Chaos in photorefractive four-wave mixing with a single grating and a single interaction region

Wiesław Królikowski*

Electrooptics Technology Center, Tufts University, Halligan Hall, Medford, Massachusetts 02155

Milivoj R. Belić

Institute of Physics, P.O. Box 57, 11001 Belgrade, Yugoslavia

Mark Cronin-Golomb

Electrooptics Technology Center, Tufts University, Halligan Hall, Medford, Massachusetts 02155

Aleksander Błędowski*

Fachbereich Physik, Universität, PF 4469, D-4500 Osnabruck, Federal Republic of Germany

Received June 26, 1989; accepted February 9, 1990

We show that the standard model of four-wave mixing in a photorefractive crystal predicts the appearance of deterministic chaos. In this model there is a single (transmission) grating and no external or internal (intracavity) feedback. The intensity of the phase-conjugate wave is found to exhibit a period-doubling route to chaos on variation of the intensity of the probe beam and the linear absorption coefficient. The crucial elements in obtaining chaotic behavior are operation above the threshold for self-oscillation and the presence of an external electric field, which causes a shift in the optical frequency of the phase-conjugate wave.

The importance and potential applications of optical phase conjugation today seem to be universally accepted.[1] Nonetheless, before a reliable high-gain device can be made and put to use, possible regions of its unstable operation should be explored. Such regions seem to abound, for example, in barium titanate ($BaTiO_3$), one of the most interesting photorefractive crystals available.[2] At the same time, regions of instabilities with possible transitions to chaos in any novel nonlinear system present an interesting subject for investigation. With this in mind, in this paper we set out to investigate instabilities and the transitions to chaos in single-grating optical phase conjugation in a photorefractive crystal. Such a simple geometry seems to be especially interesting in view of the recent developments in the field.[2–4]

The initial observations of deterministic chaos in $BaTiO_3$ phase-conjugate mirrors without external feedback involved a self-pumped geometry of multiple interaction regions and multiple gratings,[3,4] which is easy to realize experimentally but sufficiently complicated to obscure the real mechanisms for chaotic behavior. Further, the occurrence of chaos has been demonstrated in other resonator geometries, in which one mirror is a phase-conjugating element. In such geometries the presence of all necessary ingredients for dissipative chaos is easily ensured: Nonlinearity arises from the phase-conjugate mirror, feedback is contained in the cavity, and driving is provided by the pumps. Indeed, a wealth of chaotic phenomena has been observed in phase-conjugate resonators.[5]

In this paper, however, we will display the emergence of chaos in a model for an externally pumped phase-conjugate mirror that is formed by single-grating four-wave mixing (4WM) with a single interaction region. No other feedback mechanisms, such as internal corner reflection (cat mirror[6]) and external mirrors, are used in our model. We show that even ordinary 4WM, with optical feedback provided only by the energy and phase transfer between the waves, is sufficiently nonlinear to produce unstable phase-conjugate output. In previous studies[7] on instability in externally pumped phase conjugation and in wave mixing in Kerr-like media, it was shown that the relation between the material response time and the transit time of light in the nonlinear medium was important for the instabilities observed (the instabilities were the largest when these times were comparable). We note that, because of the considerable slowness of the photorefractive effect, the light transit time plays no appreciable role here. The results reported here are obtained by numerical integration of coupled wave equations (in the slowly varying envelope approximation), augmented by a time-dependent equation for grating formation.

We consider 4WM in photorefractive crystals, which is produced by a transmission grating in the regime of orthogonally polarized pumps.[8] The geometry that we assume is shown in Fig. 1. The photorefractive crystal is illuminated by two counterpropagating pump waves, A_1 and A_2 (orthogonally polarized), and by probe wave A_4 (polarization the same as that of A_1). The generated phase-conjugate wave (PCW) A_3 propagates in a direction opposite that of the probe, and these two waves are orthogonally polarized. We assume that an external electric field is applied across the crystal (through high voltage, V in Fig. 1). In our studies of the dynamics of this system we will start with the probe beam A_4 equal to zero and then

0740-3224/90/071204-06$02.00

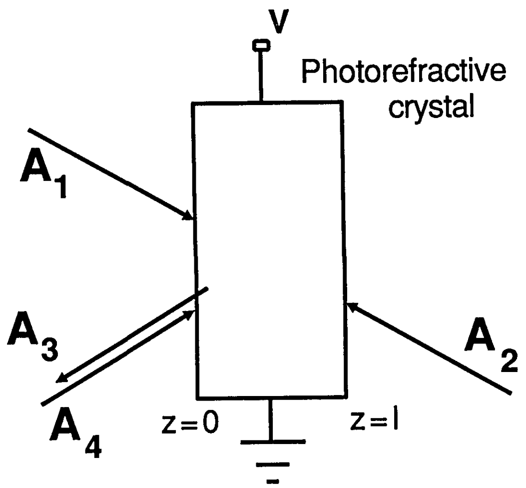

Fig. 1. Four-wave mixing geometry considered in the text. The pump waves A_1 and A_2 are orthogonally polarized, A_4 is the probe, and A_3 is the phase-conjugate output. V is the high-voltage source of the electric field.

increase its amplitude in order to examine its effect on the phase-conjugate output. The equations to be solved are of the form

$$\partial A_1/\partial z = QA_4 - \alpha A_1, \tag{1a}$$

$$\partial A_4^*/\partial z = -QA_1^* - \alpha A_4^*, \tag{1b}$$

$$\partial A_2^*/\partial z = -QA_3^* + \alpha A_2^*, \tag{1c}$$

$$\partial A_3/\partial z = QA_2 + \alpha A_3, \tag{1d}$$

$$\tau\partial Q/\partial t + \frac{E_D + E_q + iE_0}{E_M + E_D + iE_0}Q = \frac{\gamma_0}{I_0}\frac{E_q + E_D}{E_D}\frac{E_D + iE_0}{E_M + E_D + iE_0} \times (A_1A_4^* + A_2^*A_3), \tag{1e}$$

where Q is the complex amplitude of the index grating (with the corresponding relaxation time τ), $I_0 = \Sigma|A_j|^2$ is the total light intensity, E_D, E_q, and E_M are the characteristic electric fields describing the crystal (according to the theory of Kukhtarev et al.[9]), E_0 is an external electric field, α is the linear absorption coefficient, and γ_0 is the coupling constant, which depends on the material parameters (electro-optics coefficients) and geometrical factors.

Equations (1) are derived from Maxwell's equations accompanied by the theory of the photorefractive process. An exhaustive discussion of these equations as well as the role of all material parameters can be found in Ref. 10. Here we note only the specific role of the electric field, which appears in Eq. (1e) in a complex (imaginary) parameter (E_0 itself is real). When $E_0 = 0$, the phase shift between the index grating Q and the interference pattern $(A_1A_4^* + A_2^*A_3)$ is exactly $\pi/2$. The phases of waves do not change in the interaction, but energy is transferred between the waves. When the electric field E_0 is present, the photorefractive phase shift is no longer equal to $\pi/2$, which results in the change of the phases of the waves during interaction (phase transfer). This effect has a profound consequence in the energy transfer process, which becomes strongest when the waves have different optical frequencies.[11] When the waves have different optical frequencies, the interference pattern moves in the crystal. The extra phase shift induced by the time lag between the grating and the interference pattern can restore the $\pi/2$ phase shift and maximal energy transfer.

In Eqs. (1a)–(1d), time derivatives have been neglected since propagation delay is short compared with the time needed for grating formation. One can regard the wave amplitudes as following the grating evolution adiabatically. The procedure for solving Eqs. (1) is as follows. Owing to the form of the equations, temporal and spatial integrations can be separated. Since the crystal response is slow, one can divide the marching variable (time) into small intervals t_N in which the 4WM process may be considered as diffraction of waves by the quasi-stationary index grating $Q(t_N, z)$. This effect is governed by Eqs. (1a)–(1d), which actually break into two sets of coupled linear equations for amplitudes A_1 and A_4 [Eqs. (1a) and (1b)] and A_2 and A_3 [Eqs. (1c) and (1d)]. These sets of equations may be easily solved numerically (by using, for instance, the Runge–Kutta method) with the following boundary conditions: $A_1(t_N, z = 0) \equiv A_{10}$, $A_2(t_N, z = l) \equiv A_{2l}$, $A_3(t_N, z = l) \equiv 0$, and $A_4(t_N, z = 0) \equiv A_{40}$, where A_{10}, A_{2l}, and A_{40} are the amplitudes of the fields that are incident upon the crystal (in the planes $z = 0$ and $z = l$). Energy exchange between the waves leads to a change in the wave interference term $A_1A_4^* + A_3A_2^*$, which in turn modifies the index grating Q through Eq. (1e). This new grating in the next time interval t_{N+1} causes further energy exchange, a new interference pattern, and so on.[10] In numerical calculations the value of the time increment $\Delta t = (t_{N+1} - t_N)$ must be much smaller than the characteristic time scale of the photorefractive process, which is given by $(E_M + E_D + iE_0)/(E_D + E_q + iE_0)\tau$. We performed integration while decreasing time step Δt until the difference between results was less than the required accuracy.

It is well known from the steady-state theory of 4WM that a PCW may be generated even without a probe beam.[12] This effect, known as self-oscillation, occurs only when the nonlinear coupling is sufficiently large. In our numerical simulations we always assume that the coupling constant exceeds the threshold for self-oscillation, i.e., the coupling strength $\gamma_0 l$ (l is an interaction length) without E_0 is chosen to be -4, while the self-oscillation threshold is -2.[13] If there were no electric field applied to the crystal, the generated conjugate wave would have the same optical frequency as that of the pumps. The presence of the external electric field qualitatively changes the mixing process. First, it increases the magnitude of the coupling constant. Second, this field induces an additional phase shift in the coupling constant, which results in a frequency shift of the generated PCW in the pure self-oscillation case ($A_{40} = 0$).[14] The value of this frequency shift is such that the full phase shift between the index grating and the interference pattern in the steady state is $\pi/2$. We are going to examine the effect of raising the probe intensity (I_{40}) above zero and will consider the case in which all input fields have the same frequency and all boundary values are independent of time. For the characteristic electric fields in the examples considered, we assume that $E_D = 1$ kV/cm, $E_q = 5$ kV/cm, and $E_M = 100$ kV/cm, which are consistent with the experimental values for $BaTiO_3$. The external electric field is taken to be $E_0 = 3$ kV/cm in one example and $E_0 = 2$ kV/cm in the other.

The results of numerical simulations reveal that for the case discussed here, i.e., operation above the self-oscillation threshold, the presence of the nonzero input probe beam leads to unstable behavior of the intensity of the conjugate wave. The ensuing dynamics depends strongly on the value of the parameters and may become quite complex. We show the instances of this behavior in Fig. 2, where we present the time dependence of the output intensity $I_3(t, z = 0)$ as well as the phase-space portrait of the PCW amplitude $A_3(t, z = 0)$. It should be noted that in the case of pure self-oscillation ($A_{40} = 0$) the phase-space portrait reduces to the circle that indicates the constant frequency shift of the conjugate wave that is caused by the electric field. Taking the extremes of the output intensity (after any transients have decayed) for each value of the control parameter, we obtain the bifurcation diagrams shown in Figs. 3–5. In Fig. 3 we present the bifurcation diagram obtained by variation of the probe intensity I_{40} (all intensities are normalized to total pump intensity $I_{10} + I_{2l}$). It is evident that the diagram does not form a single branch, even for very weak input. The reason is that for the parameters used the effective coupling strength is very large, $|\gamma l| = |\gamma_0 l[(E_q + E_D)/E_D][(E_D + iE_0)/(E_D + E_q + iE_0)]| \simeq 12$, and therefore even for very small probe intensities the output conjugate wave is unstable. However, integration of Eqs. (1) for the case of pure self-oscillation, e.g., when $A_{40} = 0$, gives a stable intensity for the PCW.

When the electric field is smaller, as in Fig. 4 ($E_0 =$ 2 kV/cm), self-oscillations are clearly seen to start as single branch or, rather, a single point and to give rise to a time-independent steady-state intensity. This steady state readily bifurcates as the (initial) intensity of the probe (I_{40}) is increased. However, at some even higher intensity, after chaos has been reached, the intensity of the conjugated signal remerges into a single branch. Such inverse bifurcations have already been observed in similar optical systems, such as optical bistable devices[15] and phase-conjugate resonators.[5] Remerging bifurcations are characteristic of low-dimensional systems with multidimensional parameter space and with symmetries in this space that leave the equations of motion invariant.[16] The

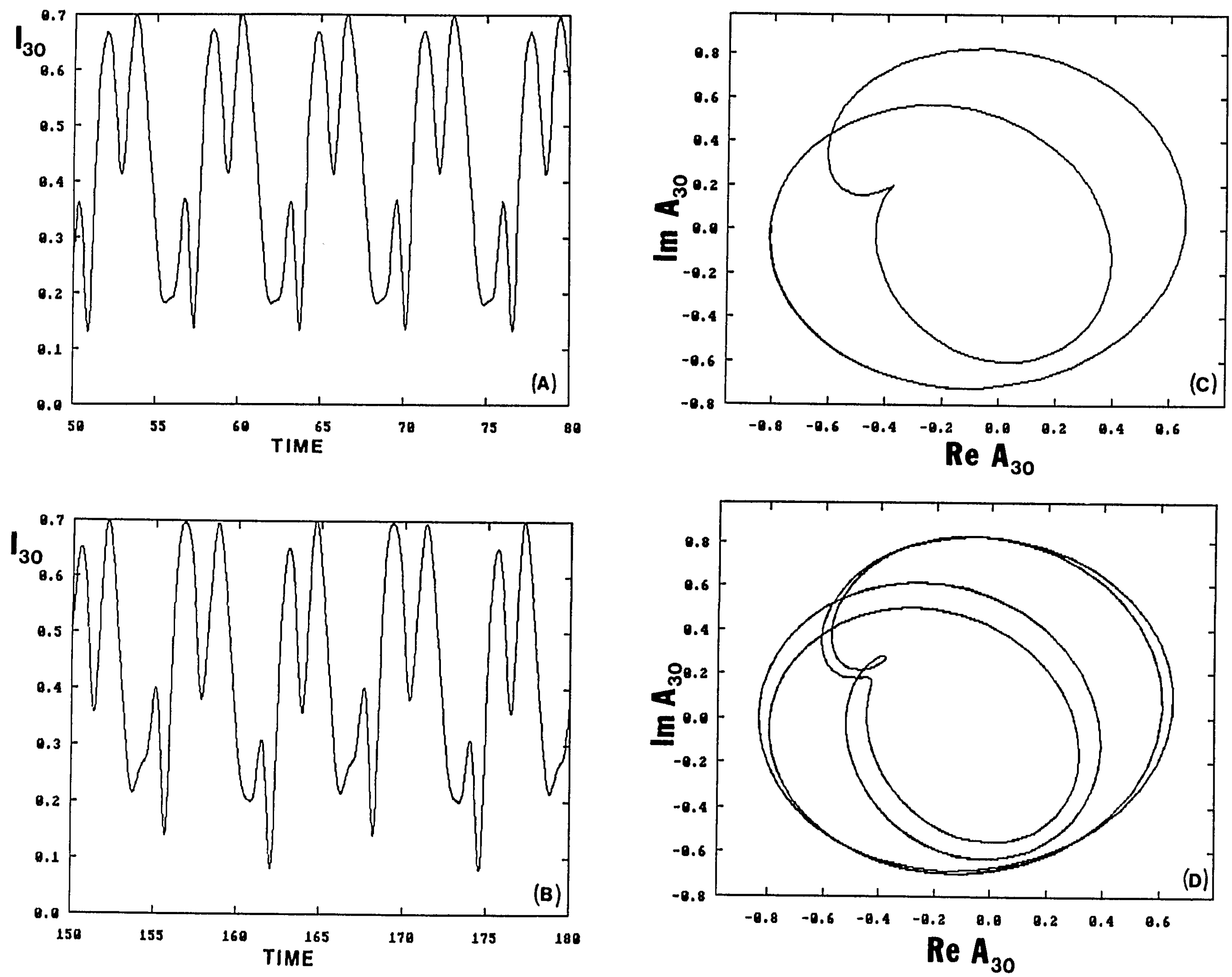

Fig. 2. (A), (B) Output intensity and (C), (D) amplitude of a conjugate wave in the unstable regime (regular pulsations): $E_0 = 3$ kV/cm, $I_{10} = 0.3$, $I_{2l} = 0.7$, $\alpha = 0$, $\gamma_0 l = -4$. Note the period doubling when the probe intensity is increased from (A), (C) $I_{40} = 0.015$ to (B), (D) $I_{40} = 0.03$.

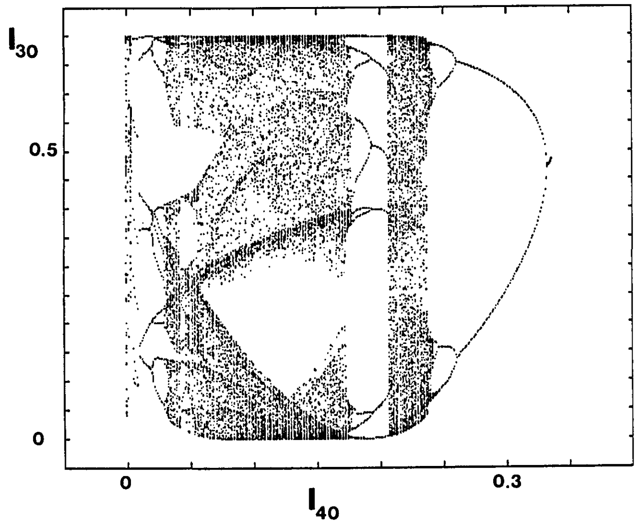

Fig. 3. Bifurcation diagram representing PCW intensity I_{30} as a function of probe intensity I_{40}. The parameters used for this diagram are $\gamma_0 l = -4$, $\alpha = 0$, $I_{10} = 0.3$, $I_{2l} = 0.7$ (intensities are normalized to total pump intensity).

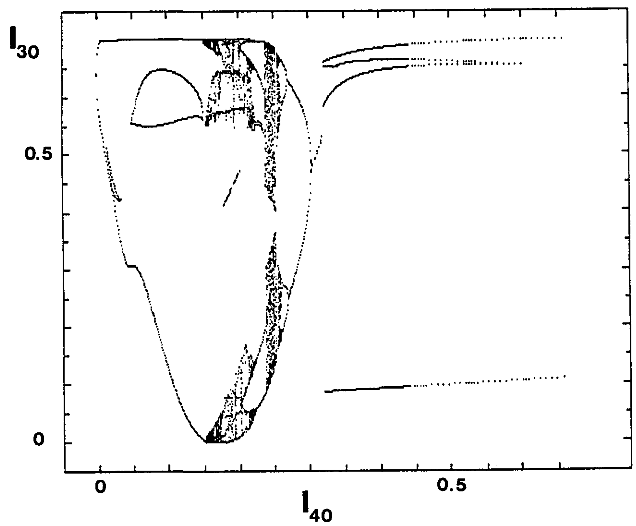

Fig. 4. Same as Fig. 3 except that the external electric field has been changed from $E_0 = 3$ kV/cm to $E_0 = 2$ kV/cm.

route to chaos is seen to be period doubling (direct or inverse) with periodic windows, which is characteristic of chaos in few dimensions. We have measured the fractal dimension of the attractor at $I_{40} = 0.13$ by using the correlation integral and embedding technique of Grassberger and Procaccia[17] to find $D \simeq 2.4$ (Fig. 6).

The origin of chaos in this simple geometry seems to be different from that in the geometries reported earlier,[3,4] for which the existence of multiple interaction regions or cavities formed by coated C faces of the electro-optic crystals seemed to be crucial. Further, those geometries enforced multiple-grating operation, which makes theoretical analysis difficult. In our example the strong coupling of waves, which creates a single transmission grating, is sufficient to cause self-oscillations (at a shifted optical frequency). This leads to the recording of a moving grat-

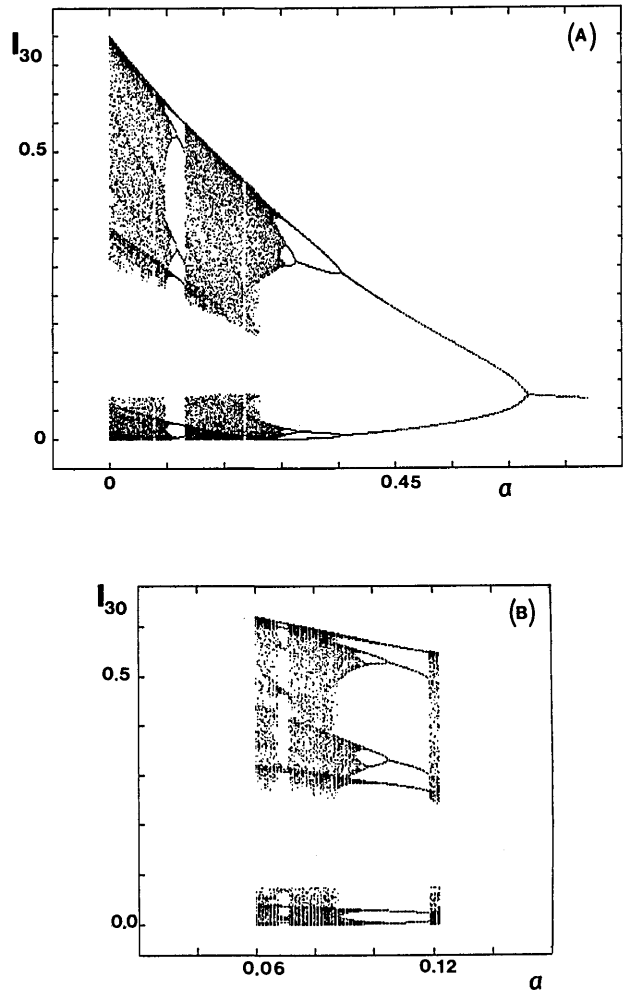

Fig. 5. Bifurcation diagram with the linear absorption coefficient as the control parameter ($\gamma_0 l = -4$, $I_{40} = 0.13$, $E_0 = 3$ kV/cm). (B) is an enlargement of the region of $0.06 \leq \alpha \leq 0.12$ from (A).

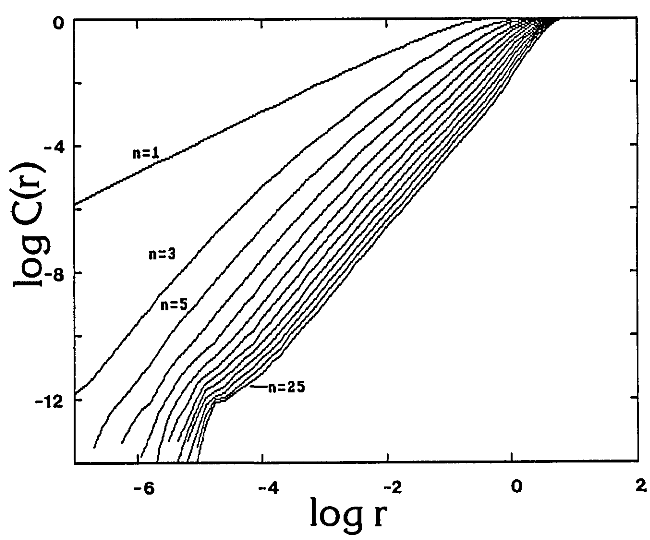

Fig. 6. Log–log plot of the correlation integral $C(r)$ versus distance r for a few embedding dimensions. The slopes of these curves in the range in which they are parallel should give the fractal dimension of the attractor.

ing in the crystal. On the other hand, the presence of the probe beam, which has a frequency equal to those of the pumps, gives rise to a stationary index grating. The PCW is always formed by the diffraction of the pump beam A_2 on the index grating. Since the contributions to the PCW that come from diffraction of the pump on different gratings (stationary and moving) have different frequencies, the output intensity is unstable. The situation is even more complicated: Each of the four waves presented in the crystal diffracts on the various gratings, which leads to the appearance of components with different optical frequencies and complex time behavior. These instabilities are then driven to chaos by changing some of the available control parameters, for example, the intensity of the probe beam, the external electric field, or even the linear absorption coefficient. In addition, in our case the presence of the external electric field plays a significant role. Our numerical simulations show that output is stable when $E_0 = 0$.[18] In general, an applied electric field increases the nonlinear coupling (through the coupling constant) and causes an additional phase shift, which leads to the formation of the moving gratings (running holograms) in the photorefractive crystal. Our system behaves as a driven pendulum does. When the driving beam is absent ($A_{40} = 0$), we have free (and stable) generation of the conjugate wave, which is degenerate in optical frequency when $E_0 = 0$ or has a shifted optical frequency when $E_0 \neq 0$. With driving ($A_0 \neq 0$) and an applied electric field, the system exhibits temporal pulsations and chaos.[19] This is qualitatively different from the situation reported by Gauthier *et al.*,[4] in which the multiple-grating model of 4WM (with real coupling constants) could not produce unstable output in single-grating regime.

From Figs. 3 and 4 it is evident that strong probe intensity suppresses instabilities of the PCW. This effect results from the nature of photorefractive coupling, which depends on the modulation of the interference pattern formed by interacting waves and not on their intensities. The presence of the probe beam A_4 changes the gain conditions for the oscillation with shifted frequency. The larger the intensity I_{40}, the smaller the modulation of the interference pattern; eventually nonlinear coupling becomes too weak to support oscillation with shifted optical frequency. The grating in the crystal, formed by waves with the same optical frequency, is stable, which leads to the stable intensity of the conjugate wave.

Finally, we present an interesting example of the bifurcation diagram in which variation of the linear absorption coefficient was the driving parameter[20] (Fig. 5). This diagram was generated in the same manner as Figs. 3 and 4, the only difference being the choice of the parameter to be varied. Figure 5 displays suppression of instabilities by linear absorption. This is a plausible behavior: Increased absorption means stronger dissipation, and strongly attenuated systems are usually more stable, with their dynamics tending to go to fixed points in the phase space. The phase-conjugate output, which is chaotic for zero absorption, is seen to remerge into a single branch with an increasing absorption coefficient. Between the cascades of inverse bifurcations, periodic windows open. One such instance is depicted in Fig. 5(B), which is an enlargement of the region of absorption $0.06 \leq \alpha \leq 0.12$, from Fig. 5(A). A period 6 attractor is seen to end up in an interior crisis, exploding into two chaotic bands. Thus, while increased absorption suppresses chaos globally, it may sometimes induce chaos locally.

In summary, we have demonstrated the appearance of chaos in a model of optical phase conjugation in a single 4WM interaction region. When a single transmission grating regime of operation is assumed, the phase-conjugate output has been driven to chaos by the variation of several different control parameters. No additional feedback mechanisms are provided other than the standard geometry of 4WM in a photorefractive medium. Low-dimensional chaos has been observed, with period-doubling routes to chaos. The crucial element in obtaining chaos is operation above the threshold for self-oscillation, i.e., strong enough coupling of the waves. Another important ingredient is the presence of an external electric field, which causes variable phase shift in the coupling constant and a shift of the optical frequency of the PCW. Once self-oscillation instabilities begin, the sequence and complexity of the bifurcation diagram that describes periodic or chaotic behavior strongly depend on which parameter is varied. If the probe intensity is varied, then we have competition of the oscillatory motions (with different frequencies), which results in complicated bifurcation diagrams. If the absorption coefficient is varied (increased), then we have gradual suppression of chaos by a series of inverse bifurcations. In both cases the PCW eventually reaches a single steady-state value.

Although we have discussed 4WM in the special case of orthogonally polarized pumps, it should be emphasized that the results presented here also apply in the conventional geometry with parallel polarized pumps. These two arrangements are completely equivalent as far as formal equations are concerned.[21] In the framework of the parallel polarized pumps, the process of inducing unstable behavior in 4WM would be a case of an externally driven, double phase-conjugate mirror. When the input probe wave has zero amplitude, we have exactly the case of the double phase-conjugate mirror (a state of stable self-oscillation).

ACKNOWLEDGMENTS

This study was supported by the National Science Foundation through a Presidential Young Investigator Award and by the U.S. Army Research Office, Durham, North Carolina.

*Permanent address, Institute of Physics, Polish Academy of Sciences, 02-668 Warsaw, Poland.

REFERENCES AND NOTES

1. See, for example, R. A. Fisher, ed., *Optical Phase Conjugation* (Academic, New York, 1983); P. Günter, Phys. Rep. 93, 99 (1982).
2. See G. C. Valley and Pochi Yeh, eds., J. Opt. Soc. Am. **5**, 1681–1821 (1988), and numerous references cited therein.
3. P. Günter, E. Voit, M. Z. Zha, and J. Albers, Opt. Commun. **55**, 210 (1985); A. M. C. Smout, R. W. Eason, and M. C. Gower, Opt. Commun. **59**, 77 (1986).
4. D. J. Gauthier, P. Narum, and R. W. Boyd, Phys. Rev. Lett. **58**, 1640 (1987).

5. G. Valley and G. Dunning, Opt. Lett. **9,** 420 (1984); G. Reiner, P. Meystre, and E. M. Wright, J. Opt. Soc. Am. B **4,** 865, (1987); G. Reiner, M. R. Belić, and P. Meystre, J. Opt. Soc. Am. B **5,** 1193 (1988).
6. J. Feinberg, Opt. Lett. **7,** 486 (1982).
7. C. Pare, M. Piche, and P. A. Belanger, J. Opt. Soc. Am. B **5,** 679 (1988); Y. Silberberg and I. Bar-Joseph, Phys. Rev. Lett. **48,** 1541 (1982).
8. S. I. Stepanov and M. P. Petrov, Opt. Commun. **53,** 64 (1985).
9. N. V. Kukhtarev, V. Markov, S. Odulov, M. Soskin, and V. Vinetskii, Ferroelectrics **22,** 949 (1979); N. V. Kukhtarev, V. Markov, and S. Odulov, Opt. Commun. **23,** 338 (1977).
10. L. Solymar and J. M. Heaton, Opt. Commun. **51,** 76 (1984); J. M. Heaton and L. Solymar, Opt. Acta **32,** 397 (1985).
11. Ph. Refregier, L. Solymar, H. Rajbenbach, and J. P. Huignard, J. Appl. Phys. **58,** 45 (1985).
12. B. Fischer, M. Cronin-Golomb, J. O. White, and A. Yariv, Opt. Lett. **6,** 519 (1981).
13. W. Królikowski, M. R. Belić, and A. Błędowski, Phys. Rev. A **31,** 2224 (1988).
14. B. Fischer, S. Sternklar, and S. Weiss, IEEE J. Quantum Electron. **25,** 550 (1989).
15. R. W. Boyd, R. M. Raymer, and L. M. Narducci, eds., *Optical Instabilities* (Cambridge U. Press, Cambridge, 1986); N. B. Abraham, L. A. Lugiato, and L. M. Narducci, eds., J. Opt. Soc. Am. B **2,** 1–264 (1985).
16. M. Bier and T. Bountis, Phys. Lett. A **104,** 239 (1984).
17. P. Grassberger and I. Procaccia, Phys. Rev. Lett. **50,** 346 (1983).
18. A. Bledowski and W. Królikowski, IEEE J. Quantum Electron. **24,** 652 (1988).
19. Although in the case discussed in this paper the external electric field is crucial for the dynamics of the system, sometimes a sufficiently strong nonlinear coupling in the absence of the electric field can cause unstable (but not chaotic) behavior of the photorefractive 4WM. See A. Błędowski, W. Królikowski, and A. Kujawski, J. Opt. Soc. Am. B **6,** 1544 (1989).
20. It has been shown recently that the absorption coefficient in photorefractive $BaTiO_3$ depends on total light intensity. See G. A. Brost, R. A. Motes, and J. R. Rotge, J. Opt. Soc. Am. B **5,** 1879 (1988).
21. K. Shaw and M. Cronin-Golomb, Opt. Commun. **65,** 301 (1988).