES

1. S A Akhmanov, M A Vorontsov and V Y Ivanov, JETP Lett. **47**,707 1988).
2. W J Firth, J. Mod. Opt. **37**, 151-154 (1990)
3. N B Abraham and W J Firth, J. Opt. Soc. Am. **B7**,951 (1990).
4. L A Lugiato, Physics Reports **219**, 293-310 (1992).
5. C O Weiss, Physics Reports **219**, 311-338 (1992).
6. G D'Alessandro and W J Firth, Phys. Rev. Lett. **66**,1597-1600 (1991).
7. G D'Alessandro and W J Firth, Phys. Rev. **A46**,537-548 (1992).
8. F Papoff, G D'Alessandro, G L Oppo and W J Firth, Phys. Rev. **A48**, 634-641 (1993).
9. M Tamburrini, M Bonavita, S Wabnitz and E Santamato, Opt. Lett. **18**, 855-857 (1993). E Ciamramella, M Tamburrini amd E Santamato, *submitted to* Appl. Phys. Lett. (1993).
10. A C Newell and J V Moloney, Nonlinear Optics, (Wiley, New York, 1992).
11. R Macdonald and H J Eichler, Opt. Commun. **89**, 289-294 (1992).
12. T Honda, Opt. Lett. **18**, 598-600 (1993).
13. G Giusfredi, J F Valley, R Pon, G Khitrova and H M Gibbs, J. Opt. Soc. Am. **B5**, 1181 (1988).
14. B Thüring, R Neubecker and T Tschudi, Opt. Commun. **102**, 111-115 (1993).

15. S A Akhmanov, M A Vorontsov, V Y Ivanov, A V Larichev and N I Zheleznykh, J. Opt. Soc. Am. **B9**, 78-90 (1992).
16. E Pampaloni, S Residori and F T Arecchi, (preprint, August 1993).
17. D Psaltis, D Brady and K Wagner, Appl. Opt. **27**, 1752 (1988).
18. J V Moloney, H Adachihara, D W McLaughlin and A C Newell, Chaos, Noise and Fractals, E R Pike and L A Lugiato (Ed.), (Hilger, London, 1987) p. 137.
19. L A Lugiato and R Lefever, Phys. Rev. Lett. **59**, 2208-2211 (1987).
20. W J Firth, A J Scroggie, G S McDonald and L A Lugiato, Phys. Rev. **A46**, R3609-R3612 (1992).
21. G Grynberg, E LeBihan, P Verkerk, P Simoneau, J R R Leite, D Bloch, S LeBoiteux and M Ducloy, Opt. Commun. **67**, 363-368 (1988).
22. A Petrossian, M Pinard, A Maitre, J-Y Courtois and G Grynberg, Europhys. Lett. **18**, 689-694 (1992).
23. P Coullet, L Gil and F Rocca, Opt. Commun. **73**, 403-408 (1989).
24. P K Jakobsen, J V Moloney, A C Newell and R Indik, Phys. Rev. **A45**, 8129-8137 (1992).