

DIAL MEASUREMENT OF CH₄, CO₂, CO and N₂O USING A TUNABLE IR SOURCE BASED ON THE Ti:SAPPHIRE LASER

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INTRODUCTION

Range-resolved differential absorption lidar (DIAL) has already been in the practical stage for the detection of naturally abundant atmospheric O₃ and H₂O. It has also been used in the measurements of some highly abundant species of pollutant molecules. Recent progress in high-power tunable IR sources makes the range-resolved detection of other important species possible. We have developed a high-power Raman laser which is continuously tunable up to 3.2 μm and pumped by a Ti: sapphire laser. Using the HITRAN database, we surveyed useful absorption lines for range resolved DIAL measurement in the 1 ~ 5 μm range, and found that CH₄, CO₂, CO and N₂O in the troposphere were detectable. A detailed simulation of the DIAL measurement of these species was made for many absorption lines, taking into account the interference from other species.

DEVELOPMENT OF TUNABLE IR SOURCE

The Ti:sapphire laser is the most efficient tunable solid-state laser, and has a wide tunable range in the near IR region. However, the tunable range is insufficient for the detection of atmospheric molecules except H₂O. We developed a high-power source tunable from 680 nm to 3.2 μm, using the Raman shifting technique of the Ti:sapphire laser.

The whole system is home-made, except for the frequency doubled Nd:YAG laser (Spectra-Physics GCR-3, 470 mJ at 532 nm, 10 Hz) used as a pump source¹⁾. The Ti:sapphire laser has an oscillator-amplifier configuration. Three SF-10 Brewster prisms and a non-coated quartz etalon (2.4 mm thick) are the tuning elements, giving a spec-

tral bandwidth of 0.2 cm⁻¹. With a specially coated wide-range coupling mirror, a tunable range of 680 ~ 930 nm was attained²⁾. After double-pass amplification with a single Ti:sapphire crystal, a maximum output energy of 100 mJ/pulse was obtained.

The output beam was focused into a 1 m Raman cell with a f=0.6m lens. The tuning curves of the fundamental wave of the Ti: sapphire laser and the stimulated Raman emission are shown in Fig. 1. Using H₂ gas at a pressure of 20 atm, we obtained tunable ranges of 973 ~ 1392 nm for the first Stokes lines (S₁) and 1673 ~ 3180 nm for the second Stokes lines (S₂). In the case of CH₄ at 15 atm, tunable ranges of 903 ~ 1131 nm for S₁ and 1302 ~ 1709 nm for S₂ were covered.

Thus, we could cover most of the wavelength range from 680 nm to 3.2 μm at a pulsed energy of more than 10 mJ. The longer wavelength region was limited by the transmittance of the quartz window used. By changing it to MgF₂, we can expect further expansion into the longer wavelength region.

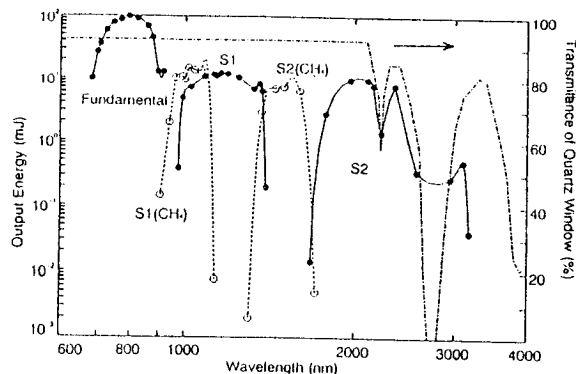


Fig. 1. Tuning curves in IR region obtained by Raman laser pumped by Ti:sapphire laser.

SIMULATION OF DIAL MEASUREMENT OF CH₄, CO₂, CO and N₂O

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We surveyed useful absorption lines for the range-resolved DIAL measurement of CH₄, CO₂, CO and N₂O using the HITRAN database for the 1 ~ 5 μm range, and made an estimation of the error in the DIAL measurement³⁾. According to the results of our simulation, the optimum absorption line strength S₀ [cm⁻¹/(mol.cm⁻²)] is approximately given by 10⁻²⁰/M, where M is the mixing ratio of the molecule in ppm. Another important factor in selecting the optimum line is that the interference from other lines is small. To evaluate this, ξ, the ratio of the absorption by other lines at the on-line wavelength to that by the line of interest was taken. ξ = ξ^s + ξ^o, where ξ^s is due to the molecule of interest and ξ^o is due to other molecules. The interference from the lines of the other molecules must be minimal because of the uncertainty of their densities. Therefore, ∑ ξ^o ≪ 0.1 was used. The sum of the absorption by all possible lines except that of the line of interest

must also be minimal so that the on-line laser light is not highly attenuated, leading to a shortening of the measurable range. Thus, the preferred lines to be used as the DIAL on-line wavelength were selected by these criteria and shown in Table 1.

The most important source of noise in the IR region is the dark current noise of the detector. We assumed the best NEP value ever obtained for each wavelength. The laser was assumed to have a linewidth of 0.1 cm⁻¹ FWHM and an output of 10 ~ 20 mJ. The measurable ranges by DIAL measurement were calculated for a range resolution of 100 m and a 10000-shot accumulation. As a result, we found that a continuously tunable laser has the possibility to greatly extend the measurable range of range-resolved DIAL in the IR region by using the optimum line as the on-line wavelength.

We are preparing to perform DIAL measurements of atmospheric CO₂ and CH₄ at 2.05 μm and 2.28 μm respectively.

References:

- 1) M. Funayama et al., Opt. Comm. 102 (1993) 457.
- 2) M. Funayama et al., Jpn. Appl. Phys. 32 (1993) L1332.
- 3) M. Uchiumi and M. Maeda, Laser Kenkyu (1994), to be published.

Table 1. Simulated detection ranges of vertical (Z_{max}) and horizontal (R_{max}) soundings by DIAL for ΔR=100m and 10000 shots.

Species	λ _{vac}	S ₀	γ	∑ _{i=1} [∞] ξ _i ^s	Strongest interfering species (ξ _{max} ^o)	∑ _{i=1} [∞] ξ _i ^o	Z _{max}	R _{max}
	μm							
CO ₂	2.075	4.0 × 10 ⁻²³	0.0714	0.869	H ₂ O (0.017)	0.032	2.96	4.89
	2.074	6.7 × 10 ⁻²³	0.0718	0.585	H ₂ O (0)	0.004	2.92	4.92
	2.048	4.2 × 10 ⁻²³	0.0714	0.265	H ₂ O (0.012)	0.021	3.20	5.80
	2.047	3.1 × 10 ⁻²³	0.0711	0.308	H ₂ O (0.017)	0.024	3.09	6.00
	1.997	7.6 × 10 ⁻²³	0.0706	2.301	H ₂ O (0.026)	0.094	2.07	2.60
CH ₄	3.451	6.9 × 10 ⁻²¹	0.0510	4.78	H ₂ O(0.035)	0.154	0.81	0.90
	2.284	4.6 × 10 ⁻²¹	0.0646	0.41	N ₂ O (0.011)	0.012	2.69	5.00
	2.262	3.9 × 10 ⁻²¹	0.0630	0.95	N ₂ O (0.025)	0.063	2.51	4.50
	2.301	4.4 × 10 ⁻²¹	0.0625	0.40	H ₂ O(0.022)	0.063	2.62	5.00
N ₂ O	4.542	4.3 × 10 ⁻²⁰	0.0823	42.94	CO ₂ (0.085)	0.495	0.40	0.41
	3.935	1.7 × 10 ⁻²⁰	0.073	2.11	H ₂ O (0.012)	0.024	1.52	2.40
	3.914	1.9 × 10 ⁻²⁰	0.0814	2.24	CH ₄ (0.020)	0.054	1.53	2.39
	3.883	2.4 × 10 ⁻²⁰	0.0775	2.09	CH ₄ (0.026)	0.131	1.60	2.41
	3.936	1.6 × 10 ⁻²⁰	0.0726	1.17	H ₂ O (0.013)	0.064	1.60	2.59
	3.933	1.8 × 10 ⁻²⁰	0.0734	2.11	CH ₄ (0.037)	0.085	1.55	2.40
CO	4.700	3.2 × 10 ⁻¹⁹	0.0667	0.02	H ₂ O (0.039)	0.244	2.00	3.05
	4.618	4.4 × 10 ⁻¹⁹	0.0625	0.003	H ₂ O (0.087)	0.54	1.81	2.48
	4.610	4.6 × 10 ⁻¹⁹	0.0611	0.0004	N ₂ O (0.100)	0.524	1.81	2.45
	4.539	1.3 × 10 ⁻¹⁹	0.0528	0	N ₂ O (0.640)	32.07	0.42	0.49