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Experimental and numerical study on chirped transient stimulated Raman scattering in dispersive medium

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ABSTRACT

Experimental and numerical study on chirped transient stimulated Raman scattering in ethanol have been performed. Negatively chirped pump pulses yielded much higher conversion efficiency than positively chirped pump pulses. The calculated results reveal that the group velocity mismatch between the pump and Stokes pulses can rapidly degrade the conversion efficiency when the pump laser generates chirped pulses. However, the combined effects of group velocity dispersion and self-phase modulation offset the negative chirps during propagation, yielding higher conversion efficiency. This is a novel mechanism for controlling transient stimulated Raman scattering and is likely to be of importance for various applications.

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1. Introduction

Stimulated Raman scattering (SRS) has been studied experimentally and theoretically since the invention of laser. In addition to investigations of nonlinear phenomena, SRS has been used in spectroscopy [1], development of novel frequency lasers [2,3], beam cleanup and combination [4], pulse amplification and compression [5], and soliton generation [6]. Much attention has been devoted to applying the SRS for generating high temporal contrast near 1 μm wavelength seed pulses for high-peak power chirped pulse amplification (CPA) laser systems [7]. Results in [8] reveal that, in CPA, the final amplified pulse contrast remains high if the seed pulse has high temporal contrast with high energy. To obtain a high-peak power, high-temporal contrast pulse, a high-temporal contrast seed pulse with high energy should first be obtained. Moreover, obtaining high SRS conversion efficiency is a key issue in many applications. Thus, conversion efficiency of SRS should be investigated experimentally and theoretically.

In the applications for generating high temporal contrast seed pulses, the pump pulse duration is ultra-short (sub-picosecond scale). When the pump pulse duration is on the order of or less than the dephasing time of molecular vibrations, T_2, SRS becomes transient. Numerous studies have investigated the transient SRS (TSRS) analytically and numerically [9–18]. In the transient regime, the threshold gain is determined by the pump energy rather than by the peak intensity, as in the case of stationary SRS. In addition, the self-phase modulation (SPM) effect becomes obvious and can decrease the conversion efficiency [9,15]. To decrease the influence of SPM and improve the conversion efficiency, chirps can be added to pump pulses [19]. TSRS modeling neglecting dispersion has shown that the phase difference between the pump, Stokes, and phonon waves quickly locks to $\pi/2$ once the Stokes field intensity has grown to roughly one order of magnitude above the initial Stokes seed intensity [10,12,13]. After the phase difference had locked to $\pi/2$, the Stokes field intensity starts to grow rapidly. However, TSRS has been mainly experimentally and theoretically studied in compressed gases, which exhibit low dispersion, pumped by picosecond-scale pulses without or with low-level chirps. In these conditions, the influence of dispersion and chirps on conversion efficiency is not significant.

Ethanol is a promising medium that is used in SRS because it has relatively large gain coefficient (5 cm/GW [20]). Moreover, its frequency shift is 2928 cm^{-1} [20], and an 800 nm pump laser can be used for directly generating nearly 1 μm high temporal contrast laser beam. However, for TSRS in ethanol (which exhibits much greater dispersion) pumped by a chirped pulse, the influence of dispersion and chirps on conversion efficiency becomes important. This paper presents experimental and numerical studies on the conversion efficiency of a first-order forward Stokes component in sub-picosecond regime chirped pulse TSRS in ethanol. The

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calculated results reveal that the effect of group velocity mismatch (GVM) can rapidly degrade the conversion efficiency when the pump laser generates chirped pulses. However, considering the combined effects of group velocity dispersion (GVD) and SPM, which can offset the chirps during propagation if the pump laser generates negatively chirped pulses, can yield much higher conversion efficiency. This behavior is a novel phenomenon and is experimentally demonstrated in the present study. The study conclusions are likely to be of importance for many applications.

2. Theoretical model

The analysis was based on a set of coupled equations for slowly varying envelopes of the pump and Stokes fields, A_p and A_s, and the vibrational coordinate Q, which represents the envelope of an optical phonon wave. Given that the pump pulse is ultra-short and is generated by an ultra-intense laser, the influence of GVM between the pump and Stokes pulses, GVD, SPM, cross-phase modulation (XPM), and the transient effect on the SRS process should be considered for better characterization of the SRS. The equations are in the coordinate system that moves with the pump group velocity [16,17]:

$$\frac{\partial A_p}{\partial z} = -\frac{\alpha_p}{2}A_p - i\frac{\beta_{2p}}{2}\frac{\partial^2 A_p}{\partial T^2} + i\gamma_p(|A_p|^2 + 2|A_s|^2)A_p - i\kappa_p A_s Q,$$
$$\frac{\partial A_s}{\partial z} = -\frac{\alpha_s}{2}A_s + \delta_{ps}\frac{\partial A_s}{\partial T} - i\frac{\beta_{2s}}{2}\frac{\partial^2 A_s}{\partial T^2} + i\gamma_s(|A_s|^2 + 2|A_p|^2)A_s - i\kappa_s A_p Q^*$$
$$\frac{\partial Q}{\partial z} = -\frac{Q}{T_2} - i\kappa_q A_p{}^* A_s, \quad (1)$$

where the subscripts p, s, and q refer to the pump, Stokes, and optical phonon waves, respectively. The parameter α is the absorption coefficient and $\delta_{ps}=1/v_{gp}-1/v_{gs}$ is the GVM coefficient. The parameter v_g is the group velocity and β_2 is the GVD coefficient. The parameter γ is the nonlinear coefficient, corresponding to SPM and XPM. The time T_2 is the vibrational dephasing time. The quantities κ represent the coefficients in nonlinear terms that describe TSRS. We set $\kappa_p = 2\pi N\omega_p c^{-1}n_p^{-1}\partial\alpha/\partial Q$, $\kappa_s = 2\pi N\omega_s c^{-1}n_s^{-1}\partial\alpha/\partial Q$, and $\kappa_q = 2^{-1}N\omega_q^{-1}\partial\alpha/\partial Q$, where N is the number density of Raman active species, $\partial\alpha/\partial Q$ is the cross-section of Raman scattering, ω is the field's angular frequency, n is the medium's refractive index, and c is the speed of light in vacuum.

In addition,

$$\kappa_s\kappa_q T_2 = \frac{g_s}{Area}, \frac{\kappa_s}{\kappa_q} = \frac{4\pi\omega_s\omega_q}{cn_s}, \gamma = \frac{n_2\omega}{cArea} \quad (2)$$

where g_s refers to the steady state gain coefficient. Area is the pump beam area, and n_2 is the nonlinear refractive index.

Eq. (1) are nonlinear and complicated. The numerical methods were extensively used to study their solutions. The temporal histories of the pump and the Stokes pulses are known because they enter the Raman cell at $z=0$. The pulses subsequent evolution, as they propagate toward large z, is determined by the medium. The medium's excitation is zero before the arrival of the pump and Stokes pulses at each point z. In Eq. (1), the time variable T is not a physical time but is shifted by z/v_{gp} at every point z. Mathematically, the first two formulae in Eq. (1) are evolution equations that can be solved by using split step Fourier method [21]. The third formula in Eq. (1) is a constraint equation and it can be solved by applying Simpson's rule.

In the experiment, the temporal duration of the compressed pump pulse was 35 fs (full width at half maximum, FWHM). Positive or negative chirps were introduced to broaden the pulses to proper durations. In the simulation, the pump pulses were assumed to be of Gaussian shape, as follows:

$$A_p = \sqrt{P}\exp\left[-\frac{(1+iC)}{2}\frac{T^2}{T_0^2}\right] \quad (3)$$

Where P is the peak power, T_0 is the pulse's temporal duration (full width at $1/e^2$), and $T_{fwhm}=1.665T_0$. The parameter C is the chirp parameter, calculated as:

$$C = \sqrt{\left(\frac{T_{chirp}}{T_{ftl}}\right)^2 - 1} \quad (4)$$

where T_{chirp} is the chirped pulse duration (FWHM) and T_{ftl} is the Fourier-transformed limit pulse duration (FWHM).

The pump spectra are shown in Fig. 1. The red and black solid lines denote the pump spectra for the experimental and simulation inputs, respectively. For simplicity, the temporal shape of the Stokes pulse is assumed to be the same as that of the pump pulse, but the wavelength is centered at 1044 nm and the initial energy is 10^{-5} pJ.

Table 1 shows the calculation's parameters.

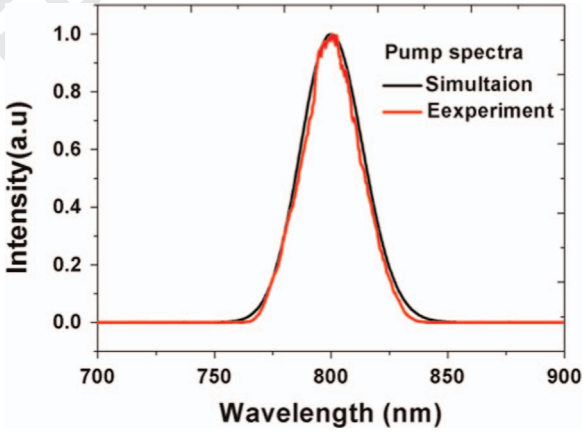

Fig. 1. Pump spectra. The red and black solid lines denote the spectra for the experimental and simulation inputs, respectively. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

Table 1
Calculation parameters.

Parameters	Value	Parameters	Value
α_s [20] (cm^{-1})	0.0164	n_2 [22] (10^{-16} cm^2/W)	7.7
α_p [20] (cm^{-1})	0.0164	T_2 (ps)	1
β_{2s} (fs^2/cm)	317	g_s [20] (cm/GW)	5
β_{2p} (fs^2/cm)	420	Area (cm^2)	0.35
δ_{ps} (fs/cm)	202.7	L (cm)	8

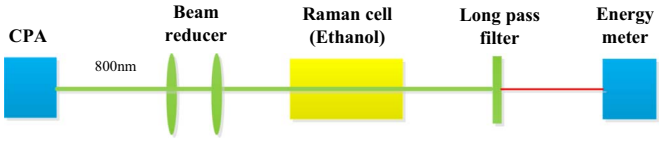

Fig. 2. Schematic of the experimental setup for SRS.

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