Observation of instabilities of laser beams counterpropagating through a Brillouin medium

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Received January 27, 1989; accepted May 22, 1989

We show experimentally that counterpropagating laser beams in a Brillouin-active medium are unstable to the growth of intensity fluctuations. The intensities of the transmitted beams are found to oscillate at the Brillouin frequency of the medium, which is equal to 7.7 GHz for our experimental conditions of excitation of carbon disulfide at a wavelength of 532 nm. The depth of modulation is found to be as large as 25%. The threshold for this instability depends on the ratio of the intensities of the counterpropagating beams. The threshold is lowest for the case of equal input intensities and is found to be as much as 33% lower than the threshold for single-beam stimulated Brillouin scattering.

It is known that extremely simple nonlinear-optical interactions can give rise to complicated dynamical behavior.[1] In particular, there has been considerable interest in the dynamical behavior of counterpropagating laser beams interacting in a nonlinear medium. It has been shown theoretically[2–5] that for a variety of nonlinear interactions the intensities of the waves can become unstable to the growth of temporal fluctuations and that under certain circumstances these instabilities can be chaotic in nature. Such instabilities were recently observed for the case of interaction by means of the nonlinear response of an atomic-sodium vapor.[6,7] In this paper we present experimental results regarding instabilities resulting from coupling that is due to the Brillouin interaction.

Several authors[4,5,8] have presented theoretical analyses that predict that instabilities can develop in the intensities of equal-frequency counterpropagating waves in a Brillouin-active medium. In addition, experimental and theoretical studies have been reported for the case of counterpropagating waves of different frequencies.[9] Instabilities of counterpropagating waves have been shown[10] to be related to the large reflectivities that are achievable with Brillouin-enhanced four-wave mixing.[11] Here we present experimental confirmation of many of these predictions. In our previous work[5] we showed that the complex amplitudes E_f and E_b of forward- and backward-going waves are coupled by means of their interaction with an acoustic wave of amplitude ρ according to the set of equations

$$\frac{\partial E_f}{\partial z}+\frac{1}{(c/n)}\frac{\partial E_f}{\partial t}=i\kappa\rho E_b, \tag{1a}$$

$$-\frac{\partial E_b}{\partial z}+\frac{1}{(c/n)}\frac{\partial E_b}{\partial t}=i\kappa\rho^* E_f, \tag{1b}$$

and

$$\frac{\partial^2\rho}{\partial t^2}+\Gamma\frac{\partial\rho}{\partial t}+\Omega^2\rho=\frac{q^2\gamma}{8\pi}E_f E_b^*, \tag{1c}$$

where the complex amplitudes are related to the physical fields through

$$E_{\mathrm{TOT}}(z,t)=\tfrac{1}{2}E_f(z,t)\exp[i(kz-\omega t)]+\tfrac{1}{2}E_b(z,t)\exp[i(-kz-\omega t)]+\mathrm{c.c.} \tag{2a}$$

and

$$\rho_{\mathrm{TOT}}(z,t)=\tfrac{1}{2}\rho(z,t)e^{iqz}+\mathrm{c.c.} \tag{2b}$$

Here c/n denotes the velocity of light in the medium, $\kappa=\gamma\omega/4\rho_0 nc$ is a coupling coefficient, ω is the angular frequency of the light waves, γ is the electrostrictive constant, ρ_0 is the mean density of the medium, $q=2\omega n/c$ is the wave vector of the Brillouin-resonant acoustic wave, $\Omega=qv$ is the angular frequency of the acoustic disturbance, v is the velocity of sound, and Γ is the Brillouin linewidth. The set of Eqs. (1) yields the simple steady-state solution

$$\rho^0(z)=\frac{\gamma}{8\pi v^2}E_f^0(z)E_b^{0*}(z), \tag{3a}$$

$$E_f^0(z)=E_f^0(0)\exp\left(ig\frac{\Gamma}{\Omega}I_b z\right), \tag{3b}$$

and

$$E_b^0(z)=E_b^0(L)\exp\left[ig\frac{\Gamma}{\Omega}I_f(L-z)\right], \tag{3c}$$

where $g=\gamma^2\omega^2/\Gamma nvc^3\rho_0$ is the line-center Brillouin-amplitude gain coefficient and $I_f=(nc/8\pi)|E_f^0(0)|^2$ and $I_b=(nc/8\pi)|E_b^0(L)|^2$ are the input intensities of each wave. [The threshold for the usual single-beam stimulated Brillouin scattering (SBS) process is usually taken as the condition

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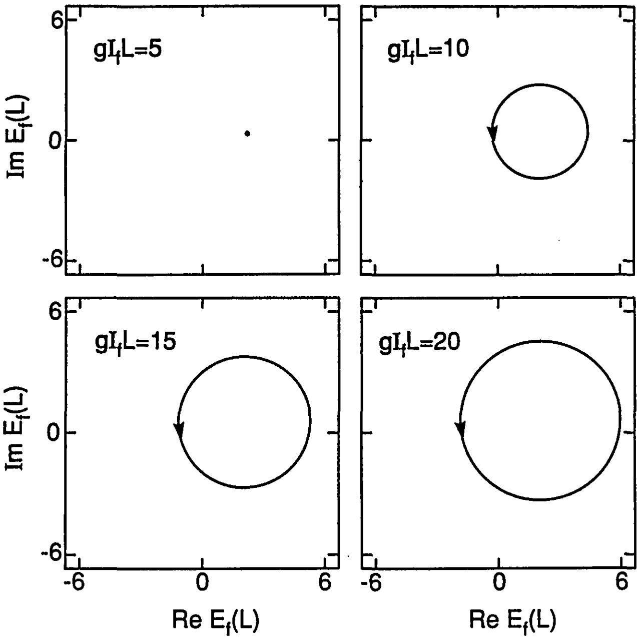

Fig. 1. Phase-space trajectories of the complex field amplitude of the transmitted forward-traveling wave for four different values of the input intensities for the case of $\Gamma/\Omega = 0.03$, $\Delta kL = 72$, and $I_f = I_b$. For an input intensity such that $gI_fL = 5$ the output field amplitude settles to a steady-state value. For higher input intensities (gI_fL = 10, 15, 20) the output field amplitude oscillates periodically in a circular orbit corresponding to an output wave containing the laser frequency and a Stokes sideband.

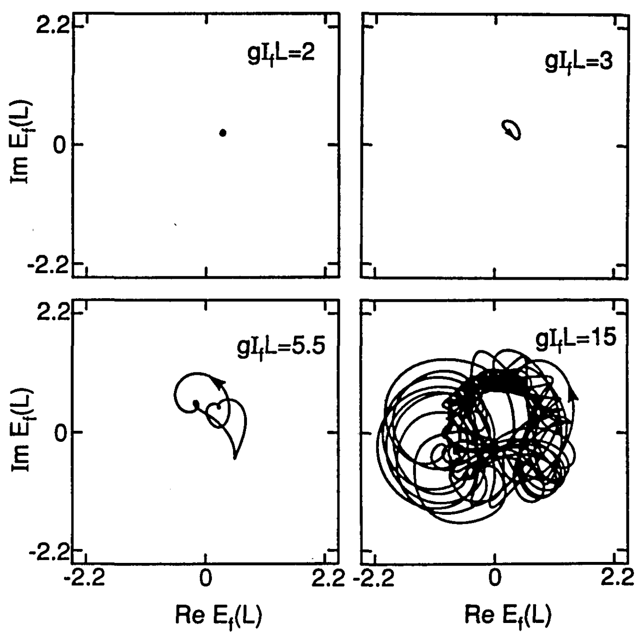

Fig. 2. Phase-space trajectories of the complex field amplitude of the transmitted forward-traveling wave for four different values of the input intensities for the case of $\Gamma/\Omega = 0.3$, $\Delta kL = 2$, and $I_f = I_b$. For an input intensity corresponding to $gI_fL = 2$ the output field amplitude settles to a steady-state value. For an input intensity such that $gI_fL = 3$ the output field amplitude oscillates periodically, as illustrated by the simple closed loop. For an input intensity corresponding to $gI_fL = 5.5$ the output field amplitude still evolves periodically but on an attractor whose period is twice that of the case $gI_fL = 3$. For the case $gI_fL = 15$ the output field amplitude evolves chaotically on a strange attractor whose fractal dimension is 2.2.

that gIL be equal to 15.] We showed in Ref. 5 that the solution of Eqs. (2) is temporally unstable to the growth of amplitude and phase fluctuations, and by means of a linear stability analysis determined the threshold values of the input intensities where this instability occurs. We also showed that for sufficiently large input intensities complicated dynamical behavior including chaotic temporal evolution can occur.

Examples of the dynamical behavior that can occur are illustrated by means of phase-space trajectories in Figs. 1 and 2. To generate these plots, we have solved the set of Eqs. (1) numerically for the case of equal input intensities and have plotted the real part versus the imaginary part of the complex field amplitude of the forward-going wave at its output from the interaction region. Figure 1 shows the case of a narrow Brillouin resonance with $\Gamma/\Omega = 0.03$ and an interaction path length L such that $\Delta kL \equiv 2\Omega L/c = 72$. These parameters correspond to those of our experimental investigation. The threshold for instability in this case, as determined by the procedure outlined in Ref. 5, occurs for $gI_fL = 6.3$. For the case of an input intensity corresponding to $gI_fL = 5$ (i.e., below the instability threshold), the trajectory reduces to a single point. For intensities above the instability threshold, the trajectory takes the form of a nearly circular orbit corresponding to an output wave consisting of a component at the laser frequency and of a Stokes sideband. Note that the amplitude of the Stokes sideband increases with increasing laser intensity.

Figure 2 shows trajectories for a case in which much more complicated dynamical behavior can occur. Here the Brillouin linewidth is much broader ($\Gamma/\Omega = 0.3$), and the interaction length is such that $\Delta kL = 2$. For the case of low input intensities ($gI_fL = 2$), the system is seen to be stable, and the trajectory again reduces to a single point. Slightly above the threshold for instability ($gI_fL = 3$), the output fields oscillate sinusoidally about their steady-state values, and the phase-space trajectory takes the form of a closed loop. For still higher input intensities ($gI_fL = 5.5$), the trajectory is still periodic but with a period equal to twice the fundamental Brillouin period (i.e., the evolution is period 2), and the phase-space trajectory takes on a complicated form. Finally, for the case $gI_fL = 15$ the system evolves chaotically. Analysis using the method of Grassberger and Procaccia[12] shows that this trajectory is chaotic with a fractal dimension of 2.2.

In our previous paper[5] we adopted the criterion that the threshold for instability occurred when the temporal growth rate of a small perturbation (more precisely when the real part of one of the eigenvalues of the growth rate) becomes positive. For comparison with experimental results, a stricter criterion must be adopted, namely, that the growth rate be sufficiently large that the instability will grow to an appreciable amplitude in a time interval comparable with the laser-pulse duration. We quantify this notion by requiring that the product of the amplitude growth rate $\mathrm{Re}(\lambda)$ and the laser-pulse duration τ_p must acquire some nonzero, positive value, which typically would be of the order of 15. Figure 3 shows how the threshold for instability depends on the criterion for the instability threshold used, i.e., the value of $\mathrm{Re}(\lambda)\tau_p$. We note that this experimental threshold is nearly twice as large as the absolute threshold corresponding to a

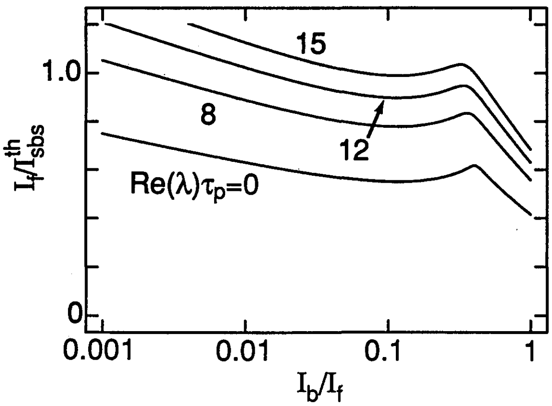

Fig. 3. Theoretically predicted intensity for the threshold of instability plotted as a function of the ratio of intensities of the backward and forward waves, for several different choices of the threshold criterion, quantified by the product of the temporal growth rate $Re(\lambda)$ and the laser-pulse duration τ_p. The results are normalized to the threshold for single-beam SBS, which we take as $I_{SBS}^{th} = 15/gL$.

vanishingly small growth rate [i.e., $Re(\lambda)\tau_p = 0$]. The sharp decrease in the instability threshold for nearly equal pump intensities can be attributed to distributed feedback, which couples the forward and backward Stokes waves. This distributed feedback arises from scattering off the nonlinear-index grating created by the interference of the two strong waves.[5]

Our experimental investigation of the stability of counterpropagating laser beams was conducted using a frequency-doubled Nd:YAG laser that was operated in a single transverse and a single longitudinal mode. The laser produced a smooth output pulse of 22-nsec duration [full width at half-maximum (FWHM) intensity] containing as much as 30 mJ of energy. The output beam was collimated with a diameter of 1 mm (FWHM intensity) and was split into two beams that were directed counterpropagating into a 15-cm-long cell containing carbon disulfide. The intensities of the two beams could be adjusted independently by using polarizing optics, as shown in Fig. 4.

We determine the threshold for instability relative to that for normal, single-beam SBS by means of the following procedure. We block one of the beams and slowly increase the intensity of the other beam (which we will call the forward-going beam) until the threshold for SBS is reached. The occurrence of SBS is signaled by the generation of backscattered Stokes radiation. The measured threshold intensity for single-beam SBS was found to be $I_{SBS}^{th} = 42$ MW/cm.^{2} The other (backward) beam is then unblocked, and the intensities of the two beams are gradually decreased (at fixed intensity ratio) until the Stokes component no longer appears in the output. The intensity of the forward-going beam at which this occurs gives the instability threshold relative to that of SBS. The results of these measurements are shown in Fig. 5. These results show that for the case of equal input intensities the threshold for instability is approximately 67% of that for single-beam SBS. These results also show that the presence of a counterpropagating beam whose intensity is even 10% of that of the pump wave is sufficient to lead to a measurable reduction of the threshold for instability. The solid curve is obtained by numerically integrating the set of Eqs. (1) for our experimental conditions. The laser-pulse shape is modeled as a Gaussian with a FWHM intensity of 22 nsec. We take the threshold for instability to be the point at which the energy of the backward Stokes pulse is 1% of that of the forward pump energy. This method simulates the conditions under which our experimental data were taken. Our measured value of the threshold intensity for single-beam SBS (42 MW/cm^2) is consistent with the usual threshold condition $gIL = 15$ if the value of g is 0.022 cm/MW. This value is smaller by a factor of 0.37 than the value quoted in the literature.[13] It is known, however, that the presence of impurities in carbon disulfide can lead to an increased phonon-damping rate Γ and hence to a decreased value of the Brillouin gain coefficient.[14] We believe that such impurities were present under our experimental conditions. We therefore assume that the ratio (Γ/Ω) is larger by a factor of (1/0.37) than the literature value of 0.012 (which is obtained by scaling the values measured at 0.6943 μm) and hence has the value $\Gamma/\Omega = 0.032$ under our experimental conditions. This value is very close to the value 0.03, which gives the best fit shown in Fig. 5 to our experimental data.

The temporal evolution of the intensity of the light leaving

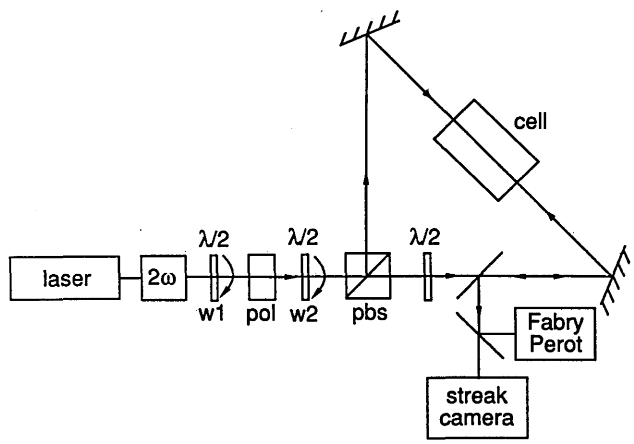

Fig. 4. Experimental arrangement used to study counterpropagating light waves in a cell containing carbon disulfide: w1, w2, half-wave plates; pol, polarizer; pbs, polarizing beam splitter.

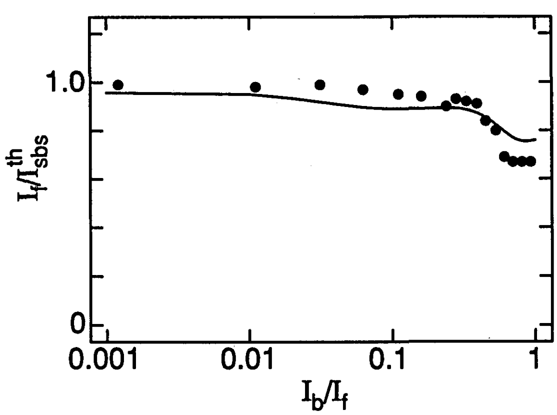

Fig. 5. Experimental measurement of the threshold for instability relative to the threshold for single-beam SBS plotted as a function of the ratio of intensities of the backward and forward waves. The solid curve gives the result of a numerical simulation.

the interaction region was measured with a streak camera having a resolution of 2 psec. Experimental results for the case of equal intensities of the two input beams are shown in Fig. 6. In Fig. 6(a) the input intensity of each beam is equal to 0.66 I_{SBS}^{th}, which is just below the threshold for instability. Figures 6(b) and (c) show the time evolution for input intensities equal to 0.8 I_{SBS}^{th} and 0.87 I_{SBS}^{th}, respectively. These input intensities are above the threshold for instability but well below the threshold for single-beam SBS. Hence the oscillations seen in the experimental data provide evidence for the occurrence of the instability predicted in Ref. 5. In Fig. 6(d) the input intensity is equal to 1.04 I_{SBS}^{th}. The frequency of the observed oscillations is in all cases equal within our measurement accuracy to the value[15] 7.7 GHz, which is the Brillouin frequency of carbon disulfide at 0.53 μm. Using a Fabry–Perot interferometer, we also examined the spectral content of the radiation leaving the interaction region. For all cases in which we observed the instability, radiation was observed only at the laser and the first Stokes frequencies. These results (that the Stokes intensity greatly exceeds the anti-Stokes intensity) agree with the predictions of the theory mentioned above.

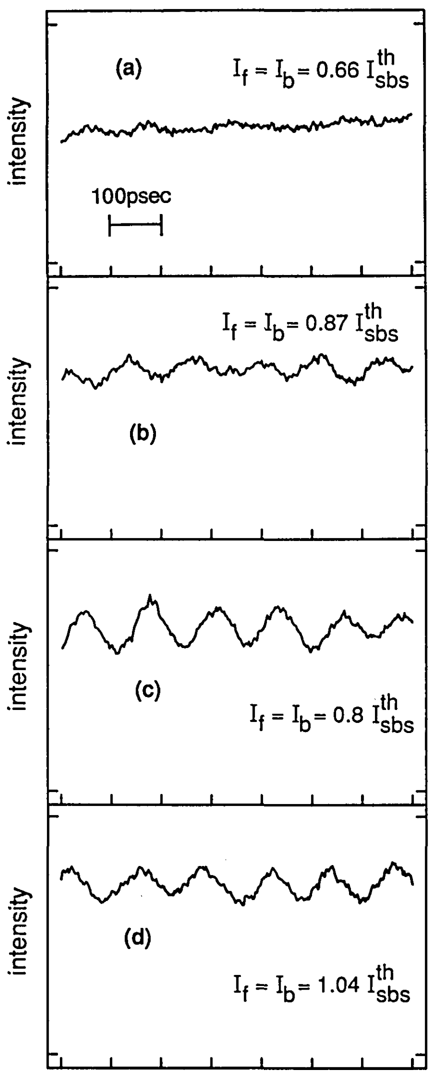

Fig. 6. Experimental data showing temporal evolution of the transmitted intensity of the forward-traveling wave for the case of equal input intensities. Just below the threshold for instability (a), the output intensity is stable. For the remaining three cases (b)–(d) the output oscillates at the Brillouin frequency (7.7 GHz). Note that in (b) and (c) the input intensities are below the threshold for single-beam SBS.

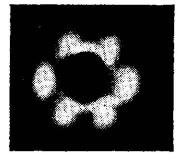

Fig. 7. Hexagonal far-field emission pattern of the Stokes emission. The central portion of the transmitted beam has been blocked.

We also examined the far-field emission pattern of the radiation leaving the interaction region. We find that under conditions of dynamical instability part of the Stokes radiation is emitted in the form of a hexagon surrounding the transmitted laser beam as shown in Fig. 7. The angle between the pump wave and this part of the Stokes radiation is 3×10^{-3} rad. We find that hexagonal emission occurs for $I_f \approx I_b$ and for I_f/I_{SBS}^{th} in the range 0.8 to 1.05. We believe that the origin of this effect is a phase-matched contribution to the nonlinear coupling, as discussed in connection with degenerate four-wave mixing in sodium vapor by Tan-no *et al.*[16] and by Grynberg.[17] The theoretical model of Grynberg assumes the case of degenerate four-wave mixing. However, it is possible that enough Stokes radiation is created along the axis of our system to produce pump waves at the Stokes frequency, which could lead to hexagonal emission according to his model.

In conclusion, we have presented experimental evidence that instabilities occur in counterpropagating laser beams in a Brillouin-active medium. Our experimental results are in good agreement with the theoretical predictions. Our theory also predicts that under certain conditions and for different material parameters chaotic evolution should be possible.

ACKNOWLEDGMENT

The portion of this research conducted at the University of Rochester was supported by National Science Foundation grant ECS-8802761 and by the sponsors of the New York State Center for Advanced Optical Technology.

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