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Stimulated Rotational Raman Scattering at Multiwavelength under TEA CO_2 Laser Pumping with a Multiple-Pass Cell[1]

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Abstract—Stimulated rotational Raman scattering (SRRS) at multiwavelength pumped by TEA CO_2 laser was demonstrated in this paper. Raman mediums were cooled by liquid-N_2 and a multiple-pass cell (MPC) with 25 passes was designed and used. When the para-H2 was pumped by single-longitudinal-mode (SLM) circular polarized TEA CO_2 laser on 10P(20), 9P(20), and 10R(20), 50 mJ 16.95 μm, 350 mJ 14.44 μm, and 536 mJ 16.9 μm radiations were obtained, corresponding to energy conversion efficiency of 1.2, 11.7, and 13.4%, respectively. When the ortho-D_2 was pumped by CO_2 laser on 10R(18), 108 mJ 12.57 μm Raman laser was obtained with energy conversion efficiency of 2.9%.

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1. INTRODUCTION

Raman effect is widely used for frequency shift of laser radiation [1–16]. By using stimulated rotational Raman scattering (SRRS) of CO_2 laser radiation, coherent far-infrared (FIR) radiation can be generated efficiently, which is necessary for laser photochemistry, remote sensing and laser isotope separation of UF_6 [17–20]. Since the potentiality of SRRS in H_2 for an efficient, high-energy, 16 μm source was presented by R.L. Byer in 1976, much attention was paid on developing a high-power H_2 Raman laser [21]. 16 μm pulses with energy of 2 μJ were demonstrated by M.M.T. Loy based on four-waves mixing in para-H_2 [22]. Using a multiple-pass cell (MPC), R.L. Byer and W.R. Trutna produced 50 mJ of 16.9 μm radiation by amplification of a signal generated by four-wave mixing [23]. 16 μm Stokes pulses with an output energy of 2.4 mJ were generated by an intracavity configuration for the para-H_2 Raman laser by A. Tsunemi in 1999 [24].

SRRS in H_2 pumped by a CO_2 laser has several useful features. For one thing, the CO_2 laser is technologically well developed, efficient, and scalable to high peak and average power. For another thing, since the CO_2 laser is tunable, the corresponding Raman shifts are also tunable. The main difficulty in obtaining SRRS in H_2 in the FIR is the relatively low Raman gain in this region of the spectrum, despite the very narrow linewidth of the Raman transition. Since H_2 is a homonuclear molecule, there are no dipole-allowed rotational or vibrational transitions. Thus, the Raman susceptibility is determined solely by the electronic transitions, which lie in the region greater than 90000 cm^{-1} above the ground state. It is therefore not surprising that the Raman susceptibility and hence the Raman gain is quite low for scattering of a CO_2 laser at 1000 cm^{-1}.

In this paper, we report a study of SRRS using a MPC. To improve the gain, the Raman mediums were cooled by liquid-N_2 and a MPC with 25 passes was designed and used. When the para-H_2 was pumped by single-longitudinal-mode (SLM) circular polarized TEA CO_2 laser on 10P(20), 9P(20), and 10R(20), 50 mJ 16.95 μm, 350 mJ 14.44 μm and 536 mJ 16.9 μm radiations were obtained, corresponding to energy conversion efficiency of 1.2, 11.7, and 13.4%, respectively. When the ortho-D_2 was pumped by CO_2 laser on 10R(18), 108 mJ 12.57 μm Raman laser was obtained with energy conversion efficiency of 2.9%.

2. EXPERIMENTAL SETUP

The experimental arrangement is shown in Fig. 1. The experimental system consisted of a high power TEA CO_2 pump laser system, and a liquid-N_2-cooled MPC and detection equipments. The Raman scatting section of H_2 for circular polarization pumping laser is about 1.5 times than linear polarization laser, so the high power, tunable, circular polarization SLM CO_2 laser is needed for application in isotope separation. In experiments, SLM CO_2 laser was obtained by using a TEA CO_2 oscillator and a low-pressure gain module.

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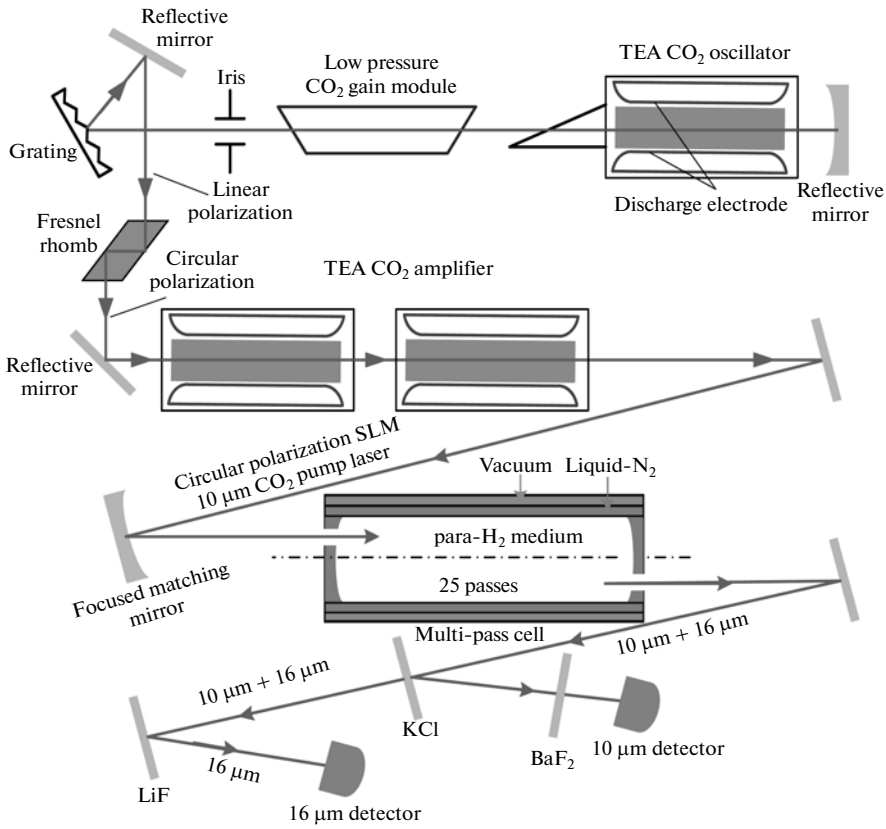

Fig. 1. Experimental setup of a para-H_2 Raman laser system under TEA CO_2 laser pumping.

The laser cavity was formed by a concave reflective mirror and a grating, and the output wavelength can be tuned by rotating the grating. An internal Iris limited oscillation to the TEM_{00} transverse mode. Brewster plate was used for sealing the TEA CO_2 oscillator and generating a linear polarization output. A Fresnel rhomb changed the polarization from linear to circular for maximum Raman gain. The circular polarized beam was amplified by two-stage TEA CO_2 amplifiers. The maximum energy at the input to the multiple-pass cell was 4.5 J, with a temporally smooth pulse of 90 ns full width at half maximum and a divergence angle of 2.2 mrad. The MPC cell had a triple metallic structure, and two inner layers were made by oxygen-free copper and the outside layer was aluminum. The 4-m-long inner cell was cooled by liquid-N_2 and thermally isolated from the outer cell by vacuum insulation. A pair of 2-m-radius copper mirrors separated by 3.77 m was mounted in the inner cell, and the reflectivity at 10.6 μm of these two mirrors was 97%. The pump beam with proper intensity was coupled into the MPC through a 2-cm-diameter hole on the mirror, and then bounced back and forth between two mirrors with 25 passes, and the pump beam was focused at the central of the cell at every pass. The Stokes and depleted pump pulses were extracted from a coupling hole on the opposite mirror. Para-H_2 gas with pressure of 300 Torr was introduced to the inner cell and was cooled to approximately 100 K. The Stokes pulses were filtered by using the reflection from LiF and the depleted pump pulses by using the transmission from BaF_2. The Stokes and pump pulses were simulta-

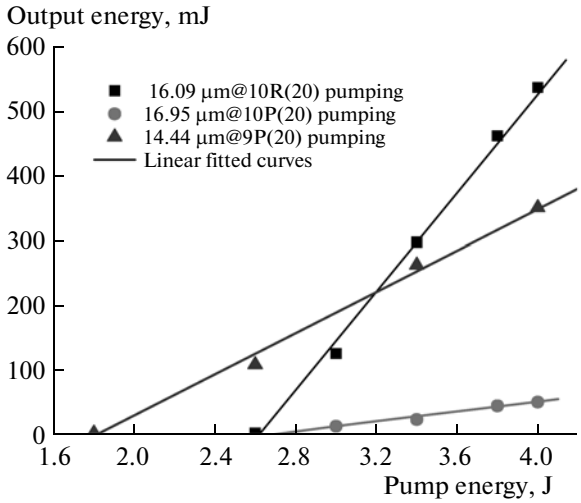

Fig. 2. Output energy of Raman laser versus the pump energy.

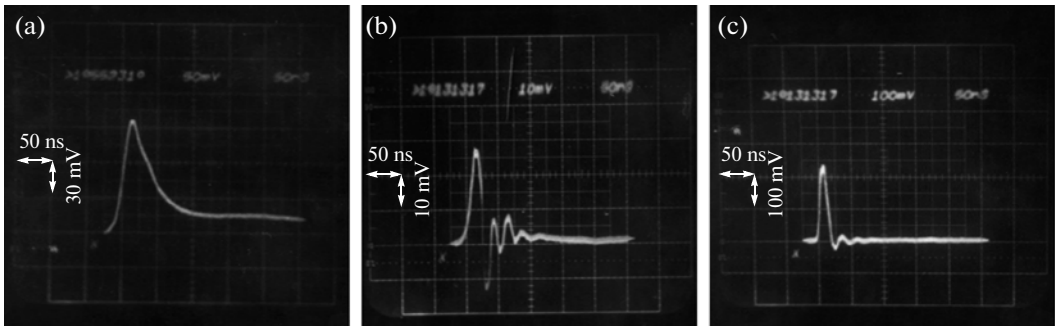

Fig. 3. Pulse shape of TEA CO_2 laser (a), depleted pump laser (b), and Raman laser (c).

neously detected with photon drag detectors and were displayed on an oscilloscope.

3. EXPERIMENTAL RESULTS AND DISCUSSION

Pumped by TEA CO_2 laser oscillating at P or R lines, SRRS in para-H_2 operating at different wavelength were obtained. For the 10P(20), 9P(20), and 10R(20) lines pumping, threshold energies were measured to be 2.7, 1.8, and 2.6 J, respectively. Above the threshold, the Stokes energy linearly increased with the increase of the pump energy. At a pump energy of 4 J on 10P(20), 16.95 μm Raman laser with the maximum output energy of 50 mJ was obtained, with an energy conversion efficiency of 1.2%. At the pump energy of 4 J on 9P(20), up to 350 mJ 14.44 Raman laser was obtained, with an energy conversion efficiency of 11.7%. At the pump energy of 4 J on 10R(20), as much as 536 mJ 16.09 μm Raman laser was obtained, with an energy conversion efficiency of 13.4%. When the linear polarized CO_2 laser was used as pump source, no Raman laser was generated even the pump energy was higher than 5 J. In experiments, laser induced plasma was observed in the MPC, so the seal of Raman MPC was very important. As shown in Fig. 2, we can see that the pump laser below the threshold has no contribution to Raman laser, the depleted pump pulses and the Stokes pulses was coincidence with the peak pulse of pump laser, and the tail pulse contribute less to generation of Raman laser. The pulse width of Raman laser was measured to be 30 ns.

When the para-H_2 was replaced by ortho-D_2 in MPC, Raman laser was also obtained with the TEA CO_2 laser pumping. The pressure of ortho-D_2 was 300 Torr and it was also controlled at a temperature of 100 K. At a pump energy of 3.7 J on 10R(18), 108 mJ 12.57 μm Raman laser was obtained, with an energy conversion efficiency of 2.9%. Therefore, FIR raman laser with different wavelength can be obtained conveniently by changing the pump wavelength of CO_2 laser or the Raman medium in MPC.

4. CONCLUSIONS

Multiwavelength SRRS pumped by TEA CO_2 laser was presented. Raman mediums were cooled by liquid-N_2 and a MPC with 25 passes was designed and used. When the para-H_2 was pumped by SLM circular polarized CO_2 laser on 10P(20), 9P(20), and 10R(20), 50 mJ 16.95 μm, 350 mJ 14.44 μm, and 536 mJ 16.9 μm radiations were obtained, corresponding to energy conversion efficiency of 1.2, 11.7, and 13.4%, respectively. When the ortho-D_2 was pumped by CO_2 laser on 10R(18), 108 mJ 12.57 μm Raman laser was obtained with energy conversion efficiency of 2.9%.

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