ARTICLE

Effect of Thermal Defocusing on Backward Stimulated Raman Scattering in CH$_4$

Xiao-qing Hua, Jing Leng, He-ping Yang, Guo-he Sha*, Cun-hao Zhang

State Key Laboratory of Molecular Reaction Dynamics, Dalian Institute of Chemical Physics, Chinese Academy of Sciences, Dalian 116023, China

The conversion efficiency of stimulated Raman scattering (SRS) in CH$_4$ is studied by using a single longitudinal mode second-harmonic Nd:YAG laser (532 nm, linewidth 0.003 cm^{-1}, pulse-width (FWHM) 6.5 ns). Due to the heat release from vibrationally excited particles, SRS processes often suffer from the thermal defocusing effect (TDE). In view of 6.5 ns laser pulse width is much shorter than the vibrational relaxation time of CH$_4$ molecules, TDE can only affect the SRS processes afterwards. In the cases of low laser repetition, TDE will be not serious, because it will be removed by the thermal diffusion in Raman medium before the next pulse arrives. At the laser repetition rate 2 Hz, CH$_4$ pressure 1.1 MPa and pump laser energy 95 mJ, the quantum conversion efficiency of backward first-Stokes (BS1) has attained 73%. This represents the highest first-stokes conversion efficiency in CH$_4$. Furthermore, due to the relaxation oscillation, the BS1 pulses are narrowed to about 1.2 ns. As a result, the BS1 peak power turns out to be 2.7 times that of the pump. Its beam quality is also much better and is only slightly affected by TDE. This reason is that BS1 represents a wave-front-reversed replica of the pump beam, which can compensate the thermal distortions in Raman amplify process. Under the same conditions, but pump laser repetition rate as 10 Hz, the conversion efficiency of BS1 goes down to 36% due to TDE. From this study, we expect that a well-behaved 630 nm Raman laser may be designed by using a closed CH$_4$/He circulating-cooling system, which may have some important applications.

Key words: Nd:YAG laser, Stimulated Raman scattering, Thermal defocusing effect, CH$_4$ gas

I. INTRODUCTION

It is well known that in the stimulated Raman scattering (SRS) processes, the laser beam often will suffer from the thermal defocusing effect (TDE) due to the heat release from vibrationally excited molecules [1,2]. However, since the laser pulse-width (a few ns) is usually much shorter than that of the vibrational relaxation time, the TDE may only affect the SRS processes afterwards. In the cases of low laser repetition rate, because of the elimination of the density gradient in the medium by the thermal diffusion, the TDE will be gentle, but it will become more drastic with the repetition rate increasing. In this paper we will report our study on the TDE of SRS in CH$_4$ by using a single longitudinal mode Nd:YAG 532 nm pump laser. We have found that in the laser repetition rate lower than 2 Hz, there is a very intense backward first-Stokes (BS1) with a quantum conversion efficiency of 73% at 1.1 MPa CH$_4$ pressure and pump pulse energy 95 mJ. As we know, this represents the highest SRS conversion efficiency of CH$_4$ reported in the literatures. Moreover, the BS1 peak power turns out to be 2.7 times that of the pump and its beam quality is also much better than that of the

pump beam. With the increase of laser repetition rate from 2 Hz to 10 Hz, BS1 output decreases drastically with a conversion efficiency of 36%. But we found surprisingly that the beam quality of BS1 is different from that of FS1 and only slightly degrades. As evidenced by our experiments, this is because that BS1 represents a wave-front-reversed replica of the pump beam [3-6], in which the TDE is compensated. We expect that a well-behaved CH$_4$ Raman laser with both high efficiency and high beam quality may be designed by a closed CH$_4$/He circulating-cooling system.

II. EXPERIMENTAL SETUP

The experimental apparatus is illustrated in Fig.1. The second harmonic of a Q-switched Nd:YAG laser at 532 nm (Continuum, PR 8000) is used as the pump source which operates on a single longitudinal mode (0.003 cm^{-1}) by means of an injection seeder. The pump laser repetition rate can be varied from 1 to 10 Hz, The pump laser is focused by a 180 cm focal length lens (L$_1$) into a stainless steel Raman cell of 180 cm length, 27 mm internal diameter and equipped with 38-mm diameter by 10-mm-thick quartz windows. The pump laser focal point is located at the Raman cell center. The forward output laser beam that consists of the residual pump (S$_0$), forward first-Stokes (FS1) (630 nm), forward second-Stokes (FS2) (771 nm) and

---

*Author to whom correspondence should be addressed. E-mail: ghsha@dicp.ac.cn, Tel: 0411-84379311

anti-Stokes (FAS1) (461 nm) lines, are collimated by the lens L_2 of focal length 65 cm and then dispersed by the prism (P_1). The backward first-Stokes (BS1) (630 nm) and SBS (532 nm) are reflected by a dichroic mirror (DM) and collimated by the lens L_3 of focal length 100 cm and then dispersed by the prism (P_2). The measurement of pump laser energy is made at the entrance to the cell and the energies of other various components are measured respectively at positions D_1 and D_2 by a calibrated power meter (PowerMax thermopile probe, Molectron, PM30V1). The actual energies in the Raman cell are then obtained by correcting for the losses due to the windows and prisms. The temporal waveforms of incident pump, Stokes, SBS and residual pump are detected by a photodiode (ET 2000, KINGS KC-79-35, Electro-Optics Technology, Inc., rise time <200 psec and fall time <350 psec) and displayed on a digital oscilloscope (TDS3054B, Tektronix). The waveforms of the laser output from exit window are detected at position D_1, while that of BS1 and SBS are detected at position D_2. The time origin for the waveforms is set as the time for the pump pulse peak to arrive the Raman cell center, i.e., the pump laser focus point. To measure the SRS thermal defocusing, a small plane object (P) is inserted into the pump beam (see Fig.1), so that the laser spot shape as well as their light intensity distribution can be recorded by a thermal sensitive paper plus a digital camera.
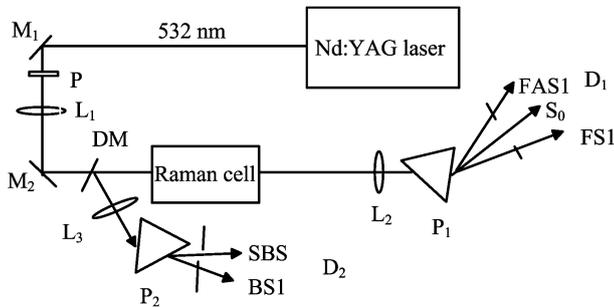

FIG. 1 The diagram of experimental setup.

III. RESULTS AND DISCUSSION

Figure 2 (a) shows the pump energy dependence of the FS1 and BS1 quantum conversion efficiency in 1.5 MPa CH_4 under 2 Hz pump laser repetition rate. The threshold energy for BS1 is about 8 mJ and its conversion efficiency first rapidly goes up with pump energy and then flats over a peak value of 73% at pump energy of 103 mJ. In contrast to BS1, the conversion efficiency of FS1 gets to a maximum value of 22% at pump energy of 20 mJ and then goes down to 15% at pump energy 100 mJ. This big difference between BS1 and FS1 was not caused by their gain coefficients difference.
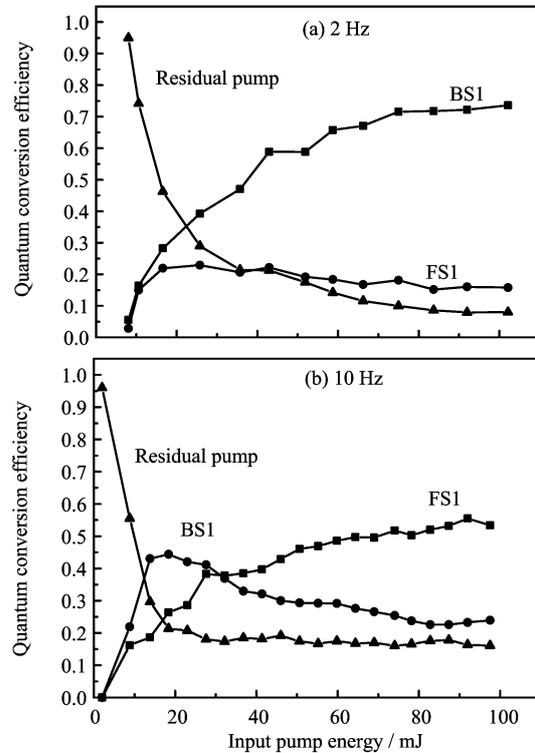

FIG. 2 Quantum conversion efficiency of forward first-Stokes (FS1) and backward first-Stokes (BS1) and residual ratio as a function of pump energy under CH_4 pressure of 1.5 MPa and pump laser repetition rate of (a) 2 Hz, (b) 10 Hz.

For CH_4 pressure 1 to 5 MPa, the Raman linewidth for CH_4 is about 0.35 cm^{-1} [7] which is much larger than that of the single longitudinal Nd-YAG laser, so that the BS1 Raman gain coefficient is only slightly lower than that of FS1. In fact, the high output of BS1 is due to propagating from the focus region counter to the pump beam, so that BS1 pulse front edge is always amplified by the intact or only little depleted pump [8]. As a result, BS1 pulse front abstracts the most parts of pump energy and the pulse is greatly narrowed to about to 1.2 ns with a very steep front edge as shown in Fig.3 (a). Furthermore, since BS1 pulse has accumulated the major parts pump energy; its output power attains 2.7 times that of the pump. It is obvious that due to the competitive suppression of BS1, FS1 is weak and it appears only after BS1 pulse has left out of the focus region as we can see in Fig.3 (a).

Figure 2 (b) displays the experimental results under the same conditions except pump laser repetition rate as 10 Hz. We see the case is just opposite to that described above, while the FS1 conversion efficiency continuously goes up with pump energy to a value of 55% at pump energy of 90 mJ, BS1 maximum conversion efficiency is only 43% and then down to ~25% at pump energy 90 mJ. Due to pump beam quality degrades by TDE,
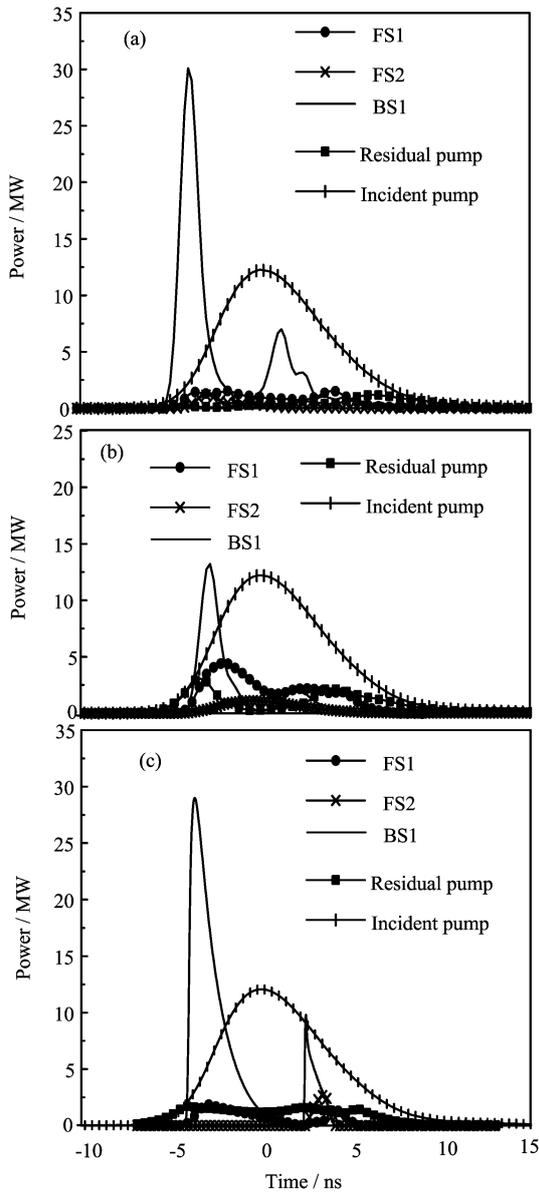
FIG. 3 Measured temporal waveforms for forward first-Stokes (FS1), forward second-Stokes (FS2), backward first-Stokes (BS1), residual pump and incident pump under pump energy of 90 mJ and laser repetition rate of (a) 2 Hz and (b) 10 Hz; (c) Calculated temporal waveforms.

the BS1 peak appears 1.2 ns latter than that under 2 Hz as shown in Fig.3 (b).

The decline of the BS1 conversion efficiency is a result of TDE. However, the influence of the TDE on BS1 and FS1 is different, because BS1 is more sensitive to the pump beam quality. As proposed by Byer [9], to meet the phase-match condition for SRS processes, the coherence length (l_c) must be comparable with the

pump laser Rayleigh length:

$$l_c \approx \frac{c\pi}{2(n_p \pm n_s)\Delta\omega_L} \qquad (1)$$

where the plus and minus signs refer to backward and forward Raman scattering respectively, n_p and n_s are the refractive indices at the pump and the first Stokes frequencies, $\Delta\omega_L$ is the pump laser bandwidth.

From the Eq.(1), we see that, the l_c for BS1 is much shorter than that of FS1 so that BS1 is very susceptible to TDE.

Figure 4 (a) displays the variations of SRS conversion efficiency with CH_4 pressure under pump energy of 95 mJ for 2 Hz pump laser repetition rate. The threshold pressure for FS1 is 0.1 MPa and its conversion efficiency approximately linearly increases with CH_4 pressure achieving a peak value of 40% under 3.0 MPa pressure, while the threshold pressure for BS1 is 0.05 MPa and its conversion efficiency rapidly goes up with CH_4 pressure attaining a maximum value of 73% under ∼1.1 MPa pressure and then goes down to 44%. This reason is that thermal diffusivity decreases with pressure increasing. The thermal diffusion time
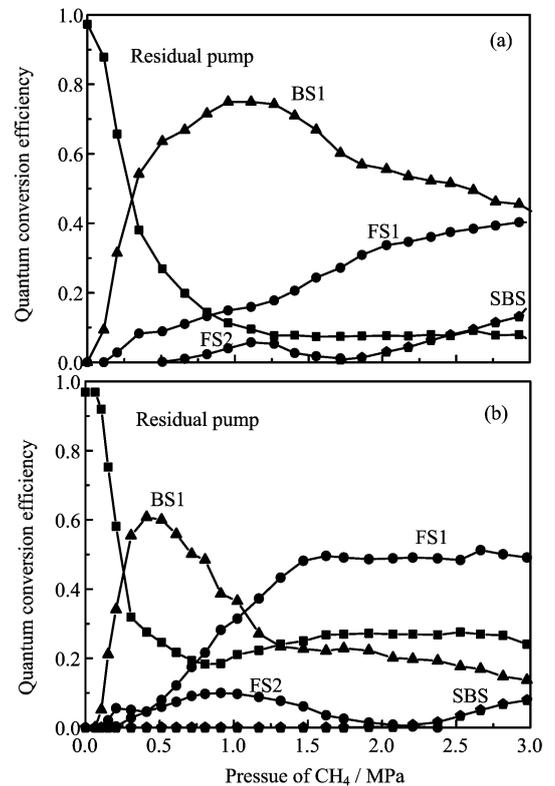

FIG. 4 Quantum conversion efficiencies for forward first-Stokes (FS1), forward second-Stokes (FS2), backward first-Stokes (BS1), stimulated Brillouin scattering (SBS) versus CH_4 pressure under pump energy of 95 mJ and laser repetition rate of (a) 2 Hz and (b) 10 Hz.

(τ) can be expressed by the formula [1,2]

$$\tau = \frac{pC_pR_g{}^2}{4RT\lambda_c} \qquad (2)$$

where p is pressure, R_g is pump beam radius in the Rayleigh region, T is temperature, λ_c is thermal conduction coefficient, R is gas constant.

We have measured R_g of the pump laser in air being 0.35 mm. Considering TDE, we estimate it would be 1.1 mm, then we get the τ being 0.2 s under CH_4 pressure 1.2 MPa so that there is no drastic TDE for pump energy 100 mJ and laser repetition 2 Hz. From the Eq.(2) we can explain the decline of BS1 conversion efficiency at higher pressure. The increasing of FS1 is due to the weaker competitive suppression of BS1 obviously. Under the same conditions, but pump laser repetition as 10 Hz, more serious TDE has been observed so that BS1 maximum conversion efficiency reduces to 60% at CH_4 pressure $\sim$0.5 MPa (Fig.4 (b)).

In a quasi-two-dimension computer program, the same SRS rate equations and algorithm have been adopted [10,11] and the Raman heat release is not considered. The calculated temporal pulse waveforms under pump energy of 90 mJ and CH_4 pressure of 1.0 MPa are shown in Fig.3 (c). We can see that the BS1 and FS1 energy conversion efficiencies and the peak power and BS1 pulse width are well accordant with the experiments at 2 Hz repetition rate (Fig.3 (a)). This result means that TDE can be neglected for the low repetition rate and low CH_4 pressure.
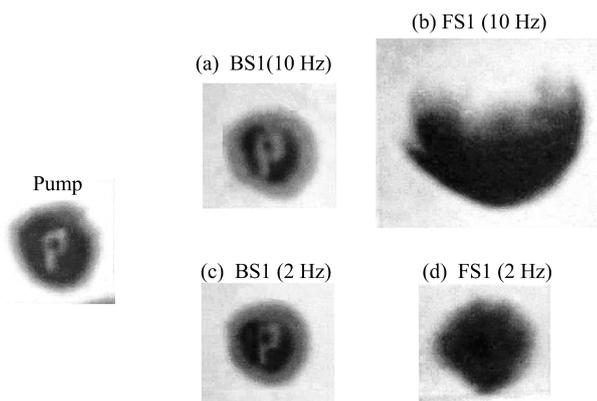

FIG. 5 The laser spot shape of pump, forward first-Stokes (FS1) and backward first-Stokes (BS1) under pump energy of 93 mJ and laser repetition rate of 2 and 10 Hz respectively.

To measure the effect of TDE to laser beam quality, a small plane object (P) was inserted into the pump beam. Under pump energy of 93 mJ and CH_4 pressure of 2.0 MPa, the laser spot of FS1 (Fig.5 (b) and (d)) become blurred, moreover, at repetition rate as 10 Hz it defocuses more seriously and appears as a crescent shape bending up. The reason is that, for high CH_4 pressure and high pump energy, gas density in low area is high due to convection so that FS1 beam distorts and deflects downward. On the other hand, the laser spots of BS1 show the same shape as the pump with the clear P letter and there is no distinct difference between 2 and 10 Hz. This gives evidence that the beam quality of BS1 is not seriously affected by TDE. Raman medium resembles a phase conjugation mirror which reflects BS1 backward so it represents a wave-front-reversed replica of the pump beam, in which the TDE is compensated.

IV. ACKNOWLEDGMENT

[1] T. B. Raymond, R. C. Francis and P. David, Walter J. Chem. Sci. Farady Trans. II **76**, 633 (1980).
[2] T. B. Raymond, R. C. Francis, P. David, Walter. J. Chem. Sci. Farady Trans. II **77**, 1387 (1981).
[3] B. S. Robert and T. V. K. Robert, IEEE J. Quantum Electron. **25**, 2142 (1989).
[4] I. V. Tomov, R. Fedosejevs, D. C. D. McKen, C. Domier and A. A. Offenberger, Opt. Lett. **8**, 9 (1983).
[5] A. I. Sokolovskyaya, G. L. Brekhovskikh and A. D. Kudryavtseva, Opt. Comm. **24**, 74 (1978).
[6] B. Y. Zel'dovich and V. V. Shkunov, Sov. J. Quantum Electron. **7**, 610 (1977).
[7] Y. I. K. Taira and H. Takuma, Chem. Phys. Lett. **91**, 299 (1982).
[8] X. Q. Hua, J. Leng, H. P. Yang, G. H. Sha and C. H. Zhang, Chin. J. Chem. Phys. **18**, 861 (2005).
[9] W. R. Trutna, Y. K. Park and R. L. Byer, IEEE. J. Quantum Electron. **15**, 648 (1979).
[10] J. C. Heuvel and F. J. M. Putten, IEEE. J. Quantum Electron. **29**, 2267 (1993).
[11] C. G. Parazzoli and W. W. Buchman, IEEE. J. Quantum Electron. **24**, 872 (1988).