Observation of Brillouin chaos with counterpropagating laser beams

O. Kulagin and G. A. Pasmanik

Institute of Applied Physics, Academy of Sciences of the USSR, Niznij Novgorod, USSR

Alexander L. Gaeta, Thomas R. Moore, Glenn J. Benecke, and Robert W. Boyd

Institute of Optics, University of Rochester, Rochester, New York 14627

Received February 4, 1991; revised manuscript received May 13, 1991

We have observed chaotic fluctuations in the transmitted intensities of laser beams counterpropagating in Freon-113 ($C_2Cl_3F_3$). As the input intensities are increased, the system enters the chaotic regime by means of the period-doubling route, in accordance with theoretical predictions.

Counterpropagating laser beams in nonlinear-optical media can give rise to temporal instabilities and deterministic chaos. These effects are significant because many optical devices use counterpropagating waves. Also, the occurrence of these instabilities demonstrates that a conceptually simple nonlinear-optical interaction can exhibit complicated dynamical behavior. Silberberg and Bar-Joseph[1] first predicted that, for input intensities above a certain threshold value, the amplitudes of counterpropagating fields in a nonlinear Kerr medium can become temporally unstable. These instabilities can display chaotic behavior as the input intensities are increased. Similar behavior is predicted for interactions within a collection of two-level atoms,[2] and oscillatory instabilities were observed by Khitrova *et al.*[3] in atomic sodium vapor. Gaeta *et al.*[4] showed that, when the tensor nature of the Kerr nonlinearity is considered, the polarizations of the counterpropagating waves can become chaotic. This behavior was observed experimentally by Gauthier *et al.*[5] The transverse profiles of counterpropagating laser beams in Kerr media are also predicted to exhibit temporally instabilities,[6] and these instabilities were observed in the form of conical emission by Grynberg *et al.*[7]

Laser beams counterpropagating through a Brillouin-active medium are also predicted to exhibit a wide range of dynamical behavior.[8–10] The intensities necessary for observation of temporal instabilities with counterpropagating waves can be several times lower than the threshold intensity for normal, single-beam stimulated Brillouin scattering (SBS). For the case in which the ratio of the Brillouin linewidth Γ to the Brillouin frequency shift Ω is less than 0.3, the temporal evolution of the system in the instability regime does not undergo chaotic evolution but exhibits oscillations at a frequencies close to the Brillouin frequency. This oscillatory behavior was observed[11] in carbon disulfide, for which the value of Γ/Ω is approximately equal to 0.01. However, for Brillouin media that possess a broad Brillouin linewidth it has been predicted that the system will display a period-doubling route to chaos as the input intensities are increased.[9]

The experimental apparatus is shown schematically in Fig. 1. A frequency-doubled Nd:YAG laser, operating in a single transverse and a single longitudinal mode, generates pulses that are ~25 ns in duration (FWHM) with a repetition rate of 10 Hz. Each pulse is split into two beams of nominally equal energy by a thin-film polarizer. After the plane of polarization of one of the beams is rotated such that the two beams have parallel polarization, the beams are focused from opposite sides into a cell containing Freon-113 ($C_2Cl_3F_3$) at room temperature. Each beam is focused by a 15-cm-focal-length lens, and care is taken to ensure that the beams are counterpropagating and overlap completely within the cell. An uncoated glass wedge reflects a portion of one of the transmitted beams for observation. In our geometry the threshold for single-beam SBS is measured to be ~1.4 mJ. (All energy measurements are accurate to ±20% absolute and to ±5% relative.) The temporal evolution of the pulse is observed by using a streak camera with a 2-ps resolution, and spectral data are obtained by using a Fabry–Perot interferometer with a free spectral range of 0.2 cm^{-1}. The measured values of $\Gamma/2\pi$ and $\Omega/2\pi$ for Freon-113 at a wavelength of $\lambda = 0.53$ μm are found to be 870 MHz and 2.4 GHz, respectively. These values yield a value of 0.35 for the ratio Γ/Ω, which is larger than the minimum value of 0.3 that theoretical analysis predicts to be necessary for observing chaos.

The temporal evolution of the power of one of the transmitted beams is shown in Fig. 2 for various input pulse energies. In each case a 2-ns window surrounding the peak of the laser pulse is shown. For an input energy of 0.7 mJ [Fig. 2(a)] the output is seen to oscillate with a period (T = 310 ps) that is shorter than the period associated with the Brillouin frequency ($T_B = \Omega^{-1} = 412$ ps). This shift is in agreement with the theoretical prediction that the frequency at which the instability grows most rapidly can be pulled to a frequency that is larger than the Brillouin frequency of the medium. Note that the threshold energy (0.7 mJ) for the onset of the temporal instability shown in Fig. 2(a) is approximately one half

0740-3224/91/102155-03$05.00

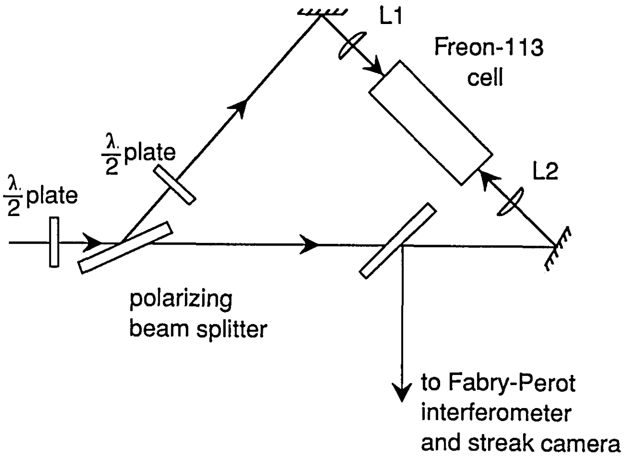

Fig. 1. The experimental apparatus. Lenses L1 and L2 each have a focal length of 15 cm.

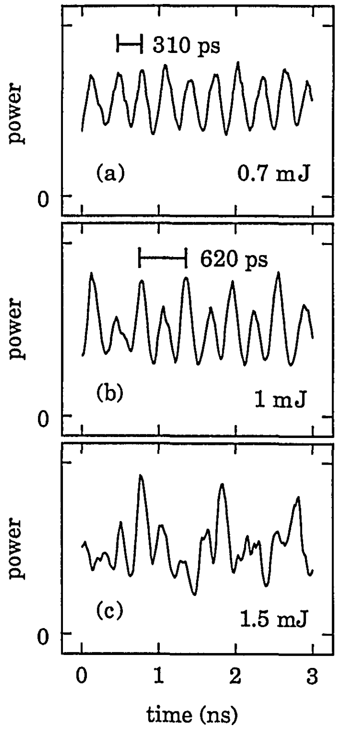

Fig. 2. Temporal evolution of the power of one of the transmitted laser beams for different values of the input energy in each beam. The sequence displays a period-doubling route to chaos.

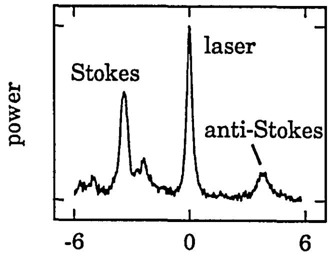

Fig. 3. Spectrum of the one of the transmitted beams in the chaotic regime. Both Stokes and anti-Stokes radiation are observed.

the threshold energy for the occurrence of single-beam SBS. For a measured input energy of 1 mJ the output is seen to oscillate with a period twice the period shown in Fig. 2(a). At higher input energies [Fig. 2(c)] the output becomes highly erratic. The observation of a period-doubling sequence in Figs. 2(a) and 2(b) suggests that the observed temporal evolution in Fig. 2(c) is chaotic. However, as a result of the relatively short time series that is obtained from a single laser pulse, determination of the Kolmogorov–Renyi entropy[12] is not possible.

Figure 3 shows a typical spectrum of the transmitted beam in the chaotic regime. The spectrum is seen to contain a component at the incident laser frequency, a Stokes component, and an anti-Stokes component. The appearance of the anti-Stokes component is in agreement with theoretical predictions.[9] The anti-Stokes component of the wave traveling in one direction is driven by the four-wave mixing process that involves the Stokes component of the wave traveling in the opposite direction and each of the counterpropagating pump waves. On the Stokes side of the laser frequency the light contains a strong component that is separated from the laser frequency by 3.0 GHz. A weaker component at the Brillouin frequency (2.4 GHz) is also present, and we believe that it is due to the occurrence of normal, single-beam SBS in a region just outside the focal volume.

We have also investigated the dynamical behavior for the case in which the beams are cross polarized. The threshold for instability is found to be the same as for the case in which the beams are polarized parallel to one another. However, in the regime of instability chaotic behavior is not observed, and the output oscillates with a period that is close to the Brillouin period. This behavior suggests that the distributed feedback from the nonlinear index grating associated with the standing-wave pattern between the counterpropagating beams[9] is critical to the dynamics of the system.

In conclusion, we have demonstrated that the transmitted intensities of counterpropagating beams in a Brillouin-active medium can exhibit chaotic behavior by means of a period-doubling route. These results, along with the observation of an anti-Stokes component in the transmitted spectrum, are in good agreement with the theoretical predictions.

We gratefully acknowlege discussions with B. Ya. Zel'dovich and P. Narum. The majority of the experiments reported here were performed in Rochester and were supported by the U.S. Army through the University Research Initiative and by National Science Foundation grant ECS-8802761. R. W. Boyd also acknowledges the receipt of a NATO travel grant.

REFERENCES

1. Y. Silberberg and I. Bar-Joseph, Phys. Rev. Lett. **48,** 1541 (1982).
2. I. Bar-Joseph and Y. Silberberg, Phys. Rev. A **36,** 1731 (1987); D. J. Gauthier "Instabilities and chaos of laser beams propagating through nonlinear optical media," Ph.D. dissertation (University of Rochester, Rochester, New York, 1989).
3. G. Khitrova, J. F. Valley, and H. M. Gibbs, Phys. Rev. Lett. **60,** 1126 (1988).
4. A. L. Gaeta, R. W. Boyd, J. R. Ackerhalt, and P. W. Milonni, Phys. Rev. Lett. **58,** 2432 (1987).

5. D. J. Gauthier, M. S. Malcuit, and R. W. Boyd, Phys. Rev. Lett. **61,** 1827 (1988).
6. W. J. Firth and C. Pare, Opt. Lett. **13,** 1096 (1988); G. Grynberg and J. Paye, Europhys. Lett. **8,** 29 (1990); G. G. Luther and C. J. McKinstrie, J. Opt. Soc. Am. B **7,** 1125 (1990).
7. G. Grynberg, E. LeBihan, P. Verkerk, P. Simoneau, J. R. R. Leite, D. Bloch, S. Le Boiteux, and M. Ducloy, Opt. Commun. **67,** 363 (1988).
8. B. Ya. Zel'dovich and V. V. Shkunov, Sov. J. Quantum Electron. **12,** 223 (1982).
9. P. Narum, A. L. Gaeta, M. D. Skeldon, and R. W. Boyd, J. Opt. Soc. Am. B **6,** 1709 (1988).
10. R. G. Harrison, J. S. Uppal, A. Johnstone, and J. V. Maloney, Phys. Rev. Lett. **65,** 167 (1990).
11. A. L. Gaeta, M. D. Skeldon, R. W. Boyd, and P. Narum, J. Opt. Soc. Am. B **6,** 1709 (1989).
12. P. Grassberger and I. Procaccia, Phys. Rev. Lett. **50,** 346 (1983).