Polarization Instabilities of Counterpropagating Laser Beams in Sodium Vapor

Daniel J. Gauthier, Michelle S. Malcuit, and Robert W. Boyd
The Institute of Optics, University of Rochester, Rochester, New York 14627
(Received 16 June 1988)

We have observed temporal instabilities in the polarizations of counterpropagating laser beams in atomic sodium vapor. For intensities slightly above the instability threshold, the polarizations fluctuate periodically. For higher intensities, the fluctuations are chaotic and the system evolves on a strange attractor whose fractal dimension increases with increasing laser intensity.

PACS numbers: 42.50.Tj, 42.65.−k

One of the conceptually simplest processes in nonlinear optics is the mutual interaction of two counterpropagating laser beams in a nonlinear medium. However, recent work has shown that this interaction can give rise to extremely complicated dynamical behavior, including chaotic temporal evolution of the intensities and polarizations of the transmitted waves. The possibility that instabilities can occur in the interaction of laser beams are very important from a practical point of view, because instabilities can lead to the degradation of processes that utilize counterpropagating laser beams, such as optical bistability, phase conjugation, squeezing, and nonlinear laser spectroscopy.

Silberberg and Bar-Joseph[1] first pointed out theoretically that instabilities can occur in counterpropagating laser beams for the case of scalar waves interacting in a nonlinear Kerr medium characterized by noninstantaneous response. The origin of this instability is the combined action of gain due to four-wave mixing and distributed feedback due to scattering from the grating formed by the interference of the incident laser beams. Quite recently, Khitrova, Valley, and Gibbs[2] have observed an instability in the intensities (but not the polarizations) of counterpropagating laser beams in sodium vapor. Gaeta *et al.*[3] have pointed out that when the vector nature of light is included in the theoretical formulation for the case of a nonlinear Kerr medium, the nature of the predicted instability is quite different in that the state of polarization and not necessarily the intensity of the light becomes unstable. Furthermore, these authors showed that, in general, the threshold for the polarization instability is lower than that predicted by the scalar theory of Silberberg and Bar-Joseph,[1] and that the polarization instability can exist even for the case of a medium with instantaneous response. Slightly above the threshold for this instability, periodic fluctuations of the polarization state were predicted, while chaotic behavior was predicted for input intensities sufficiently far above threshold.

In this Letter, we present experimental results for the case of sodium vapor showing that the polarizations of counterpropagating laser beams are temporally unstable. We have measured the time evolution of the resulting fluctuations and hence have studied the dynamical evolution of the system. We have determined the route to chaos for the system, have constructed the attractors describing the evolution of the system in phase space, and have determined the fractal dimensions and metric entropies characterizing these attractors. The nonlinearity of sodium vapor cannot be totally explained by the Kerr-medium model; however, our results are in good qualitative agreement with the predictions of Ref. 3. For example, the threshold that we have measured for the polarization instability is considerably lower than that reported by Khitrova, Valley, and Gibbs[2] for the intensity instability. These results suggest that polarization instabilities of the type predicted in Ref. 3 are not unique to the Kerr nonlinearity but can occur whenever vector waves interact in a nonlinear medium. There have been a number of additional experimental studies showing that instabilities and chaotic behavior can occur in passive nonlinear optical systems; however, such studies have required the use of external feedback[4] or complicated geometries[5] to induce the instability.

Our experimental investigation of the stability of counterpropagating laser beams made use of the highly nonlinear response of the sodium $3S_{1/2} \rightarrow 3P_{1/2}$ atomic transition. Counterpropagating beams were derived from the output of a continuous-wave dye laser whose frequency was tuned through the atomic transition frequency. The input power of one of the beams (which we will call the forward beam) was held fixed at a value of 160 mW and measurements were made for various input powers in the other beam (the backward beam). Polarizing beam splitters were placed in each beam so that the input beams were linearly polarized with parallel polarizations. Our experiment involved measuring for the forward beam the fluctuations and average power of the light emitted in the polarization orthogonal to that of the input beams. It was found that the threshold for the instability depends sensitively on the atomic number density, the buffer gas pressure, and the precision to which the two beams were counterpropagating. We found that the instability threshold decreased with decreasing buffer gas pressure, and for this reason all of the data reported here were collected using a low helium-buffer-gas pressure of

~ 15 mTorr. For an interaction length of 5 cm and a beam diameter of 750 μm, the number density which produced the largest conversion into the orthogonal polarization was found to be $\sim 2\times 10^{13}$ atoms cm^{-3}.

We first characterized the instability in terms of its dependence on the laser frequency. We measured the average power generated in the orthogonal] polarization for the forward beam as a function of the detuning of the laser frequency from the atomic resonance. Figure 1 displays this dependence for an input power of 150 mW in the backward beam and a sodium number density of $\sim 2\times 10^{13}$ atoms cm^{-3}. The tick marks indicate the positions of the four hyperfine components of the $3S_{1/2}\rightarrow 3P_{1/2}$ sodium transition. We see that as much as 20% of the incident power can be converted into the orthogonal polarization. The excitation spectrum is also seen to be very rich, containing spectral features that are much narrower than the $\sim$2-GHz Doppler width. We have found that the generated light associated with each of these features has a distinct optical spectrum and temporal behavior. The spectrum of the light generated in the orthogonal polarization was measured by a scanning Fabry-Perot interferometer having a resolution of ~ 250 MHz and by a spectrum analyzer having a frequency range of 100 Hz to 1 GHz. The temporal evolution of the intensity was measured with a ten-bit transient digitizer with a sampling rate of 200 MHz.

For the central feature of Fig. 1 [i.e., for a laser detuning of 1.01 GHz to the high-frequency side of the $3S_{1/2}(F=2)\rightarrow 3P_{1/2}(F=2)$ transition], the generated light was found to be at the same frequency as the input light. For this value of the laser detuning, the instability threshold corresponds to a power of 15 mW in the backward beam. Slightly above this threshold, the power emitted in the orthogonal polarization was observed to be nearly constant in time, corresponding to the dc instability predicted by the Kerr-medium model. For powers well above the threshold value, weak oscillations were observed in the generated signal with a frequency ~ 10 MHz.

For the spectral feature of Fig. 1 to the low-frequency side of the central feature, the emission spectrum contains three frequencies: a component at the input laser frequency and two sidebands symmetrically displaced by 1.8 GHz, which is the ground-state hyperfine splitting.

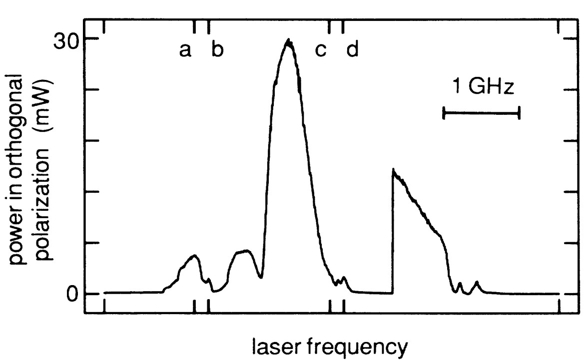

FIG. 1. Laser-frequency dependence of the instability in the polarizations of counterpropagating waves. The average power generated in the polarization orthogonal to that of the input beams is plotted as a function of the laser frequency for a power of 160 mW in the forward beam and 150 mW in the backward beam. The four tick marks on the horizontal axis, labeled a–d, indicate the positions of the $(F=2)\rightarrow(F=1)$, $(F=2)\rightarrow(F=2)$, $(F=1)\rightarrow(F=1)$, and $(F=1)\rightarrow(F=2)$ hyperfine components of the $3S_{1/2}\rightarrow 3P_{1/2}$ sodium transition, respectively.

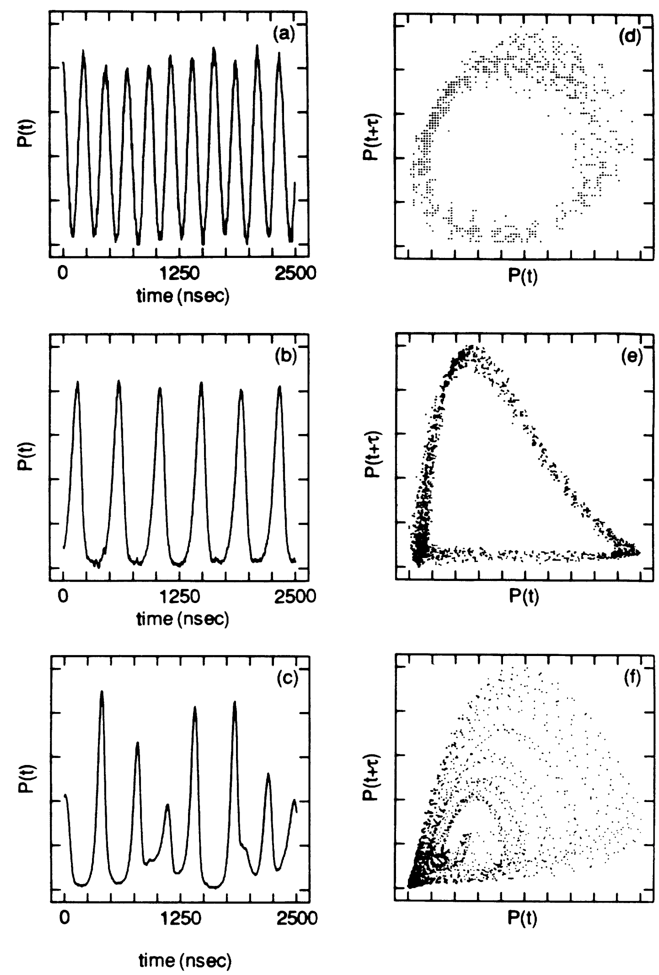

FIG. 2. Temporal evolution of the light generated in the orthogonal polarization. The laser was detuned 310 MHz to the low-frequency side of the $3S_{1/2}$ $(F=2)\rightarrow 3P_{1/2}(F=2)$ transition, the input power of the forward beam was 160 mW, and the input power in the backward beam was (a) 25 mW, (b) 27 mW, and (c) 31 mW. The attractors shown in (d)–(f) correspond to the time series shown in (a)–(c). These attractors are reconstructed by our plotting the power emitted in the orthogonal polarization at time $t+\tau$ vs that emitted at time t for time delays τ equal to (d) 55 ns, (e) 100 ns, and (f) 50 ns.

The majority of the power was emitted in the low-frequency (Stokes) sideband. This observation suggests that for certain laser frequencies, stimulated hyperfine Raman scattering[6] is the relevant nonlinear coupling mechanism. The spectrum of the strong (trapezoid-shaped) feature to the high-frequency side of the central feature has similar frequency components as the low-frequency feature, with the majority of the power contained in the anti-Stokes sideband.

Rich temporal behavior was observed for the low-frequency feature. Figure 2 displays this behavior for a detuning of 310 MHz to the low-frequency side of the $3S_{1/2}(F=2) \rightarrow 3P_{1/2}(F=2)$ transition. In this case the instability threshold corresponds to a power of 24 mW in the backward beam. For a power of 25 mW, oscillatory evolution was observed, as shown in Fig. 2(a). With small increases in the power, the evolution changed to self-pulsing [Fig. 2(b)], and then to wildly fluctuating oscillations [Fig. 2(c)].

To characterize the nature of the time behavior shown in Figs. 2(a)-2(c), we have studied the evolution of the system in a two-dimensional, time-delay phase space.[7] In each of the three cases illustrated in Figs. 2(d)-2(f), the trajectory is seen to fill phase space in a highly nonuniform manner, suggesting that the observed fluctuations are not due to random noise. The trajectories shown in Figs. 2(d) and 2(e) [corresponding to the time evolutions shown in Figs. 2(a) and 2(b), respectively] form closed loops, broadened somewhat by noise introduced by our detection system, demonstrating that the evolution of Figs. 2(a) and 2(b) is periodic. The trajectory of Fig. 2(f), which corresponds to the wildly fluctuating time series of Fig. 2(c), appears not to form a closed curve. This behavior suggests that the evolution is chaotic and that the phase-space trajectory forms a strange attractor.

To verify these conclusions quantitatively, we have calculated the correlation dimension, D_2, and the order-2 Reyni entropy, K_2, of our experimental attractors using both the fixed-mass method of Badii and Politi[8] and the fixed-volume method of Grassberger and Procaccia.[9] In Fig. 3, we show the dependence of D_2 and K_2 on the laser power for a detuning of 310 MHz to the low-frequency side of the $3S_{1/2}(F=2) \rightarrow 3P_{1/2}(F=2)$ transition. It is seen that a Hopf bifurcation occurs at the instability threshold of 24 mW in the backward beam, where the evolution changes from that of a fixed point to that of a limit cycle. For powers of 25 and 27 mW, the dimension of the attractors is nearly unity and the entropy is approximately zero, showing that the evolution is periodic in these cases. This result confirms the assessment given above based on a visual inspection of the attractors. For a power of 31 mW, we find that the correlation dimension is equal to 2.7 and the order-2 Reyni entropy is equal to 6×10^7 bits/s. The fact that the order-2 Reyni entropy is greater than zero proves that the system is chaotic in this case. For still higher laser powers, the calculated dimensions grow rapidly with increasing laser power. The error bars become large as the dimension increases because, for both the fixed-mass and the fixed-volume methods, the scaling region[10] is small because of the limited size of our 16384-point data sets. The increase of the dimension of the attractor with increasing laser power shows quantitatively that the dynamics becomes more complicated as the system is driven further into the nonlinear regime.

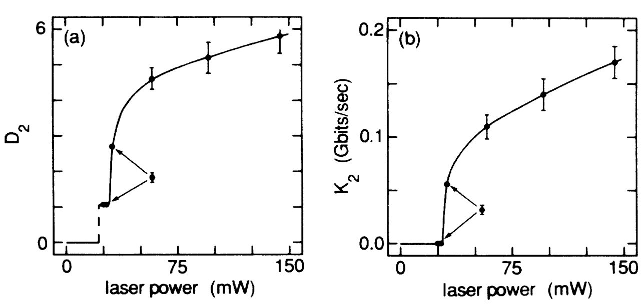

FIG. 3. (a) Correlation dimensions, D_2, and (b) order-2 Renyi entropies, K_2, of the attractors reconstructed from time series data for a detuning of 310 MHz to the low-frequency side of the $3S_{1/2}(F=2) \rightarrow 3P_{1/2}(F=2)$ transition and for a power of 160 mW in the forward beam as a function of the power in the backward beam. For powers greater than 27 mW, the order-2 Renyi entropy is greater than zero showing that the system is chaotic.

In conclusion, we have shown that counterpropagating laser beams in an atomic vapor can be unstable to the growth of periodic or chaotic temporal fluctuations in the polarizations of the transmitted beams.

We gratefully acknowledge useful discussions of this work with I. Bar-Joseph, M. Beck, A. Gaeta, I. McMackin, P. Narum, and Y. Silberberg. Support for this work was provided by the National Science Foundation, the Office of Naval Research, and the U.S. Army Research Office.

[1] Y. Silberberg and I. Bar-Joseph, Phys. Rev. Lett. **48**, 1541 (1982), and J. Opt. Soc. Am. B **1**, 662 (1984).

[2] G. Khitrova, J. F. Valley, and H. M. Gibbs, Phys. Rev. Lett. **60**, 1126 (1988).

[3] A. L. Gaeta, R. W. Boyd, J. R. Ackerhalt, and P. W. Milonni, Phys. Rev. Lett. **58**, 2432 (1987).

[4] R. Bonifacio, M. Gronchi, and L. A. Lugiato, Opt. Commun. **30**, 129 (1979); K. Ikeda, H. Daido, and O. Akimoto, Phys. Rev. Lett. **45**, 709 (1980); H. M. Gibbs, F. A. Hopf, D. L. Kaplan, and R. L. Shoemaker, Phys. Rev. Lett. **46**, 474 (1981); H. Nakatsuka, S. Asaka, H. Itoh, K. Ikeda, and M. Matsuoka, Phys. Rev. Lett. **50**, 109 (1983); F. Mitschke, J. Mlynek, and W. Lange, Phys. Rev. Lett. **50**, 1660 (1983); R. G. Harrison, W. S. Firth, C. A. Emshary, and I. A. Al-Saidi, Phys. Rev. Lett. **51**, 562 (1983); G. C. Valley and G. J.

Dunning, Opt. Lett. **9**, 513 (1984); H. Nakajima and R. Frey, Phys. Rev. Lett. **54**, 1798 (1985); P. Salieri, G. Giusfredi, S. Cecchi, and F. T. Arecchi, J. Opt. Soc. Am. B **5**, 1166 (1988).

[5]D. J. Gauthier, P. Narum, and R. W. Boyd, Phys. Rev. Lett. **58**, 1640 (1987).

[6]P. Kumar and J. H. Shapiro, Opt. Lett. **10**, 226 (1985).

[7]N. H. Packard, J. P. Crutchfield, J. D. Farmer, and R. S. Shaw, Phys. Rev. Lett. **45**, 712 (1980); H. Froeling, J. P. Crutchfield, D. Farmer, N. H. Packard, and R. Shaw, Physica (Amsterdam) **3D**, 605 (1981).

[8]R. Badii and A. Politi, Phys. Rev. Lett. **52**, 1661 (1984), and J. Stat. Phys. **40**, 725 (1985).

[9]P. Grassberger and I. Procaccia, Phys. Rev. Lett. **50**, 346 (1983), and Phys. Rev. A **28**, 2591 (1983), and Physica (Amsterdam) **9D**, 189 (1983).

[10]E. J. Kostelich and H. L. Swinney, in *Chaos and Related Nonlinear Phenomena,* edited by I. Procaccia and M. Shapiro (Springer-Verlag, New York, 1987); G. Broggi, J. Opt. Soc. Am. B **5**, 1020 (1988).