Observation of Deterministic Chaos in a Phase-Conjugate Mirror

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Deterministic chaos in the intensity of the beam produced by a barium titanate self-pumped phase-conjugate mirror has been observed. The correlation exponent of the strange attractor is found to depend on the crystal orientation and to lie within the range 1.2 to 2.4, and the order-two Renyi entropy is found to increase with increasing laser intensity and to be as large as 22 bits/sec. A standard model of self-pumped phase conjugation due to four-wave mixing has been generalized to include time dependence. This model predicts frequency shifts and chaotic behavior for the reflectivity.

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There has recently been great interest in determining the extent to which optical nonlinearities can cause optical systems to become unstable in their operating characteristics.[1] In this Letter we show experimentally and theoretically that certain phase-conjugate mirrors can be operated in a regime in which the output fluctuates temporally in a chaotic manner. This result is of interest both in demonstrating a limitation to the performance characteristics of phase-conjugate mirrors (PCM's) and in demonstrating that a passive nonlinear optical system can display deterministic chaos.

Unstable operation has long been observed in lasers, and it has been shown that under certain conditions these instabilities are a manifestation of deterministic chaos.[2] There has recently been considerable interest in determining the conditions under which passive nonlinear optical systems can also display chaotic behavior. Feedback by an external resonator often plays an important role in establishing such instabilities.[3] However, Silberberg and Bar-Joseph[4] have shown theoretically that the mutual interaction of two counterpropagating waves in a Kerr medium characterized by a noninstantaneous response can lead to chaotic behavior of the transmitted intensities. Furthermore, a number of authors[5] have shown that the polarization state of counterpropagating beams in a Kerr medium can also show chaotic behavior. Since strong counterpropagating waves are present in the standard geometry of phase conjugation by degenerate four-wave mixing, these predictions suggest that chaotic behavior may be expected for certain PCM's.[6]

Under special conditions, phase conjugation by four-wave mixing can occur without the use of externally applied pump waves. In the most common realization of a self-pumped PCM,[7] a laser beam is focused into a poled barium titanate crystal and an output beam is generated by means of a nonlinear interaction. The standard model[8] of self-pumped phase conjugation assumes the geometry shown in the inset to Fig. 1. The incident laser beam of field amplitude A_4 breaks up into new spatial components due to a self-focusing process known as *self*-beam fanning, creating a beam of amplitude A_1. This beam undergoes total internal reflection at the faces of the crystal and is thereby redirected so that it intersects the incident laser beam a second time. Analogously, a second beam of amplitude A_1' is generated at this intersection point and traverses the loop in the opposite direction. At each of the two interaction regions counterpropagating waves are therefore present, and the

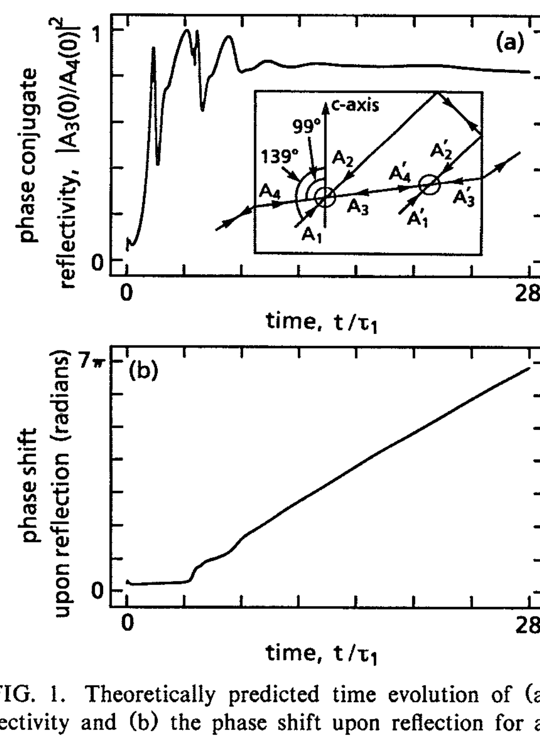

FIG. 1. Theoretically predicted time evolution of (a) the reflectivity and (b) the phase shift upon reflection for a self-pumped phase-conjugate mirror based upon four-wave mixing in two coupled interaction regions, whose geometry is shown in the inset. The values of the coupling coefficients appropriate for low laser intensities were used. Following an initial transient, the reflectivity reaches a constant value and the phase of the output wave increases linearly in time, implying that the conjugate wave is shifted in frequency with respect to the input.

phase-conjugate output wave is generated by the usual four-wave mixing process. Since the four interacting waves are strongly coupled within each of the interaction regions, it might be expected that this system would show instabilities, and in fact such instabilities have been observed experimentally.[9] The intent of the present Letter is to show that these instabilities are chaotic and to present a theoretical model that describes the nature of them.

Our model of a self-pumped PCM is an extension of that of MacDonald and Feinberg.[8] In their model, only one of the four gratings that contribute to the nonlinear coupling among the waves was explicitly included. In this approximation, they were able to find an analytic solution for the steady-state intensity of the phase-conjugation signal. In order to explore the stability characteristics of a self-pumped PCM, we have extended the MacDonald-Feinberg model by including the temporal evolution of the optical nonlinearity and by including the contributions of all four gratings. In particular, we consider the geometry shown in the inset to Fig. 1 in which the field amplitudes A_i are coupled according to

$$\left(\frac{\partial}{\partial z}+\frac{1}{c}\frac{\partial}{\partial t}\right)A_1=Q_1A_4-Q_2A_3-Q_3A_2, \tag{1a}$$

$$\left(\frac{\partial}{\partial z}-\frac{1}{c}\frac{\partial}{\partial t}\right)A_2=Q_1^*A_3-Q_2^*A_4-Q_3^*A_1, \tag{1b}$$

$$\left(\frac{\partial}{\partial z}-\frac{1}{c}\frac{\partial}{\partial t}\right)A_3=-Q_1A_2-Q_2^*A_1-Q_4A_4, \tag{1c}$$

$$\left(\frac{\partial}{\partial z}+\frac{1}{c}\frac{\partial}{\partial t}\right)A_4=-Q_1^*A_1-Q_2A_2-Q_4^*A_3, \tag{1d}$$

where Q_j denotes the amplitude of the grating created by the interference between two of the waves and is given by

$$\tau_1\dot{Q}_1+Q_1=(\gamma_1/I_0)(A_1A_4^*+A_2^*A_3), \tag{2a}$$

$$\tau_2\dot{Q}_2+Q_2=(\gamma_1/I_0)(A_1A_3^*+A_2^*A_4), \tag{2b}$$

$$\tau_3\dot{Q}_3+Q_3=(\gamma_3/I_0)A_1A_2^*, \tag{2c}$$

$$\tau_4\dot{Q}_4+Q_4=(\gamma_4/I_0)A_3A_4^*. \tag{2d}$$

A similar set of equations for the second interaction region are obtained through the substitutions $A\rightarrow A'$, $Q\rightarrow Q'$, and $z\rightarrow z'$. Here $I_0=\sum_j|A_j|^2$ is the total intensity, and γ_j is the photorefractive coupling coefficient. From low-intensity measurements of the two-beam coupling strength, we have determined that the ratios of these coefficients are given by $\gamma_1{:}\gamma_2{:}\gamma_3{:}\gamma_4=1{:}0.48{:}0.49{:}0.17$. These values are in good agreement with previously reported[10] values and are within a factor of 2 of those predicted on the basis of theory.[11] We have also found that for laser intensities greater than 10 W cm^{-2} the coupling coefficients are intensity dependent and that γ_2, γ_3, and γ_4 can even change sign. In deducing the form of Eqs. (2) we have assumed that the nonlinear response of the medium obeys a Debye relaxation relation, with a time constant τ_j which scales with the grating wave vector as predicted by theory.[12] For the geometry of Fig. 1, the ratios of these time constants are given by $\tau_1{:}\tau_2{:}\tau_3{:}\tau_4=1{:}7.1{:}7.2{:}7.2$. The coupling between the two interaction regions is described by the boundary conditions $A_1(l)=A_2'(l)$, $A_2(l)=A_1'(l)$, $A_3(l)=A_3'(0)$, and $A_4(l)=A_4'(0)$.

In order to investigate the dynamics of the self-pumped phase-conjugation process, we have performed a numerical integration of the set of equations given by (1) and (2). When this integration is performed with use of the low-intensity coupling coefficients given above and a short interaction length l, the solution predicts that the reflectivity reaches a stable steady-state value. However, when the integration is performed with use of a longer path length such that $\gamma_1 l=10$, the solution predicts that the phase of the generated wave increases linearly with time, although the intensity of the generated wave still reaches a constant value, as shown in Fig. 1. This result explains the frequency shift between the incident and conjugate waves which has been observed by many workers.[9,13] The value of the frequency shift can be modified by a change in the ratio of the coupling coefficients, which is accomplished experimentally by a rotation of the crystal. Frequency shifts in an external ring resonator due to mode-pulling effects have been observed[14] and theoretically analyzed.[15] It has been postulated that this mode-pulling effect is the origin of the frequency shift observed in the self-pumped PCM; however, our numerical results show that the nonlinear interaction alone is sufficient to produce instabilities in the phase of the output beam.

Qualitatively different behavior is predicted by our model when the integration is performed with use of values of the coupling coefficients appropriate to the higher laser intensities used in our experiment. Figure 2(a) shows the predicted behavior when the values $\gamma_1 l=6$, $\gamma_2 l=-3.7$, $\gamma_3 l=-3.8$, and $\gamma_4 l=-1.3$ are used. The predicted phase-conjugate intensity is seen to fluctuate wildly in time. Our computer simulations do not predict chaotic behavior or frequency shifts when only one grating is included in the model. For this reason, we believe that the origin of these instabilities is the interplay between the four contributions to the nonlinear coupling, each of which is characterized by a different time constant.

The experiment that demonstrates chaotic behavior in the output intensity of a PCM was performed with a barium titante crystal having dimensions of $4.8\times4.2\times4.1$ mm^3 and an unusually large free-charge density of 2×10^{17} cm^{-3}. The experiment was conducted in a thermally insulated enclosure on a floating optical table to prevent environmental effects from influencing the results. A single-mode argon-ion laser operating at 5145

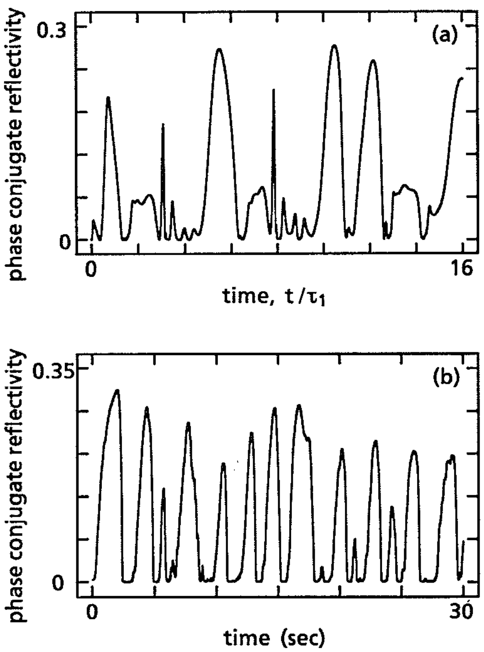

FIG. 2. (a) Theoretically predicted and (b) experimentally observed chaotic time evolutions of the reflectivity of a self-pumped phase-conjugate mirror. Values of the coupling coefficients appropriate for high laser intensity were used in the theoretical simulation.

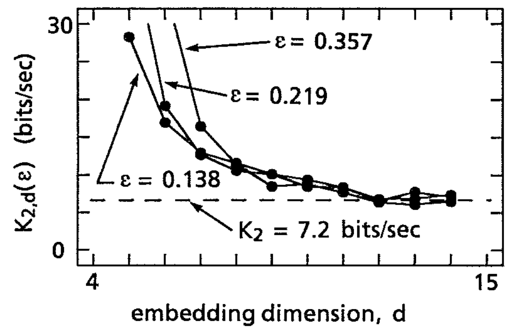

FIG. 3. Determination of the order-two Renyi entropy K_2 for the experimental signal shown in Fig. 2(b). The quantity $K_{2,d}(\epsilon)$ is seen to converge to the positive value $K_2 = 7.2$ bits/sec in the limit of large embedding dimension d and small hypersphere radius ϵ, implying that the signal is chaotic.

Å and stabilized to provide less than 0.1% intensity fluctuations was focused into the crystal to a spot size of $\simeq 80$ μm to provide the input field. The intensity of this field was in the range 50 to 1000 W cm^{-2}, where chaotic behavior is expected. To prevent instabilities due solely to feedback into the exciting laser,[13] a Faraday isolator[16] with an isolation ratio greater than 60 dB was used to isolate the laser from the beam returning from the barium titanate crystal. The output spectrum of the laser was monitored with a Fabry-Perot interferometer to ensure that the laser ran in a single longitudinal mode. The phase-conjugate intensity was recorded with a twelve-bit digitizer and stored in a microcomputer.

We find that for a broad range of laser intensities and crystal orientations, the intensity of the phase-conjugate wave fluctuates wildly in time. Typical results are shown in Fig. 2(b). We have determined that the fluctuations in the phase-conjugate signal are uncorrelated from the small fluctuations in the laser intensity; specifically, the normalized cross covariance is less than 3%.

In order to determine that these fluctuations are chaotic, we have calculated the correlation exponent ν and the order-two Renyi entropy K_2 of the associated strange attractor from the time series of measured reflectivities $\{R(t_j) \mid j = 1, \ldots, N\}$.[17] We form d-dimensional time-delay vectors

$$\mathbf{Y}_j = \{R(t_j), R(t_j+\delta), \ldots, R(t_j+(d-1)\delta) \mid j = 1, 2, \ldots, N-d\},$$

where $n = 16000$ and the time delay δ is determined from the first minimum in the autocorrelation of the phase-conjugate signal. These vectors are used to calculate the correlation integral $C_d(\epsilon)$ which is proportional to $\exp[\nu - K_{2,d}(\epsilon)\tau d]$, where ϵ is the hypersphere radius and τ is the sampling interval. The order-two Renyi entropy is determined by

$$K_2 = \lim_{d \to \infty} \lim_{\epsilon \to 0} K_{2,d}(\epsilon).$$

We have analyzed the experimental data in Fig. 2(b) and find that $\nu = 1.3$. In Fig. 3 we plot $K_{2,d}(\epsilon)$ as a function of the embedding dimension d for several different values of ϵ. It is seen that for large values of d, $K_{2,d}(\epsilon)$ converges to the *positive* value of 7.2 bits sec^{-1}, implying that the signal is chaotic. In addition, we have determined that $\nu = 1.2$ for the attractor associated with the theoretical curve shown in Fig. 2(a) and that $K_2 = (13.9 \text{ bits})\tau_1^{-1}$.

We have found experimentally that the measured value of the correlation exponent ν depends sensitively upon the crystal orientation. Exponents in the range 1.2 to 2.4 have been observed. We have also conducted an experiment in which we varied the input intensity between 150 and 1000 W cm^{-2} while holding the crystal position fixed. We find that ν remains nearly constant, while K_2 varies linearly with input intensity between the values 3 and 22 bits sec^{-1}. These observations suggest that geometrical factors related to the nature of the self-pumped phase-conjugation process determine the trajectory in phase space, whereas the laser intensity determines the time scale of the chaotic evolution.

Kaplan and Yorke[18] have conjectured that for continuous chaotic systems the fractal dimension D must be greater than 2. The correlation exponent ν, which is a lower bound on D, is often a good estimator of D. However, the measured and theoretical values of ν for our system are under some conditions less than 2, suggesting that at least under these conditions ν does not accurately predict D. It is seen from Fig. 2 that the phase-conjugate reflectivity is often near zero, implying that some regions of the attractor are visited more frequently than others. Since the fractal dimension is insensitive to the rate of visitation, whereas the correlation exponent is sensitive to this effect,[17] the two dimensions are expected to have different numerical values.

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