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Raman-assisted spatial cross phase modulation in carbon disulfide

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We report on the observation of spatial modifications of a weak beam copropagating with a strong beam due to Raman-assisted cross phase modulation. This effect, due to the interference between nonlinear polarizations associated with the third-order susceptibility, is enhanced when the difference between the frequencies of the incident beams is resonant with the frequency of a vibrational mode of CS_2. In principle, the phenomenon may be observed in a large variety of materials and can be exploited in applications such as spatial soliton switching.

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The study of nonlinear optical transverse effects such as spatial self-phase modulation (SPM) of a laser beam has been a subject of active research in the last three decades [1–8]. Among the various phenomena related to SPM are self-trapping and filamentation [1,2], light bending [3], generation of spatial solitons [4], and pattern formation [5]. In principle, all kind of media may exhibit SPM and a large amount of results have been reported for different systems such as atomic vapors [5], solids [6], liquid crystals [7], and photorefractive crystals [8]. Theoretically, these phenomena may be described by the wave equation which governs the propagation of a laser beam inside a nonlinear medium. The effect of the nonlinearity is included through an intensity-induced change in the sample's refractive index and/or an intensity-dependent absorption.

In general, the previous studies on transverse spatial effects are related to phenomena governed by the frequency degenerate third-order nonlinear susceptibility $\chi^{(3)}$ and effects due to the nondegenerate susceptibility have not received much attention. However, the investigation of such effects is worthwhile because its knowledge may allow beam profile manipulations in a variety of situations of fundamental interest [9] and as well as dual-wavelength all-optical spatial switching applications [10].

In this paper we report changes in the spatial profile of an optical beam induced by another beam due to Raman-assisted cross phase modulation. The effect is enhanced when the difference among the frequencies of the two incident beams is in resonance with the frequency of a vibrational Raman transition of carbon disulfide.

The two-color Z-scan technique [11] was used to characterize the phenomenon. This technique is based on the spatial cross phase modulation induced in a probe laser beam (frequency ω_1 and wave number k_1) by a pump laser (frequency ω_2 and wave number k_2) when they propagate collinearly in a nonlinear medium. As in the conventional Z scan [12], the two-color Z-scan technique is based on the transformation of the probe beam wave front during beam propagation through a nonlinear sample, to amplitude distortion of the beam through a small aperture placed in the far-field region. The modulus and sign of $\mathrm{Re}\chi^{(3)}(\omega_1,\omega_2,-\omega_2,\omega_1)$ and $\mathrm{Im}\chi^{(3)}(\omega_1,\omega_2,-\omega_2,\omega_1)$ may be obtained by analyzing the transmitted beam intensity as a function of the sample distance from the focusing lens.

The present experiments were performed using two dye lasers which consist of an oscillator plus one stage of amplification, transversely pumped by the second harmonic of a pulsed (5 Hz) neodymium-doped yttrium aluminum garnet (Nd:YAG) laser, delivering pulses of 9 ns and 100 kW peak power. The oscillators were operated with a grazing incidence grating in the range of 560–640 nm with linewidths $\leq 0.02\ \mathrm{cm}^{-1}$.

The two incident beams with parallel polarizations were focused onto a cell (10 mm length) containing spectroscopic-grade CS_2. The sample was placed along the propagation direction of the laser beams (the Z axis) and the spatial changes in the probe laser transmission was recorded for different Z positions and laser frequencies. Caution was taken to make the pump and the probe beams focus on the same point. The measured radius of the pump beam at the focus was $w_2=92\ \mu\mathrm{m}$ and the radius of the probe beam, w_1, was adjusted to be 65 or 92 μm. The probe beam intensity was weak enough and did not induce detectable nonlinear phase change but was much stronger than the Raman signal excited by the pump beam. On the other hand, the peak power of the incident pump beam (40 kW) was strong enough to induce a detectable cross phase modulation. Under these excitation conditions, the profile intensity change of the probe beam at the far-field region was clearly visible, which made the adjustment of the beams' overlap and the measurements quite easy. The probe frequency was fixed at $16\,920\ \mathrm{cm}^{-1}$ and the pump frequency was tuned in such a way that $\omega_2-\omega_1$ was set to different values close to resonance with the vibrational mode of CS_2 ($\omega_R=655.7\ \mathrm{cm}^{-1}$). A photodiode was placed at the center of the probe beam profile 100 cm behind the position $Z=0$. The aperture in front of the photodiode had a diameter of 2 mm.

Z-scan measurements were carried out for different values of the frequency detuning $\Delta=\omega_2-\omega_1-\omega_R$. The transmittance signal collected by a lens positioned close to the sample is almost flat indicating that nonlinear absorption, such as two-photon absorption, is negligible and

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thus we consider that $\mathrm{Im}\chi^{(3)}(\omega_1,\omega_2,-\omega_2,\omega_1)\approx 0$ in agreement with previous measurements [13]. The small aperture two-color Z-scan transmittances for two values of Δ are shown in Figs. 1(a) and 1(b). In the case where $\Delta=3000\ \mathrm{cm}^{-1}$, Fig. 1(a) shows a signal profile with a valley followed by a peak which indicates an effective positive real nonlinearity (induced focusing). The Z ordinate is measured along the beams' direction and $Z<0$ corresponds to locations of the sample between the focusing lens and its focal plane. The experiments were performed with $a=k_2w_2^2/k_1w_1^2=1$ and 2, and a larger sensitivity was observed when $a=2$. This means that when the radius of the pump beam at the focus is fixed, the more tightly the probe beam is focused, larger probe transmission changes can be obtained, in accordance with the theoretical results presents in Ref. [11(b)]. Figure 1(b) shows the two-color Z-scan signal for zero detuning $(\omega_2-\omega_1=\omega_R)$. In this case a large change in the Z-scan profile is observed and the result mimics the case where nonlinear absorption occurs [12]. However, in the present case, as in Fig. 1(a), $\hbar(\omega_1+\omega_2)$, $2\hbar\omega_1$, and $2\hbar\omega_2$ are off resonance with excited states and two-photon absorption is negligible [13]. The strong change in the probe beam wave front is attributed to the Raman contribution and the theoretical curve traced with parameters previously obtained for CS_2 [12,14] shows good agreement with the experiment, as discussed below.

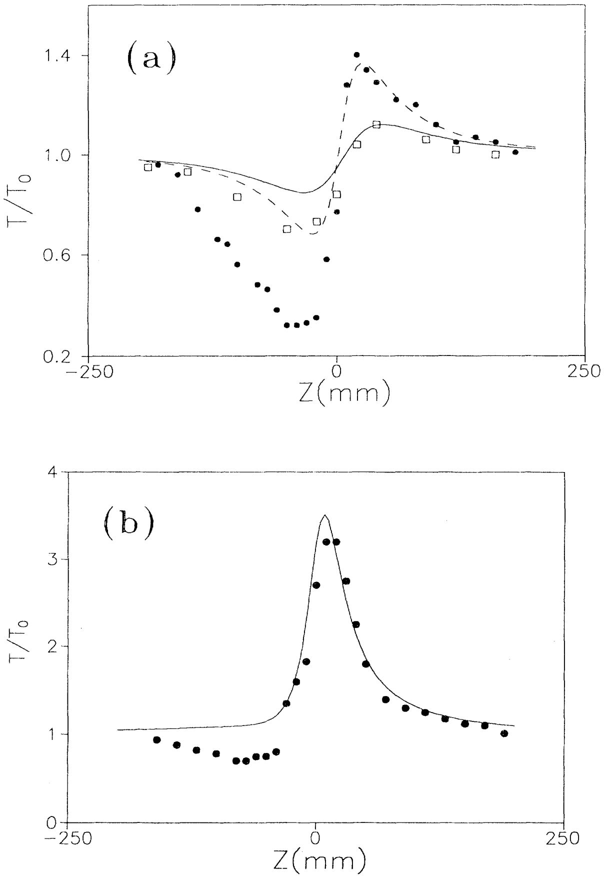

FIG. 1. (a) Normalized transmission for $\Delta=3000\ \mathrm{cm}^{-1}$ as a function of Z with $a=1$ and 2. T_0 is the transmission of the probe beam when the pump laser is blocked. The dots and the dashed curve illustrate experimental and theoretical results, respectively, for $a=1$. The open squares and the solid curve correspond to the experimental and theoretical results, respectively, when $a=2$. (b) Normalized transmission as a function of Z when $\Delta=0$ and $a=2$.

The enhancement of the cross-phase-modulation effect due to the Raman resonance is clearly demonstrated in Fig. 2 where the intensity transmission change is shown as a function of the Raman detuning. For this measurement the sample was positioned at $Z=32$ mm and the frequency of the pump laser was varied while the probe-laser frequency was fixed.

The normalized transmitted intensity (T/T_0) is calculated following the usual procedure [11,12]. First, we consider that the probe field is proportional to $\exp[-i\Delta\phi(z,r,t)]$ at the exit surface of the sample, where $\Delta\phi$ is the phase variation of the probe due to the pump laser. The far-field intensity distribution is then calculated using diffraction theory [15] and the expression obtained for T/T_0 includes the laser beam spatial parameters and pulse widths. The influence of the Raman resonance is included in the calculations through $\Delta\phi(r,z,t)$ which is a complex quantity and contains contributions from the real part of $\chi^{(3)}(\omega_1,\omega_2,-\omega_2,\omega_1)$ as well as from its imaginary part which originates from the Raman dephasing relaxation.

The solid lines in Figs. 1 and 2 illustrate the theoretical results obtained assuming that the main nonlinear contributions are due to the Kerr and Raman effects. Therefore, the optical nonlinearity was expressed by

$$\chi^{(3)}(\omega_1,\omega_2,-\omega_2,\omega_1)=\chi_{\mathrm{NR}}^{(3)}+\frac{R}{\omega_R-(\omega_2-\omega_1)+i\Gamma_R}\,, \tag{1}$$

where the value of the nonresonant part $\chi_{\mathrm{NR}}^{(3)}$ is obtained from the measurements of the nonlinear refractive index [12], and $\chi_{\mathrm{NR}}^{(3)}/R=4.36$ cm and $\Gamma_R=0.2\ \mathrm{cm}^{-1}$ as determined in Ref. [14].

The fact that the amplitude of the valley signal in Fig. 1(a) is larger than the peak signal indicates that other mechanisms are also contributing to the results. Notice

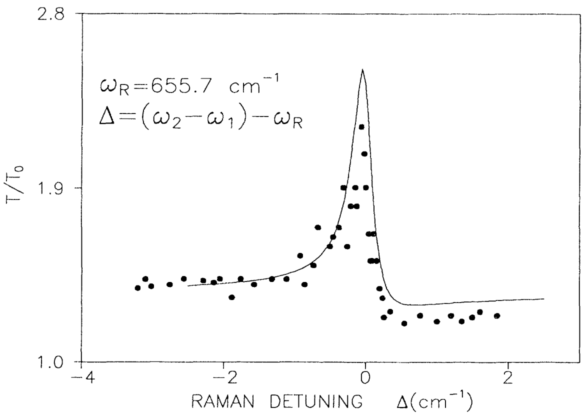

FIG. 2. Normalized transmission for $a=2$ as a function of the Raman detuning when the sample is placed at $Z=32$ mm.

that this experiment was performed with a large Raman detuning ($\Delta = 3000$ cm^{-1}) and thus the Raman contribution to the Z-scan profile is negligible. A qualitative understanding of the results may be attained in the following way. First we note that the strong pump beam may stimulate density waves due to the large electrostrictive effect in CS_2 [2]. In fact, when very high intensities are used, a characteristic noise can be heard as originating from the CS_2 cell and the incident beams may be scattered by the density waves along various directions. Although the results of Fig. 1(a) have been obtained using not very high intensities, the probe beam is scattered due to the effect of the pump beam, and the intensity passing through the aperture, placed in the far-field region, decreases. This process is particularly important when the sample is in the $Z<0$ region because the scattering phenomenon contributes in the same way as the Kerr and the electrostrictive nonlinearities, and so a deeper valley signal is observed. At the $Z>0$ region the collimation of the probe beam due to the positive electrostrictive nonlinearity is compensated by the scattering effect. Thus the Z-scan signal at the $Z>0$ region is less affected than for $Z<0$ and the Kerr nonlinearity dominates the results. It was also noted that the electrostrictive and scattering contributions are power dependent. When high pump powers are used the Z-scan peak decreases and the valley becomes deeper. On the other hand, if low powers are used, the agreement between the experimental and the theoretical results based on Eq. (1) is improved. In the cases illustrated by Figs. 1(b) and 2 the frequency detuning Δ is such that the Raman contribution is enhanced. The good agreement between the experimental results and the theory shows that the Kerr and the resonant Raman susceptibilities provide the main contributions for the cross-phase-modulation process in these cases.

Finally, we emphasize that the transverse nonlinear phenomenon presented here is quite general and can be observed in any material with allowed Raman transitions. In particular, for CS_2 where fundamental studies of spatial solitons have been performed [16], we expect that exploitation of the Raman-assisted cross-phase-modulation effect will open new possibilities to control spatial soliton trapping and switching. Moreover, this variation of the Z-scan technique provides a nonlinear method for spectroscopical studies of Raman resonances in condensed matter.

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