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Generation of UV laser light by stimulated Raman scattering in D_2, D_2/Ar and D_2/He using a pulsed Nd:YAG laser at 355nm*

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A pulsed Nd:YAG laser at 355nm is used to pump Raman cell filled with D_2, D_2/Ar and D_2/He. With adequately adjusted parameters, the maximum photon conversion efficiency of the first-order Stokes light (S_1, 396.796nm) reaches 33.33% in D_2/Ar and the stability of S_1 in pure D_2 is fairly high, the energy drift being less than 10% when the pump energy drifts in the range of 5%. The conversion efficiency and stability, which are functions of the composition and pressure of the Raman medium and the energy of pump laser, are investigated. The result has been used to optimize the laser transmitter system for a differential absorption lidar system to measure NO_2 concentration profiles.

Keywords: stimulated Raman scattering (SRS), conversion efficiency, conversion stability
PACC: 7830, 6300

1. Introduction

Raman scattering experiments have been conducted extensively, especially those on Raman spectra in different materials, such as CN_x nanotubes,[1] InAs nano-grains,[2] H_2, CH_4,[3,4] and so on. On the basis of this, stimulated Raman scattering (SRS) theory was developed and used in some fields. Raman lidar is a successful example because of the good performance as wavelength shifters for differential absorption lidar (DIAL)[3,4] measurements.

Raman cells pumped by second[5]/third harmonic Nd:YAG laser are used in laser ranging,[6] submarine communications, DIAL measurement, and so on. The Stokes and anti-Stokes light caused by SRS are listed in Table 1. And SRS experiments pumped by third harmonic Nd:YAG laser (355nm) in D_2 were reported by Papayannis[7] (lens focal length f=0.5m, range of pressure 50.5–353.5kPa, pump energy 75mJ). In that paper it was reported that the photon conversion efficiency on S_1 was 27%. We did SRS experiments pumped by laser at 355nm in D_2, D_2/Ar and D_2/He in order to obtain higher and more stable conversion efficiency, and they were improved indeed.

Table 1. The wavelength of Stokes and anti-Stokes light in deuterium(D_2) pumped by second/third harmonic Nd:YAG laser (532nm/355nm).

	S_1/nm	S_2/nm	S_3/nm	AS_1/nm
Second harmonic(532nm)	632.77	780.487	1018.17	459.02
Third harmonic(355nm)	396.796	450.231	520.296	320.679

The ith Stokes, ith anti-Stokes, and input pump beams are denoted by S_i, AS_i, and p, their frequencies by $\nu_i^{\rm S}$, $\nu_i^{\rm AS}$, and $\nu_{\rm p}$, respectively. The S_i and the AS_i frequencies are given by $\nu_i^{\rm S} = \nu_{\rm p} - i\nu_{\rm R}$ and $\nu_i^{\rm AS} = \nu_{\rm p} + i\nu_{\rm R}$, where $\nu_{\rm R}$ is the Raman transition frequency. In the case of gases, the plane-wave steady-state Raman gain coefficient $G_{\rm R}$ is given by[8]

$$G_{\rm R} = \frac{{\rm d}\sigma}{{\rm d}\Omega}\frac{2\lambda_{\rm S}^2 \Delta N}{\pi h c^2 \nu_i^{\rm S} \Delta\nu_{\rm R}} I_{\rm p}, \qquad (1)$$

where ${\rm d}\sigma/{\rm d}\Omega$ is the differential Raman cross section (cm^2/sr) at λ=355nm, h is the Planck constant (Js), c is the speed of light (m/s), $\Delta\nu_{\rm R}$ is the Raman linewidth (FWHM in cm^{-1}), $\lambda_{\rm S}$ is the Stokes wavelength (cm), ΔN is the population difference between the initial and the final energy levels (cm^{-3}), and $I_{\rm p}$ is the initial pump intensity. In the case of

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D_2 we have $\Delta\nu_R$=3.67/p+3.58p,[9] where pressure p is given in atm (1atm=101kPa). We can conclude that the Stokes intensity along the Raman cell is $I_S(z) = I_S(0)\exp(G_R Z)$, where Z is the distance, $I_S(0)$ is the initial Stokes intensity.

Four-wave mixing (FWM) is a third-order nonlinear process that coexists with SRS.[9] S_1 is caused by SRS at low pressure or low pump energy, when the energy of S_1 reaches the threshold for S_2 generation, the latter is produced by SRS and the FWM process. Then if the energy of S_2 is high enough, they will produce S_3. In this way higher-order Stokes beams would be produced. The cascade process causes the distribution of the pump energy in different components. In the absence of FWM, the main part of the converted energy would be restricted to 1 or 2 Stokes orders.[10] So we can weaken the strength of FWM to strengthen S_1 and improve its conversion efficiency.

2. Experimental set-up

Figure 1 shows the experimental set-up of the SRS system used. The Nd:YAG laser system has the following features: repetition rate is 10Hz, maximum laser energy emitted per pulse at 355nm is 85mJ and laser pulse duration 18ns.

The Nd:YAG laser beam at 1064nm produces the laser beam at 355nm after passing through a harmonic generator H. A spectral filter F eliminates laser frequencies at 1064 and 532nm. Then the laser beam at 355nm is obtained and is focused into a 1.5m-long stainless steel Raman cell after passing through a variable attenuator VA and a beamsplitter BS. The Raman cell output laser beam is recollimated by a lens at the end of the Raman cell. The two lenses of Raman windows are plane-convex quartz lenses of 75cm focal length with 1.8cm diameter. The pressure inside the Raman cell is indicated by a calibrated high-pressure gauge PG. The purity of D_2 is 99.99%. For Ar and He, which are used as buffer gases, the purities are 99.99% and 99.999 %, respectively.

The laser of various wavelengths generated by the SRS effect are separated by a quartz prism P and a diaphragm D located at the exit of the Raman cell, as shown in Fig.1. The output laser energy at each wavelength is measured by the precise detector 1 (molectron detector, EM500 single channel joulemeter) connected with a personal computer. The beamsplitter BS placed before the Raman cell is used to reflect a small part (13%) of the incoming laser energy to detector 2 (same as the former) connected with another personal computer. The two computers are connected by the control bus, and record simultaneously the input and output energies for each pulse. Taking the ratio of the two detectors' signals and the absorption and reflection losses at the various optical elements used (Raman cell windows, prism P) into account, we obtain the photon conversion efficiency and the stability of the forward SRS effect.

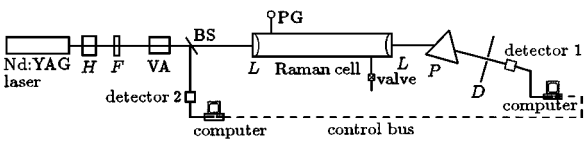


Fig.1. The experimental set-up of the SRS experiment.

3. Experimental results and discussion

The results presented in this paper will be limited to forward Raman scattering, and the photon conversion efficiencies of the 355nm laser radiation to S_1, S_2, AS_1 and the residual pump laser.

We measured SRS conversion efficiency in pure D_2 in certain pressures range (404 to 2626kPa). Figure 2 shows the results for a given pump energy of 63mJ. The photon conversion efficiency on S_1 increased dramatically to 24% while the pressure increased to 1010kPa from 404kPa. Then it climbed up slowly to the maximum 25% until the pressure was 2626kPa, at the same time, the maximum on S_2 reached 16%. The course of change in the case of S_2 was similar to that of S_1, it also increased with the pressure, but more slowly, due to its lower gain. In this experiment the maximum values of the photon conversion efficiency on S_1 and S_2 in pure D_2 were slightly higher than those we obtained before, for we used a much longer Raman cell (1.5m) and elongated focusing geometry at the entrance of the cell. That caused the reduced potential of vector phase matching with the reduced numerical aperture.[11] These two experimental factors (Raman cell length and focusing geometry) increased the Raman gain coefficient and suppressed the build up of FWM process. So the conversion efficiencies of the SRS conversion on S_1 and S_2 were enhanced. This result is understood from the Eq.(1), too. The AS_1 occurred when the pressure increased to 505kPa. During the whole process the residual pump energy decreased because of the constancy of the total energy. The SRS energy con-

version efficiency has not been studied at the pressure higher than 2626kPa because of the limited experimental conditions.

The addition of a buffer gas (i.e. Ar, He) in the Raman active medium plays a twofold role: it reduces simultaneously the FWM processes (by increasing the wavevector mismatch) and the Raman gain (by increasing the Raman linewidth without increasing the number density of the active gas).[11] The Raman linewidth of a gas mixture can be approximated by $\Delta\nu = \Delta\nu_0 + \gamma p_b$,[11] where $\Delta\nu_0$ is the self-broadened linewidth, γ is the broadening coefficient, and p_b is the buffer gas partial pressure. As a result, for a carefully selected active gas/buffer pressure ratio, the addition of a buffer gas may significantly increase the first Stokes (S_1) conversion efficiency.[12]

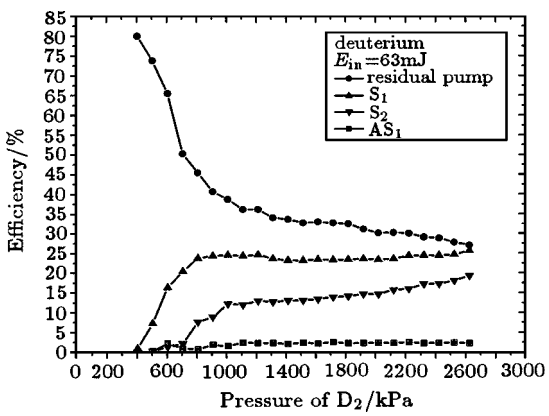


Fig.2. Energy conversion efficiency versus D_2 pressure at a constant pump energy of 63mJ.

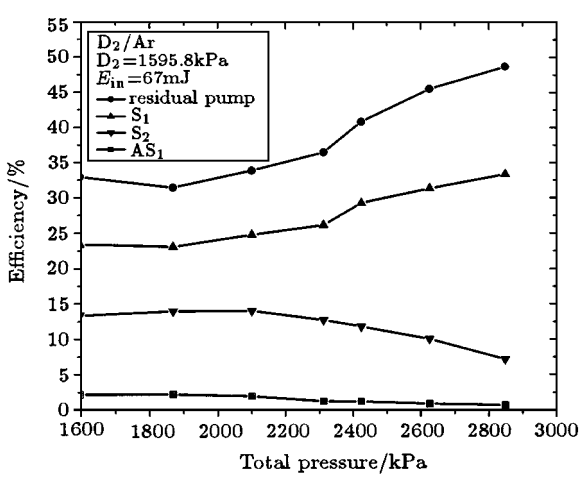


Fig.3. Energy conversion efficiency versus D_2/Ar total pressure, and the partial pressure of D_2 is 1595.8kPa.

We studied the photon conversion efficiency on S_1 in D_2/Ar with 1010kPa partial pressure of D_2, and found the maximum conversion efficiency on S_1 was 25.48%. When the partial pressure of D_2 was increased to 1595.8kPa, the maximum conversion efficiency on S_1 reached 33.33%, which is obviously higher than that in pure D_2. The results are shown in Fig.3. It is obvious that the energy conversion efficiency on S_1 increases with the total pressure, while the changing trends are contrary for S_2 and AS_1. Because increasing the pressure of Raman medium leads to greater dispersion in it, and thus the FWM processes persists, which prevents the generation of S_2 and AS_1. Then D_2/He gas mixture was studied, and the results are shown in Fig.4. The maximum conversion efficiency on S_1 reached 31.01%, and the effect was not more obvious than that with Ar. As analysed above, we can increase the energy conversion efficiency on S_1 and decrease simultaneously the others by choosing a suitable buffer gas with larger molecular mass and adjusting the pressure ratio of active gas/buffer gas. Because of the existence of the FWM process, the SRS light module competition and the instability of the pump energy, the energy of SRS light was unstable and changed during the whole course. Especially, the instability of S_1 does harm to the detection of atmosphere. So we expected to gain stable S_1 light by changing the experimental parameters. Now we assume that the stability degree $\zeta = \frac{n_1}{n_2} \times 100\%$, where n_1 is the number of S_1 pulses which drifts in the range of 10% for the average output S_1 energy, n_2 is the total number of the output S_1 pulses. In this experiment, the two computers recorded in real time the input and output energy for each pulse. It was indicated that the pump light was stable and drifted within the range of 5%. In the case of pure D_2, the stability of S_1 became higher with increasing pressure, and eventually ζ tended to a constant value asymptotically. That means nearly all of the energy of output S_1 pulses drifted in the range of 10%. The result is much better than that reported before. At a high pressure, the FWM process was reduced (and enhanced the wavevector mismatch, preventing the generation of S_2 and other higher-order Stokes light and AS_1). That strengthened S_1 and improved its conversion stability. Figure 5 shows the dependence of output S_1 energy on the pump energy at a given pressure of the Raman medium. It is evident that the changing trends are similar. When the pump energy is strong, the output S_1 energy is strong too, which is easy to understand from the Eq.(1) mentioned above.

But they are not proportional because of the existence of FWM process and SRS light module competition. Under the given conditions described in Fig.5 there was only one output S_1 pulse which drifted beyond the range of 10% among more than 300 output S_1 pulses, and ζ was greater than 99.7%.

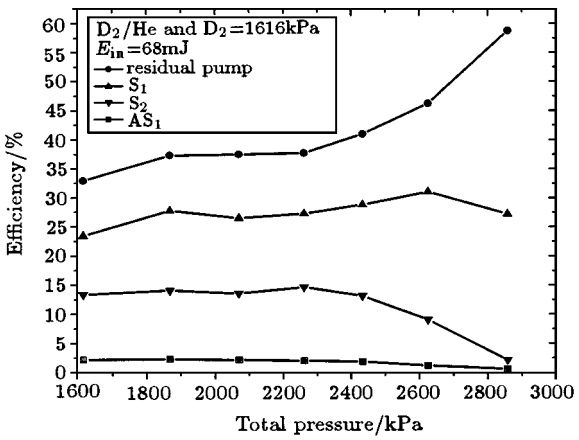


Fig.4. Energy conversion efficiency versus D_2/He total pressure, and the partial pressure of D_2 is 1616kPa.

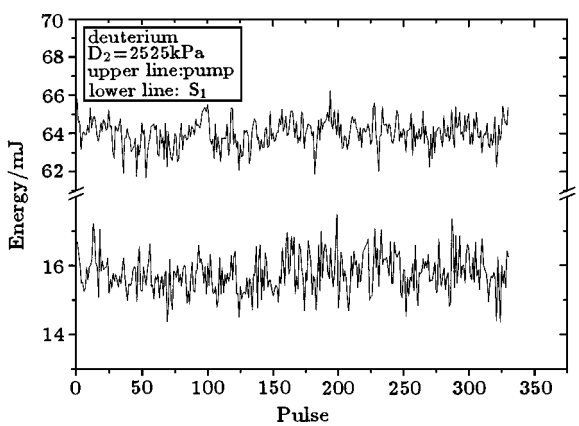


Fig.5. Real time energy curves of pump light and S_1 for each pulse at a pressure of 2525kPa with D_2.

Figure 6 shows the stability of different states in D_2, D_2/Ar and D_2/He. The results prove that the effect of stability caused by buffer gases becomes strong with their molecular mass and partial pressure. A conclusion thus can be drawn. The addition of buffer gas (Ar/He) is not only useless for stabilizing the output S_1 energy, but makes it worse compared with the pure D_2. Because the addition of buffer gas causes the relative decrease of the number density of the active gas, and the pump energy is shared by the active and buffer gas, the active gas has less pump energy than in pure D_2. The mechanism of SRS light drifts is the existence of quantum noise of the Raman medium.

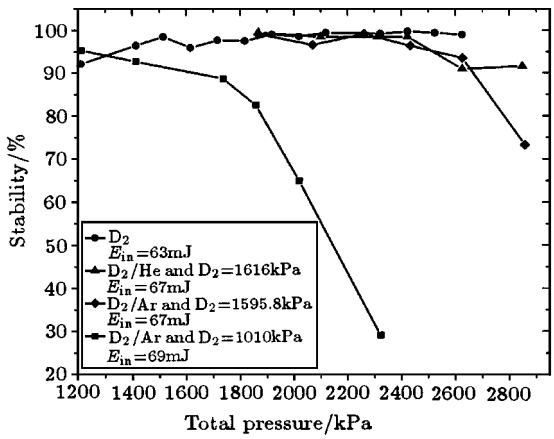


Fig.6. Stability curves on S_1 energy at different states versus pressure.

4. Conclusion

We worked over SRS experiments in pure D_2, D_2/Ar and D_2/He. And the focus was to optimize the photon conversion efficiency and stability on S_1, and to find their relationship with the composition and pressure of the Raman medium and the pump energy. A conclusion is drawn that the energy conversion efficiency and stability on S_1 are satisfactory when the pressure is 2626kPa in pure D_2. Adding buffer gases with larger molecular mass into D_2, the energy conversion efficiency on S_1 becomes little higher, but the stability becomes worse. And it is important to use longer Raman cell and higher pump power in this experiment. The results may have practical value in optimizing the laser transmitter system used in a DIAL system to measure NO_2 concentration profiles, biomedical diagnostics, submarine communications, and so on.

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