Lyapunov exponent analysis of irregular fluctuations in a self-pumped BaTiO₃ phase-conjugate mirror, establishing transition to chaotic behavior

Philip M. Jeffrey and Robert W. Eason

Department of Physics and Optoelectronics Research Centre, University of Southampton, Southampton SO9 5NH, UK

Received March 19, 1993; revised manuscript received July 21, 1993

The behavior of the phase-conjugate output from a self-pumped, BaTiO₃ phase-conjugate mirror is known to exhibit temporal instabilities under certain conditions. Approaches for investigating these experimentally observed fluctuations indicate the presence of chaotic behavior. However, a dynamic system that possesses one or more positive Lyapunov exponents is by definition chaotic. For the first time, to our knowledge, we calculate directly from experimental data the largest nonnegative Lyapunov exponent of these irregular fluctuations and report examples of definite transitions from stable to chaotic behavior.

INTRODUCTION

There has been continued interest in photorefractive crystals because of their potential suitability for optical communication devices and applications. Photorefractive barium titanate (BaTiO₃) is of particular interest because it provides a simple self-aligning phase-conjugate mirror. There are many different geometries for the operation of these phase-conjugate mirrors, such as the interaction of two mutually incoherent beams^{1,2} or, more simply, the introduction of a single beam to form a self-pumped phase conjugator.3 Recently reported increases in both the reflectivity4,5 and response time6 of self-pumped phase conjugation (SPPC) in BaTiO3 and the inherent simplicity of requiring only a single input beam makes these phase-conjugate mirrors particularly suitable for a range of optical communication devices and applications. However, all practical devices would require that the SPPC's properties exhibit both long-term spatial and temporal stability. Therefore it is desirable to investigate the nature of any such instability so that future systems can be designed to operate under optimum conditions.

Spatial instability in SPPC in BaTiO₃ has been previously investigated 7 and was shown to be due in certain cases to feedback from internal reflections from the crystal faces. Temporal instability has also been investigated and was seen to depend on factors such as the input beam position, angle, and incident laser intensity. $^{8-10}$ Power spectra observations 11 and attempts to estimate the lower bound of the Hausdorff (fractal) dimension of the temporal field 12 also reveal that the temporal instabilities are critically dependent on the input-beam position.

Kaplan and Yorke¹³ formulated an expression for the dimension of an underlying attractor in a system in terms of the Lyapunov exponents λ_i , which measure the exponential separation of nearby orbits in phase space. Measurement of the Lyapunov exponents (the largest exponent is λ_1 , but certain directions will produce smaller

exponents $\lambda_2, \lambda_3, \ldots$ with $\lambda_1 \geq \lambda_2 \geq \lambda_3 \geq \ldots$) permits quantities such as useful bounds on the dimension of an attractor to be derived. We have experimentally investigated the largest Lyapunov exponent (λ_1) for different input-beam positions in SPPC in BaTiO₃, in order to ascertain whether the increasingly unstable fluctuations in the phase-conjugate output become chaotic. Accurate calculation of the Hausdorff dimension¹³ requires knowledge of all but the most negative Lyapunov exponents, which is problematic for real systems. It has been shown that the following inequality holds^{14–19} if the points in phase space are uniformly distributed over the attractor:

$$D_2 \le D_1 \le D_0 \,, \tag{1}$$

where D_2 is the correlation dimension, D_1 , the information dimension, and D_0 , the Hausdorff dimension.

Investigations of irregular behavior in phase-conjugate resonators^{20,21} and four-wave mixing^{22,23} in BaTiO₃ have been reported to show chaotic behavior. However there are difficulties with the use of the correlation dimension to establish chaotic behavior in SPPC because the Hausdorff dimension of an attractor is independent of the frequency of visitation of the attractor, whereas the correlation exponent exhibits sensitive dependence on the rate of visitation.¹⁴ In SPPC in the unstable reflectivity regime the phase-conjugate intensity is often observed to be near zero, suggesting that some parts of the attractor are visited more often than others, and therefore the correlation dimension is an insufficient measure of the Hausdorff dimension.

Gauthier et al.²⁴ confirmed this by investigating SPPC in BaTiO₃ and reporting that the correlation dimension showed sensitive dependence on the crystal orientation. The correlation dimension was found to be effectively independent of the input intensity, suggesting that although the laser intensity determines the time scale of the chaotic evolution, geometrical factors are principally responsible for the trajectories in phase space.

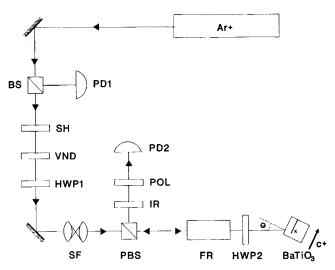


Fig. 1. Experimental setup used to investigate SPPC in BaTiO₃. BS, beam splitter; SH, shutter; VND, variable neutral-density filter; SF, spatial filter; PBS, polarizing beam splitter; IR, mechanical iris; POL, polarizer; PD1, PD2, photodiodes; FR, Faraday rotator; HWP1, HWP2, 514.5-nm half-wave plates.

In contrast to techniques that use the correlation dimension and other methods, ^{25,26} we have used the algorithm established by Wolf *et al.*^{27,28} and modified by Rand and Wilson^{29,30} to calculate directly the largest nonnegative Lyapunov exponent from an experimental time series. The advantage of this technique is that the algorithm is insensitive to the frequency of visitation of the attractor and is fairly parameter independent. Therefore this algorithm is better suited to analysis of experimental data from SPPC than are alternative methods for determining chaotic behavior.

Wolf²⁷ investigated other approaches to quantifying chaos from experimental data, such as fractal power spectra, entropy, and fractal dimension, all of which have been reported previously for SPPC in BaTiO₃. ^{11,12,24} Wolf comments that these approaches often fail to characterize chaotic data accurately and are susceptible to external noise and are sensitive to the amount of data being analyzed. The techniques also show strong parameter dependence, which can lead to large errors. However, the algorithm reported by Wolf³⁰ has been shown to provide an accurate calculation of the largest nonnegative Lyapunov exponent from experimentally produced data. The algorithm favorably shows only weak parameter dependence, with experimental results containing relatively small (5–10%) errors.

EXPERIMENTAL CONFIGURATION

The experimental configuration is shown in Fig. 1. The dimension of the $BaTiO_3$ crystal was 5 mm \times 5 mm \times 5 mm and had been electrically poled into a single ferroelectric domain before experimental investigation. The input beam was from a 5-W Ar^+ laser operating at 514.5 nm, e polarized with respect to the input crystal face. The beam was split by a 50:50 beam splitter so that the laser stability could be measured by a photodiode (PD1) connected to a Newport powermeter. The laser output was stable to <0.5%. The input-beam intensity could be varied by the variable neutral-density

filter, and the polarization was rotated to the horizontal by a half-wave plate (HWP1).

The spatially filtered beam, with beam diameter approximately 1.5 mm, was then transmitted by a high-extinction-ratio (10^4) polarizing beam splitter. The Faraday rotator (FR) rotated the plane of polarization of the beam by $+45^\circ$ before it was rotated by -45° (back to horizontal polarization) by a second half-wave plate (HWP2).

The crystal was mounted onto a rotation stage, permitting an accurate translation ($\sim 1 \mu m$) of the crystal horizontally (varying the beam position, x, along the crystal face), vertically, and rotation about the center of the crystal to vary the angle θ , between the input beam and the normal to the crystal face, as shown in Fig. 1. To minimize instabilities due to air currents and thermal effects, we mounted and thermally insulated the crystal within a black insulating container, with an entrance slit just large enough to permit the input beam to pass. The thermal stability of the crystal and the surrounding chamber was monitored with a thermocouple placed close to the crystal. The input beam was observed to fan toward the +c axis, and several self-pumped channels formed. The SPPC mechanism produced a phase-conjugate beam, which was redirected by the polarizing beam splitter after retraversal of the Faraday isolator. A mechanical iris blocked out any stray light before the phase conjugator was passed through a high-extinction polarizer aligned to transmit the vertically polarized phase conjugator, while filtering out any residual horizontally polarized light from stray reflections and scattering.

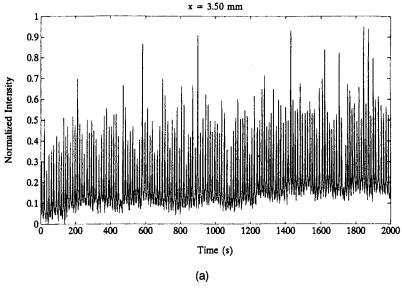
The phase-conjugate intensity was measured by a second photodiode (PD2), the output from which was amplified by a low-noise amplifier and fed into a computer-based data-acquisition system. Before any data were obtained by the computer, the crystal was illuminated by a 75-W white-light source for 5 min to ensure that all the gratings within the crystal were erased.

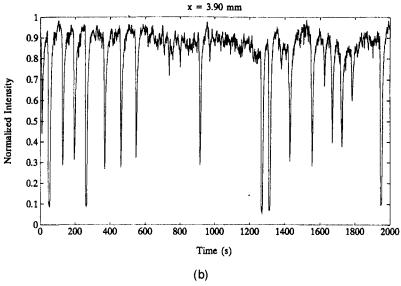
EXPERIMENTAL RESULTS

Experimental data of the phase-conjugate intensity were taken as a function of time (over periods of hours), as a function of input beam intensity (I_{in}) , horizontal beam position (x), and angle (θ). For $\theta < 40^{\circ}$ and for a narrow region across the crystal face, irregular fluctuations were observed in the phase-conjugate output (as shown in Fig. 2). As detailed elsewhere, 10 on either side of this narrow region the phase-conjugate output was stable. However, as the input-beam position was systematically tracked through this region, a definite transition from regular to irregular behavior and then back to regular behavior was observed before the phase-conjugate output finally became stable again. The experimental time series is first reconstructed into an n-dimensional phase space by the choice of a time delay τ (equal to one-quarter orbital period).31 The mean orbital period was defined by observing a dominant spectral feature in the power spectrum. The largest nonnegative Lyapunov exponent can then be calculated:

$$\lambda_1 = \frac{1}{t_M - t_0} \sum_{k=1}^{M} \log_2 \frac{L'(t_k)}{L(t_{k-1})},$$
 (2)







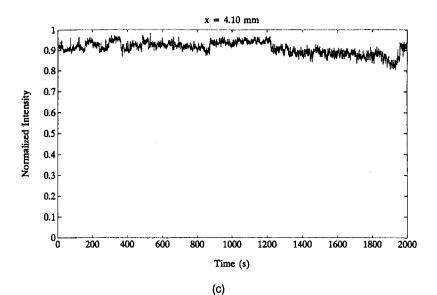


Fig. 2. Experimental results showing the variation of the phase-conjugate intensity with transverse position (x) of the input beam along the crystal face $(\theta = 2^{\circ})$. (a) x = 3.50 mm, (b) x = 3.90 mm, and (c) x = 4.10 mm.

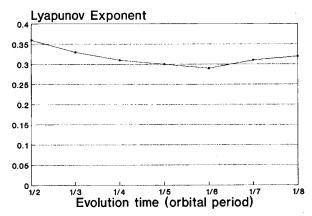


Fig. 3. Dependence of the largest nonnegative Lyapunov exponent (λ_1) as a function of the evolution time Δ for $\theta=2^{\circ}$, $I_{\rm in}=2.1~{\rm W~cm^{-2}}$, and $x=3.50~{\rm mm}$.

where M is the total number of replacement steps and the length $L(t_0)$ between the initial point of the fiducial trajectory and the nearest neighbor to the initial point is observed to have evolved to a length $L'(t_1)$ after time t_1 . The evolution time Δ , defined as

$$\Delta = t_{k+1} - t_k \,, \tag{3}$$

was held constant at one-quarter orbital period.

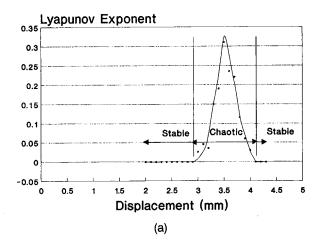
Figure 3 shows that the exponent calculation was found to be independent of the evolution time when we assumed that the orbital divergence was monitored at least a few times per orbit. Data were sampled at ~12 data points per mean orbital period, and data files contained 32,000 data points. Thus the reconstructed attractor in phase space contained $\sim 3,000$ orbits. The nature of the transition from stable phase-conjugate output to periodic behavior to irregular fluctuations and back again to stability was observed to be essentially the same for different input-beam intensities, position, and angle. Although varying the input-beam intensity changed the period of regular oscillations,8 the same transitional behavior was observed. Again, varying the input angle changed the position along the crystal where the transition to instability occurred but did not alter the transitional characteristic behavior of the phase-conjugate output. Results are presented in Fig. 4, which shows the largest nonnegative Lyapunov exponent (λ_1) obtained from experimental data of SPPC in BaTiO₃. The input-beam intensity, I_{in} = 2.1 W cm⁻², was kept constant for the three cases shown in Fig. 4. Figure 4(a) shows the results for $\theta = 2^{\circ}$. Data files were obtained at intervals of 0.1 mm along the crystal face. For x < 3.0 mm and x > 4.0 mm the Lyapunov exponent (λ_1) was observed to be zero (0.00 \pm 0.02), showing nonchaotic behavior corresponding to stable phaseconjugate output. For x = 3.5 mm a Lyapunov exponent of $\lambda_1 = 0.31$ was obtained, thereby establishing that the transition is definitely one from nonchaotic behavior to chaotic and back to nonchaotic behavior as the input beam is tracked across the crystal. Estimated errors in the calculation of the Lyapunov exponent are 5-10%.

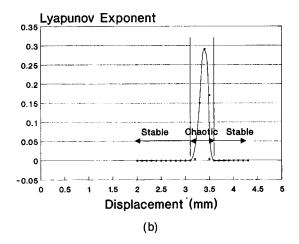
Figure 4(b) shows a similar result for $\theta = 15^{\circ}$. However, the chaotic region is reduced to a smaller region x < 3.6 mm and x > 3.1 mm. A Lyapunov exponent of $\lambda_1 =$

0.29 was obtained for x=3.4 mm. Figure 4(c) shows that for $\theta=50^\circ$ the phase-conjugate output remains stable.

CONCLUSION

For the first time, to our knowledge, we report investigations of the variation of the largest nonnegative Lyapunov exponent (λ_1) as a function of input-beam position for





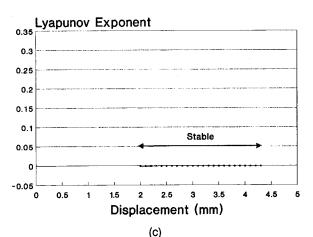


Fig. 4. Experimental results showing the calculated largest nonnegative Lyapunov exponent (λ_1) as a function of input-beam $(I_{\rm in}=2.1~{\rm W~cm^{-2}})$ position along the x axis of the crystal (5 mm): (a) $\theta=2^{\circ}$, (b) $\theta=15^{\circ}$, and (c) $\theta=50^{\circ}$.

SPPC in BaTiO₃, proving that for certain beam geometries variation in the input-beam position causes the phase-conjugated output to become increasingly chaotic within a narrowly defined region. This technique has the advantage over other methods of determining chaos because of its good parameter independence and insensitivity to the rate of visitation of the attractor, which is important for the specific case of SPPC.

The experimental analysis used in this paper could also be extended to investigations of instabilities that have been observed in other forms of phase conjugators in BaTiO₃, such as the mutually pumped phase conjugator. 32,33 We have also observed similar instabilities in SPPC at 514.5 nm in an ion-implanted planar waveguide in an impurity-doped blue BaTiO₃ crystal.⁵ This investigation is currently under way.

ACKNOWLEDGMENTS

We acknowledge the support of the Science and Engineering Research Council under grants GR/F 84256 and GR/H 21531 and the support of P. M. Jeffrey in the form of a research studentship. We also thank Alan Wolf for supplying further documentation concerning his algorithm for determining the Lyapunov exponents from a time series. This research was performed under the United Kingdom's Phase Conjugation Consortium.

REFERENCES

- 1. S. Sternklar, S. Weiss, M. Segev, and B. Fischer, "Beam coupling and locking of lasers using photorefractive fourwave mixing," Opt. Lett. 11, 528-530 (1986)
- R. W. Eason and A. M. C. Smout, "Bistability and noncommutative behavior of multiple-beam self-pulsing and self-pumping in BaTiO₃," Opt. Lett. **12**, 51-53 (1987).

 3. J. Feinberg, "Self-pumped, continuous-wave phase conjuga-
- tor using internal reflection," Opt. Lett. 7, 486-488 (1982).
- G. W. Ross and R. W. Eason, "Highly efficient self-pumped phase conjugation at near-infrared wavelengths by using nominally undoped BaTiO3," Opt. Lett. 17, 1104-1106 (1992).
- 5. G. W. Ross, P. Hribek, R. W. Eason, M. H. Garrett, and D. Rytz, "Impurity enhanced self-pumped phase conjugation in the near infrared in blue BaTiO3," Opt. Commun. 101, 60-64 (1993).
- S. W. James, K. E. Youden, P. M. Jeffrey, R. W. Eason, P. J. Chandler, L. Zhang, and P. D. Townsend, "BaTiO₃ waveguide self-pumped phase conjugator," Opt. Lett. 18, 1138-1140 (1993).
- 7. P. Günter, E. Voit, M. Z. Zha, and J. Albers, "Self-pulsation and optical chaos in self-pumped photorefractive BaTiO₃, Opt. Commun. 55, 210-214 (1985).
- 8. A. M. C. Smout, R. W. Eason, and M. C. Gower, "Regular oscillations and self-pulsating in self-pumped BaTiO3," Opt. Commun. 59, 77-82 (1986).
- 9. M. C. Gower and P. Hribek, "Mechanisms for internally selfpumped phase-conjugate emission from BaTiO3 crystals," J. Opt. Soc. Am. B 5, 1750-1757 (1988).
- A. Nowak, T. R. Moore, and R. A. Fisher, "Observation of internal beam production in barium titanate phase conjugators," J. Opt. Soc. Am. B 5, 1864-1878 (1988).

- 11. A. K. Majumdar and J. L. Kobesky, "Oscillations and possible transition to optical chaos in phase-conjugate waves in self-pumped BaTiO₃ at 514.5 nm," Opt. Commun. 75, 339-346 (1990).
- T. Rauch, C. Denz, and T. Tschudi, "Analysis of irregular fluctuations in a self-pumped BaTiO3 phase-conjugate mirror," Opt. Commun. 88, 160-166 (1992).
- 13. J. L. Kaplan and J. A. Yorke, "Chaotic behaviour of multidimensional difference equations," in Functional Differential Equations and Approximations of Fixed Points, H. O. Peitgen and H. O. Walther, eds., Vol. 730 of Lecture Notes in Physics (Springer-Verlag, Berlin, 1979).
- 14. P. Grassberger and I. Procaccia, "Measuring the strangeness of strange attractors," Physica D 9, 189-208 (1983).
- 15. J. D. Farmer, "Chaotic attractors of an infinite-dimensional dynamical system," Physica D 4, 366-393 (1982).
- 16. P. Grassberger, "Generalized dimensions of strange attractors," Phys. Lett A 97, 227-230 (1983).
- 17. H. G. E. Hentschel and I. Procaccia, "The finite number of generalized dimensions of fractals and strange attractors," Physica D 8, 435-444 (1983).
- 18. J.-P. Eckmann and D. Ruelle, "Ergodic theory of chaos and strange attractors," Rev. Mod. Phys. 57, 617-656 (1985).
- 19. P. Grassberger and I. Procaccia, "Characterization of strange attractors," Phys. Rev. Lett. 50, 346-349 (1983).
- G. Reiner, P. Meystre, and E. M. Wright, "Transverse dynamics of a phase-conjugate resonator. II: Fast nonlinear medium," J. Opt. Soc. Am. B 4, 865-874 (1987).
- 21. G. Reiner, M. R. Belić, and P. Meystre, "Optical turbulence and phase-conjugate resonators," J. Opt. Soc. Am. B 5, 1193-1209 (1988).
- W. Krölikowski, M. R. Belić, M. Cronin-Golomb, and A. Bledowski, "Chaos in photorefractive four-wave mixing with a single grating and a single interaction region," J. Opt. Soc. Am. B 7, 1204-1209 (1990).
- 23. K. D. Shaw, "Observation of chaos in off-Bragg photorefractive four-wave mixing," Opt. Commun. 97, 148-156 (1993).
- 24. D. J. Gauthier, P. Narum, and R. W. Boyd, "Observation of deterministic chaos in a phase-conjugate mirror," Phys. Rev. Lett. **58**, 1640–1643 (1987).
- 25. G. Sugihara and R. M. May, "Nonlinear forecasting as a way of distinguishing chaos from measurement error in time series," Nature **344,** 734–741 (1990).
- 26. M. Dämmig, C. Boden, and F. Mitschke, "On the detection of deterministic structures in irregular signals," Appl. Phys. B 55, 121-125 (1992).
- 27. A. Wolf, J. B. Swift, H. L. Swinney, and J. A. Vastano, "Determining Lyapunov exponents from a time series," Physica D 16, 285-317 (1985).
- 28. A. Wolf and T. Bessoir, "Diagnosing chaos in the space circle," Physica D 50, 239-258 (1991).
- 29. D. A. Rand and H. B. Wilson, "Detecting chaos: a critique of the Sugihara-May approach to time-series analysis," preprint (Warwick Preprints, University of Warwick, Coventry CV4 7AL, UK, 1991).
- 30. D. A. Rand and H. B. Wilson, "Detecting chaos in a noisy time-series," preprint (Warwick Preprints, University of Warwick, Coventry CV4 7AL, UK, 1993).
- 31. N. H. Packard, J. P. Crutchfield, J. D. Farmer, and R. S. Shaw, "Geometry from a time series," Phys. Rev. Lett. 45, 712-716 (1980).
- 32. S. W. James, G. Hussain, and R. W. Eason, "Dynamic instabilities in the mutually pumped bird wing phase conjugator,' in Technical Digest of Meeting on Photorefractive Materials, Effects, and Devices II (Sociéte Française d'Optique, Aussois, France, 1990), pp. 325-328.
- D. Wang, Z. Zhang, X. Wu, and P. Ye, "Instabilities in a mutually pumped phase conjugator of BaTiO3," J. Opt. Soc. Am. B 7, 2289-2293 (1990).