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Stimulated Raman scattering process for nonlinear Raman lidar monitoring atmospheric carbon dioxide*

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Abstract: The lidar technique based on stimulated Raman scattering (SRS) process has been extensively used in monitoring trace gas concentrations in the atmosphere. To monitor the atmospheric CO_2 concentration, a nonlinear Raman lidar based on the SRS process was devised. A third harmonic Nd:YAG laser wave (354.7 nm) was injected into the Raman cells filled with high-pressure gases, CO_2 and N_2. The first order Stokes lights 371.66 nm(CO_2) and 386.7 nm(N_2) were generated by the stimulated Raman scattering process. The energy of the first order Stokes lights was measured by changing the gas pressure in the Raman cell and the Nd:YAG laser system output energy. The optimum pressures of CO_2 and N_2 in the Raman cell were achieved, which were 0.8 MPa and 3.5 MPa respectively. The principles of this physics process were also discussed.

Key words: Nonlinear Raman lidar; Atmospheric CO_2; SRS process

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Reliably detecting trace gases, such as CO_2, is becoming more and more important due to environment problems and climate changes. Light detection and ranging (lidar) is an active remote sensing technique suitable for monitoring atmospheric species[1]. Nonlinear Raman spectroscopy techniques, such as stimulated Raman gain spectroscopy(SRGS), might be used for remote sensing of the atmosphere. Because of their simple configurations and low cost, the SRS lidar may widely be used for detecting the atmospheric CO_2[2,3].

The atmospheric volume backscatter differential coefficient of atmospheric N_2 can be exactly measured because of its stable proportion in the atmosphere. Moreover, its backscattered voltage signal returned could be used as the reference value[4].

The system involved in nonlinear Raman lidar composes of an ordinary Raman lidar and a Raman cell combined with a shifter. The Stokes light (371.66 nm) was produced when a high-energy pulse (354.7 nm) was injected into the Raman cell. While the Stokes light (371.66 nm) and the pumping laser (354.7 nm) were transmitted into the atmosphere simultaneously, the returned signal was stronger than that when only the pumping laser (354.7 nm) was transmitted through stimulated Raman gain process. The difference was used to retrieve the concentration distribution profile. The intensity and wavelength for the Stokes light (371.66 nm) are key factors for the nonlinear Raman lidar system. It is very important to produce and study the Raman light source, especially the high intensity source of 371.66 nm Stokes light.

1 Stimulated Raman scattering (SRS) process

When a high-energy light is injected into the cell filled with high-pressure gas, the SRS process occurs. Along the SRS interaction process, the interference photons of incidence are scattered by the stimulated phonons. The scattering process for S_1 in SRS is shown in Fig. 1[5]: one interference incidence photon (ω) collides with one hot oscillated phonon (ω_{mn}), which produces one Stokes photon (ω_s) and one scattering pho-

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non (ω) is added simultaneously. The added phonon collides with one photon of incidence again, which produces one Stokes photon and adds one scattering phonon. By repetition and overlap, an avalanche process comes into being, which produces lots of scattering phonons. As shown in Fig. 1, the intensity of the Stokes light is strong enough and it is comparable to that of the injected wave. The key factor for SRS process lies in the enoughness of the photons of incidence.

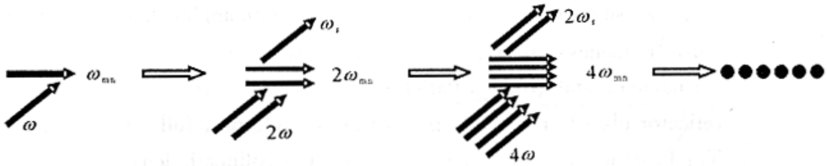

Fig. 1 Process of the SRS

In the granted steady state[6], ignoring the dissipation from the wave of incidence, the power of the Stokes light for SRS process can be expressed as[7]

$$I_s = I_s(0)\exp(g_s I_p l) \quad (1)$$

where $I_s(0)$ is the power of the Stokes wave of incidence, which can be produced by Raman signal of the spontaneous radiation in the Raman cells. I_p is the power of the wave of incidence. g_s is the gaining factor in steady state. l is the length of the Raman cell.

So the energy of the Stokes light (E_s) for SRS process can be obtained by

$$E_s = \pi(\sqrt{2}\omega_0)^2 I_s = 2\pi\omega_0^2 I_s \quad (2)$$

where ω_0 is the frequency of the pumped light. If the energy of S_1 is strong enough, it will produce a second order Stokes light and higher order Stokes lights.

2 Experimental setup and process

The experimental setup for SRS process is shown in Fig. 2. The specifications for the parts of the setup are shown in Table 1. In practice, the Nd:YAG laser emits light of 1 064 nm mixed with light of 532 nm. A third harmonic wave (354.7 nm) is produced through the harmonic crystal group (BBO). The two mirrors (M_1, M_2) filter the miscellaneous waves (532 nm and 1 064 nm), so only the pumping wave (354.7 nm) is injected into the two Raman cells filled with high-pressure gases, CO_2 and N_2 respectively. Focusing the lens (T_1, T_2) would promote the SRS process in the Raman cell. We can see S_1 when the Stokes light passes through the prismatic lens (prism).

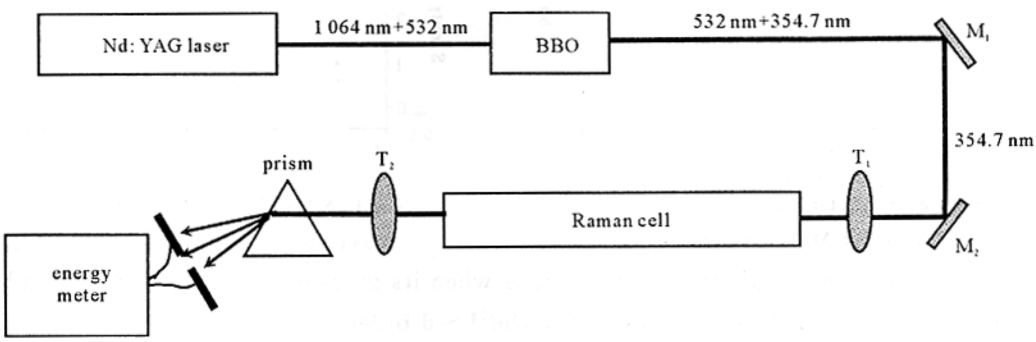

Fig. 2 Setup for measuring the S_1 energy output in SRS process

The experiment was carried out in two steps.

First, we injected the pumping wave into the Raman cell filled with high pressure CO_2 and the Stokes light was generated through the stimulated Raman scattering process. By changing regularly the gas pressure in the Raman cell and the energy of the pumping light, the variable S_1 energy was measured by the energy meter.

Secondly, we replaced the high pressure CO_2 with N_2, repeated the first step and measured the variable S_1 energy for N_2.

At the same time, in the back of the prism, the wavelength of the first Stokes lights (S_1) of the CO_2 and N_2 were measured by the ultraviolet wavelength meter (model: WDS-3). The S_1 wavelength of 371.66 nm was generated for high pressure CO_2, while S_1 wavelength of 386.70 nm for high pressure N_2 was produced.

Table 1 Specifications for the setup

Nd:YAG laser	single pulse energy: 60 mJ (355 nm); radiation angle: 0.5 mrad; radius of beam: 0.4 mm; pulse frequency: 20 Hz; pulse width: 4 ns
BBO	harmonic crystal group (β-$BaBO_4$)
M_1, M_2	reflector placed in 45°, full permeation for 532 nm, full reflection for 355 nm (>99.5%)
T_1, T_2	T_1: focus lens, focal length: 880 mm, T_2: collimatic lens
Raman cell	stainless steel tube, 1 000 mm long, 14 mm in diameter, the entrance and exit covered with flat mirrors
energy meter	model: Max500, Molectro company

3 Experimental results

The S_1 energy output varies along with the gas pressures of CO_2 and N_2 gases (see Fig. 3 and Fig. 4). The curves in Fig. 3 denote the changing of the S_1 energy output for CO_2 gas along with the pumping energy. The four curves show the variety when different CO_2 gas pressures are chosen in the Raman cell: 1 MPa, 0.8 MPa, 0.6 MPa and 0.4 MPa. The three curves in Fig. 4 show the variety when different N_2 gas pressures are chosen in the Raman cell: 3.5 MPa, 2.6 MPa and 2.3 MPa. The two figures obviously show that the S_1 energy outputs increase linearly along with the increase of the injected energy in low pumped energy stage. When the pumping energy exceeds 55 mJ, the S_1 energy outputs saturate and the downtrend appears. Moreover, by setting pumping energy intensity as a constant value (> 47.5 mJ), decreasing gas pressure (ranging from 1 MPa to 0.4 MPa) will increase the S_1 energy for CO_2 gas. When the pressure of N_2 decreases (from 3.5 MPa to 2.3 MPa), the S_1 energy output also decreases apparently.

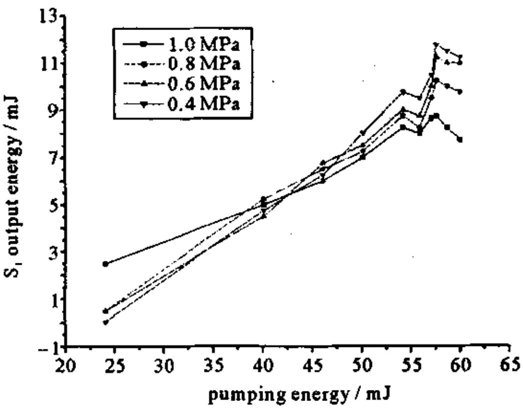

Fig. 3 S_1 energy output for SRS process of CO_2 gas. Gas pressure: 1 MPa, 0.8 MPa, 0.6 MPa and 0.4 MPa

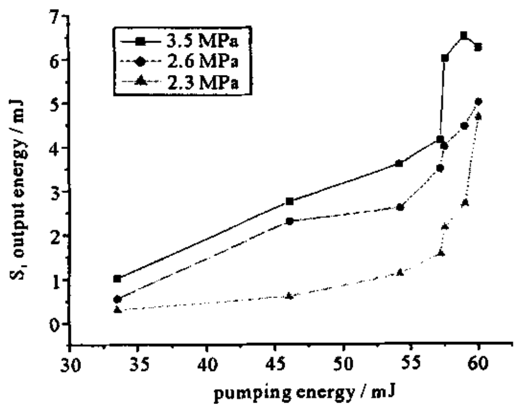

Fig. 4 S_1 energy output for SRS process of N_2 gas. Gas pressure: 3.5 MPa, 2.6 MPa and 2.3 MPa

S_1 energy output for atmospheric CO_2 is obvious when its pressure is below 1 MPa, while the N_2 pressure needs to exceed 2 MPa. In this experiment, the 1 ~ 6 order Stokes lights and 1 ~ 4 order anti-Stokes lights were obviously seen on the paper of fluorescence (N_2: 3.5 MPa; CO_2: 0.8 MPa, 55 mJ pumping energy). From the S_1 energy output, we obtained the optimum pressures: 3.5 MPa for N_2 and 0.8 MPa for CO_2.

As a pumping wave (354.7 nm) is injected into the high-pressure gases, the SRS process occurs. At the same time, other nonlinear chemical and physical processes happens, including the four waves mixing (FWM) process, laser induced breakdown (LIB) process, stimulated Brillouin scattering (SBS) process, etc, which will influence and compete with each other[8].

Altering the gas pressure in Raman cell will change the density of the photons. The optimum density will restrain the FWM, SBS and other nonlinear process, which will promote the SRS process and raises the S_1 energy output.

In actual experiment, changing the focal length and Raman cell length also influences the S_1 energy output. It is found that the S_1 energy output rises apparently with the increases of the Raman cell length and the focal length.

4 The lidar system

Based on the experimental results above, the lidar system involved stimulated Raman gain process is devised. The schematic setup for nonlinear lidar system is shown in Fig. 5.

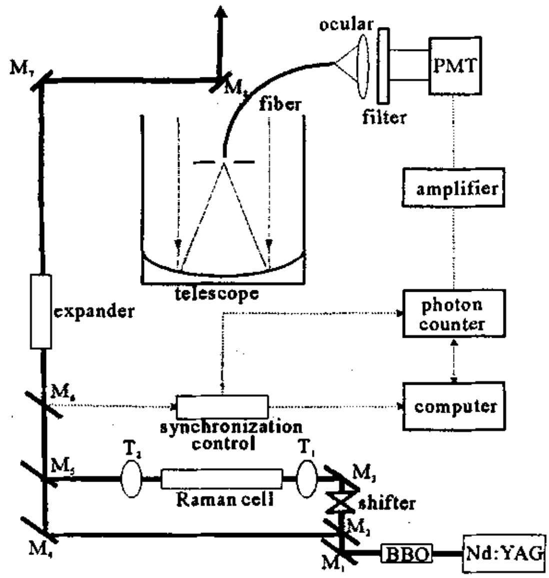

Fig. 5 Schematic of lidar system

When the shifter shuts off, only the pumping wave is transmitted into the atmosphere. The lidar system just becomes an ordinary Raman system. The back scattering signals for CO_2 and N_2 are acquired and shown in Fig. 6 and Fig. 7. The *Y*-axis presents the number of the integrating photons (10,000 shots). It is obvious that the photons for CO_2 are far fewer than that for N_2. According to the two signals, the CO_2 concentration can be retrieved after data analysis and processing.

When the shifter turns on, the Stokes light and pumping light will be transmitted into the atmosphere simultaneously and the SRGS process will occur. The lens (T_1, T_2) are key components for the whole lidar system, which are crucial apparatuses for acquiring the returned signal for SRGS process. Attaining the signal is the aim of our next step work.

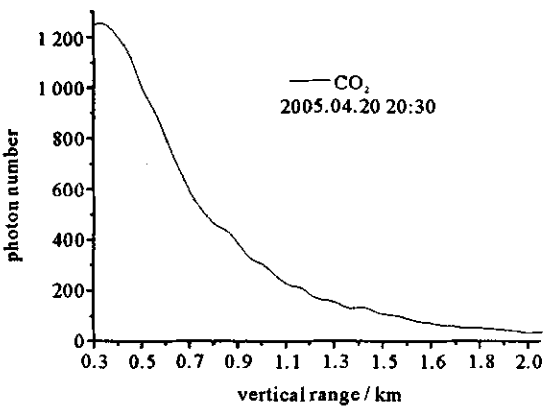

Fig. 6 Raman returned signal for CO_2 gas

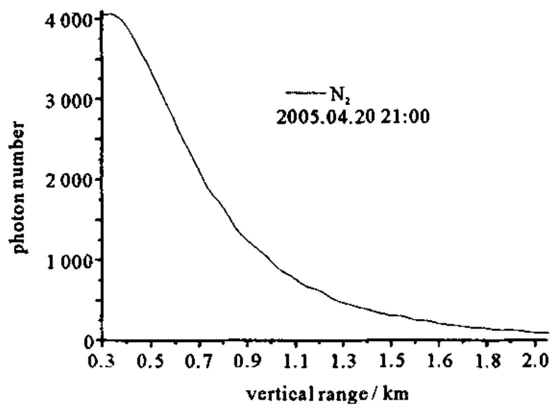

Fig. 7 Raman returned signal for N_2 gas

5 Conclusion

Studying on SRS process for high-pressure gas shows that the pressures of CO_2 and N_2 are two key factors for SRGS lidar system detecting the atmospheric CO_2. The optimum pressures of CO_2 and N_2 in the Raman cell are 0.8 MPa and 3.5 MPa respectively, which will produce efficient S_1 energy outputs. The S_1 energy is influenced greatly by gas pressure chang because of other nonlinear processes. Controlling the gas pressure in the Raman cells would restrain the other processes and promote the SRS process. The technique has applied successfully into the nonlinear lidar system. Moreover, the ordinary Raman returned signals for CO_2 and N_2 can be achieved, from which the CO_2 concentrations profile can be retrieved.

References：

[1] Zhang G X, Zhang Y C, Hu S X. Measurements of planetary boundary layer aerosols with mobile lidar AML-1[J]. *High Power Laser and Particle Beams*, 2004, **16**(3): 286—287.

[2] Immler F. A new algorithm for simultaneous ozone and aerosol retrieval from tropospheric DIAL measurement[J]. *Appl Phys B*, 2003, **76**: 593—596.

[3] Wu Y H, Yue G M, Hu H L, et al. D2 stimulated Raman scattering pumped by fourth harmonic Nd: YAG laser and its application in laser radar[J]. *Chinese Journal of Lasers*, 2000, **A27**(9): 823.

[4] Shen J Q. Laser monitoring the atmosphere[M]. Beijing: Science Press, 2001.

[5] Zhen S X. Laser Raman spectroscopy[M]. Shanghai: Science Press, 1985. 150—151.

[6] Edner H, Fredriksson K, Sunesson A, et al. Mobile remote sensing system for atmospheric monitoring[J]. *Appl Opt*, 1987, **26**(19): 4330—4338.

[7] Tao Z M, Zhang Y C, Lü Y H, et al. Effect of stimulated Raman scattering pumped by fourth harmonic Nd: YAG laser in methane and analysis of its physical processes[J]. *Acta Physica Sinica*, 2004, **53**(8): 2589.

[8] Sonoulepnikoff L D, Mitev V, Simeonov V, et al. Experimental investigation of high-power single-pass Raman shifters in the ultraviolet with Nd: YAG and KrF lasers[J]. *Appl Opt*, 1997, **36**(21): 5023.

非线性拉曼激光雷达系统检测 CO_2 气体的受激拉曼过程

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摘 要： 利用气体的受激拉曼散射增益效应的非线性雷达技术是探测大气中的 CO_2 气体的重要方法，用 Nd:YAG 固体激光器（1 064 nm）的三倍频光（354.7 nm）注入装有 CO_2 和 N_2 高压气体的拉曼管中，气体的受激拉曼散射（SRS）过程产生两种气体的一阶斯托克斯光，用来作为拉曼雷达的发射种子光源。介绍了产生光源的实验装置，论述了 SRS 中气体气压变化与一阶斯托克斯光能量输出变化的定量关系，得到最佳能量输出的优化条件，并对 SRS 中一阶斯托克斯光产生过程的物理机制进行了讨论。并根据光源的试验结果，设计了非线性受激拉曼雷达系统，对前期的普通拉曼雷达进行了实验，得到了初步的实验结果。

关键词： 非线性拉曼激光雷达； 大气二氧化碳； 受激拉曼散射过程