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STIMULATED SCATTERING OF PICOSECOND OPTICAL PULSES IN THE PRESENCE OF SELF-FOCUSING

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We have studied, for various states of polarization for the input beam, the properties of the backward-going wave generated by stimulated light scattering when 25-ps-long laser pulses are focused into CS_2. For the case of a circularly polarized input wave, the backward-going wave is generated by stimulated Rayleigh-wing scattering (SRWS). However, for the case of a linearly polarized input beam, strong filamentation occurs and the backward-going wave appears to be generated by stimulated librational scattering (SLS).

Stimulated scattering (SS) of light often leads to the generation of a backward-going optical wave, and under appropriate circumstances this generated wave has the form of the phase conjugate of the incident wave. Stimulated Brillouin scattering (SBS) is the process most often used to form the phase conjugate wave. However, SBS has a response time of the order of 1 ns and thus cannot be used to form the phase conjugate of picosecond laser pulses. Stimulated Rayleigh wing scattering (SRWS) is the SS process arising from the tendency of anisotropic molecules to become aligned in the electric field of an applied optical wave.[1] SRWS exhibits an extremely fast response time (2 ps in CS_2) and can be efficiently excited using picosecond laser pulses. Also, SRWS is a vector process, that is, it is sensitive to the state of polarization of the incident light (SBS is a purely scalar process, and thus responds only to the intensity of the applied field). These two properties make SRWS an attractive process to be used to form the simultaneous conjugate of both the phase and polarization of an incident field, a process known as vector phase conjugation. SRWS has been predicted theoretically[2] and demonstrated experimentally[3] to return the vector phase conjugate of an applied optical field.

The optical Kerr effect in a liquid of anisotropic molecules leads to an intensity dependent change in the refractive index. As a result of this index change, several effects can occur to alter the spatial and spectral properties of a laser beam propagating through such a medium. Self-focusing is a process in which the nonlinear medium alters the spatial beam profile of an incident field. Filamentation, a related process, causes the input laser beam to break up into several components. Since the intensity within these filaments is usually quite high, several

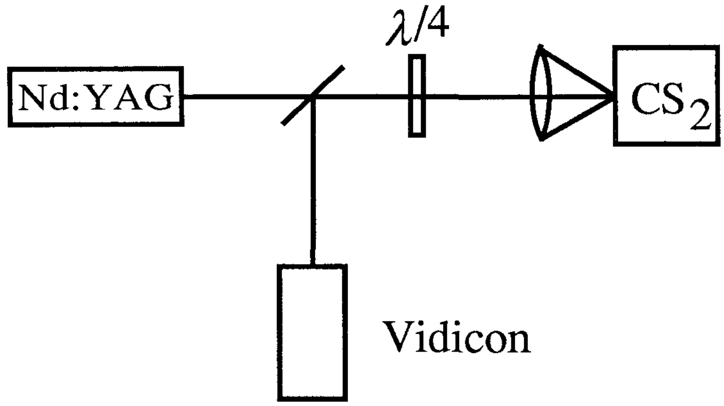

Fig. 1. Experimental setup.

different nonlinear processes can occur simultaneously within the filament to cause dramatic changes in the properties of the optical field. These processes, for example, often degrade the fidelity of the phase conjugation process. It is therefore desirable to eliminate filamentation when building SRWS phase conjugate mirrors. However, the same material parameters that lead to large SRWS gain coefficients also lead to large nonlinear refractive indices. Therefore, little is to be gained by choosing different nonlinear materials to be utilized in an SRWS generator. Instead, one must choose experimental conditions that allow SRWS rather than filamentation to occur.

In this paper, we present the results of an experimental investigation of the optical properties of the backward-going wave generated by stimulated scattering when excited by picosecond optical pulses. We find that the optical properties of the generated wave depend strongly on the state of polarization of the input wave. We feel that this behavior is the result of competition among self-focusing, SRWS and stimulated librational scattering (SLS).

Our experimental arrangement is shown in Fig. 1. The frequency-doubled output of a mode-locked and Q-switched Nd:YAG laser was focused into a 10-cm-long cell containing CS_2. The laser pulses had a wavelength of $0.53\,\mu$m and were 25 ps in duration. The laser pulse energy was in the range of $5–50\,\mu$J. When the input laser intensity exceeded the threshold value, a wave counterpropagating to the input beam was generated. This wave was then sampled by means of a 50/50 beam splitter. The polarization state of the input laser beam was controlled by a quarter-wave plate. The spatial beam profile of the return signal was recorded with a vidicon camera interfaced to a frame grabber and laboratory computer. The plane just after the sampling beam splitter was imaged onto the vidicon. The spectrum of the return signal was recorded using a 0.75-m-spectrometer interfaced to an optical multichannel analyzer giving a spectral resolution of 0.01 nm.

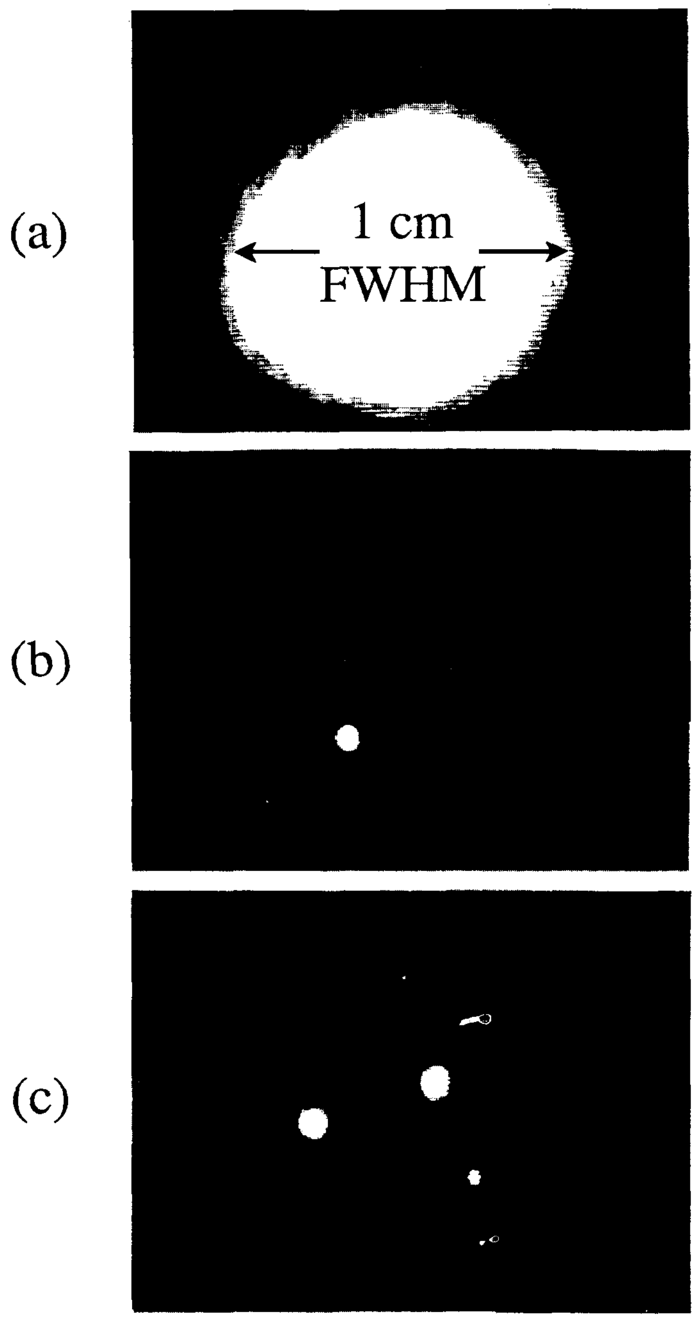

Fig. 2. Spatial intensity distributions. The linearly polarized input beam is shown in (a) while the generated backward-going wave is shown in (b) for a input intensity equal to the threshold value and in (c) for an input intensity approximately twice as large.

When the input laser beam was circularly polarized, the spatial profile of the return signal closely resembled that of the input. The return signal was also circularly polarized but with the opposite sense of rotation. These observations are consistent with the theoretical prediction that SRWS returns the vector phase conjugate of the input wave. Such a return wave can be used to remove distortions in both the phase and polarization of the incident field. Details of an experimental demonstration of this property of SRWS have been reported previously.[3] However, when the input laser was linearly polarized, the return beam did not resemble the input. Rather, the spatial profile of the backward-going beam was composed of several filaments (see Fig. 2). Although the diameter of each filament remained constant, the position of each filament varied from shot to shot. Also, the number of filaments depended strongly on the input laser power. Figure 2 shows the spatial profile of the backward-going wave generated by a linearly polarized input wave for various input laser pulse energies. We note strong filamentation with the number of filaments being approximately equal to the ratio of input power to threshold power. In our experimental geometry, the threshold power for linear polarization was approximately equal to 100 kW. Hence, we see that linear polarization does not lend itself well to phase conjugation applications.

The origin of the different behavior that is observed through the use of light of different polarizations can be understood by considering the polarization dependence of the nonlinear refractive index and the SRWS gain coefficient. The polarization dependence of the optical Kerr effect has been studied by Close et al.[4] By considering the nonlinear medium to be composed of an ensemble of anisotropic molecules characterized by the two principal polarizabilities α_1 and α_2, the average polarizability induced by an applied electric field is given by

$$\langle \alpha_{ij} \rangle = \alpha \delta_{ij} + \gamma_{ij} , \tag{1a}$$

where

$$\gamma_{ij} = \frac{(\alpha_1 - \alpha_2)^2}{45kT} \sum_{kl} [3\delta_{ik}\,\delta_{ij} - \delta_{ij}\,\delta_{kl}] \langle F_k F_l \rangle , \tag{1b}$$

and $\alpha = 1/3\ \alpha_1 + 2/3\ \alpha_2$ is the average polarizability, F_i is the local field within the medium, and < > denotes an average over many optical cycles. If we assume that the applied field is elliptically polarized with an ellipticity ε ($\varepsilon = 0$ for linearly polarized light while $\varepsilon = 1$ for circularly polarized light), it is straightforward to show that the nonlinear coefficient of the refractive index is

$$n_2 = \frac{4\pi N (\alpha_1 - \alpha_2)^2}{90kTn_0} \left(\frac{2 - \varepsilon^2}{1 + \varepsilon^2} \right) . \tag{2}$$

(Of course, elliptically polarized light simultaneously experiences self-induced ellipse rotation. However, this effect does not affect the conclusions of our calculation.) Here, N represents the number density of molecules and n_0 is the linear refractive index of the bulk material. Due to the fact that the nonlinear refractive index coefficient for a linearly polarized wave is four times larger than that of a circularly polarized wave [from Eq. (2)], a circularly polarized input beam experiences self-focusing less readily than a linearly polarized input beam. In particular, filamentation can occur only when the input laser power exceeds the critical value given by[4]

$$P_{\text{cr}} = \frac{(0.61)^2 \lambda^2 c}{n_2} . \tag{3}$$

Next, we determine the threshold power required to excite SRWS. If we assume that the single pass gain coefficient $gIL = gQ/2\lambda T$ (where Q is the pulse energy and T is the pulse width of the exciting laser pulse) must exceed a threshold value of 25, the SRWS threshold power is then given by

$$P_{\text{th}} = \frac{50\lambda}{g_{rw}(\varepsilon)} , \tag{4}$$

where $g_{rw}(\varepsilon)$ is the SRWS gain coefficient. The polarization dependence of $g_{rw}(\varepsilon)$ was studied theoretically by Chiao and Godine[1] and is given by

$$g_{rw}(\varepsilon) = -2IM\left[g_0\left\{-1 + \frac{7D}{6} + \left[1 + \frac{D}{3}\left(1 + \frac{16\varepsilon^2}{(1+\varepsilon^2)^2}\right) + \frac{D^2}{36}\left(1 + \frac{96\varepsilon^2}{(1+\varepsilon^2)^2}\right)^{1/2}\right]\right\}\right], \tag{5}$$

where $D = (1 + i\Omega\tau)^{-1}$ is the molecular response function. Equation (5) shows that the peak SRWS gain occurs at a Stokes shifted frequency $\Omega = 1/\tau$ for both circularly and linearly polarized light. Also, the SRWS gain coefficient is larger for a circularly polarized input than for a linearly polarized input by a factor of 3/2.

To examine the interaction between self-focusing and SRWS, we next define a dynamic range parameter $R = (P_{\text{cr}} - P_{\text{th}})/P_{\text{th}}$. This parameter can be interpreted in the following manner: For $R > 0$, the threshold power for SRWS exceeds that required for filamentation, and the input laser beam tends to break into filaments before SRWS is excited. For $R < 0$, the opposite is true, and the SRWS threshold in the backward direction is reached before filamentation occurs. The magnitude of R (for $R > 0$) gives an indication of the extent to which the input power can exceed the

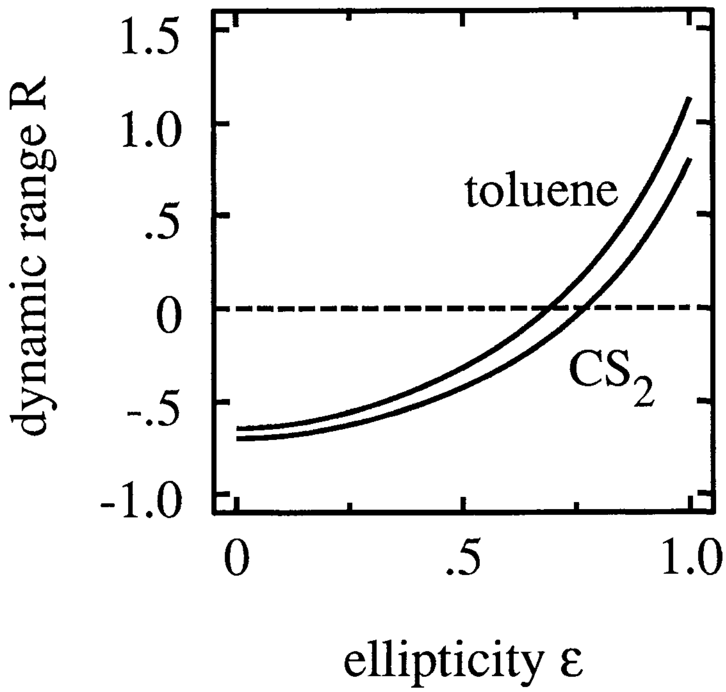

Fig. 3. Dynamic range parameter R as a function of the input laser ellipticity ε. For $R<0$, self-focusing occurs before SRWS while for $R>0$, SRWS occurs before self-focusing.

SRWS threshold power before self-focusing effects become important. Equations (1)–(5) can be combined to find that the polarization dependence of R is

$$R = Mg_{rw}(\varepsilon)\left(\frac{1+\varepsilon^2}{2-\varepsilon^2}\right) - 1, \tag{6}$$

where $M=P_{cr}^{(\text{linear})}/P_{th}^{(\text{circular})}$ is the ratio of the critical power for self-focusing of linearly polarized light to the threshold power for SRWS using circularly polarized light, and depends only on material parameters ($M=0.45$ for CS_2). The relation between R and the input laser ellipticity is shown in Fig. 3 for two different materials, CS_2 and toluene. Here, we find that for the case of a linear polarized input, R is less than 0, and filamentation is expected to be observed in the output beam profile. However, for the case of a circularly polarized input, R is greater than 0. Thus, backward SRWS is expected to generate a wave that preserves the spatial beam profile of the input wave. These predictions are in agreement with our experimental results.

We have also found that when strong filamentation of the backward-going wave is observed, the backward-going beam does not appear to be generated by SRWS. Since SRWS in the backward direction exhibits a characteristic frequency shift ($\Omega=1/\tau$) to the Stokes side of the laser frequency, the spectrum of the return signal

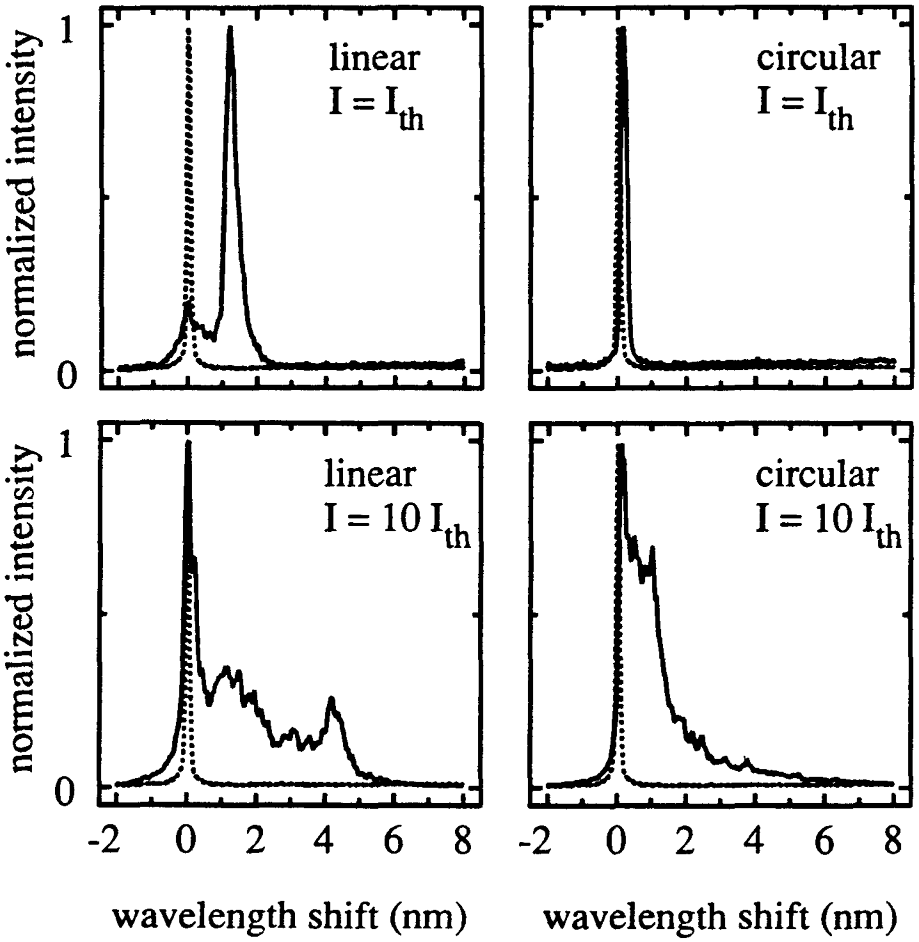

Fig. 4. Spectrum of the generated backward-going wave for different states of polarization and intensity of the input laser beam. The dashed line shows the spectrum of the input laser beam, while the solid line shows the spectrum of the generated beam.

is a unique experimental signature. Figure 4 shows the spectrum of the return signal for two different pulse energies, for both a circularly and linearly polarized input. For the case of a circularly polarized input, the spectrum is shifted to the Stokes side of the laser wavelength by 0.05 nm, indicating that SRWS is the dominant process leading to the generation of the return signal. For higher input pulse energies, the spectrum continues to broaden to the Stokes side of the laser frequency. However, for the case of a linearly polarized input wave, the spectrum is quite different. Near threshold, the return signal does not exhibit the SRWS frequency shift. For higher input laser powers, we detect a significant broadening to the Stokes side of the laser frequency and a small but measurable broadening to the antiStokes side. These results show that when filamentation is present in our system, a different nonlinear mechanism leads to the generation of the backward-going wave.

Near threshold, stimulated librational scattering (SLS) appears to be the dominant process leading to the generation of the backward-going wave. When anisotropic molecules are excited with an applied electric field, the molecules undergo a librational or rocking motion.[5–8] This molecular motion leads to a field-induced susceptibility change in accordance with the equation

$$\tau_2^2 \frac{\partial^2 \triangle \chi_{ik}}{\partial t^2} + \tau_1 \frac{\partial \triangle \chi_{ik}}{\partial t} + \triangle \chi_{ik} = \kappa (E_i E_k - \frac{1}{3} \delta_{ik} E_j E_j^*) , \qquad (7)$$

where $\triangle \chi$ is the field-induced nonlinear susceptibility change, and κ is the nonlinear coupling constant. As a result of this nonlinearity, the backward-going wave will experience gain at a frequency Stokes-shifted by $\Omega = 1/\tau_2$ from the laser frequency. In CS_2, the frequency shift is expected to be 1.8 nm,[8] in reasonable agreement with our experimental observations.

Since the steady-state gain coefficient for SLS is smaller than the steady-state gain coefficient for SRWS, it is somewhat surprising that SLS is observed in the backward direction. We suspect that the suppression of SRWS when excited by a linearly polarized wave is related to the degree of saturation (i.e. the degree of alignment of the molecules within the field) of the molecular orientation process. Strong saturation most likely occurs within the self-focused filaments. With the molecules no longer free to rotate, the SRWS gain is dramatically reduced, so that lower gain processes can be observed. It is unclear what degree of saturation is required to inhibit SRWS. Also, when filamentation occurs, the laser creates a laser-induced waveguide. Within the filament, the laser pulse propagates through a long interaction region while maintaining a high laser intensity. Therefore, large values of the single-pass gain, gIL, are obtained.

At higher input pulse energies, a significant spectral broadening occurs in the backward-going wave (see Fig. 4). These results are similar to those observed by He and Prasad for stimulated Rayleigh–Kerr scattering.[9, 10] They studied the spectral broadening of a laser pulse in the forward direction, upon propagation through a waveguide in the filled with CS_2. They proposed a new mechanism, stimulated Rayleigh–Kerr scattering which is the stimulated scattering process resulting from large field-induced rotations of anisotropic molecules (i.e. the saturated limit of SRWS) and found an empirical relationship to best fit their data.

In conclusion, we have studied the polarization dependence of the backward-going wave generated by SS when excited by picosecond optical pulses. For the case of a circularly polarized input wave, the return wave displayed the properties of SRWS. However, when the backward-going wave was excited by a linearly polarized wave, filamentation was present, and the return signal was not generated by SRWS. Stimulated librational scattering within the self-trapped filaments appears to be the origin of the backward-going wave.

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