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New solid state lasers for hydrocarbons detection by LIDAR

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I - Introduction

Current development of lidar/DIAL technology allows to obtain 3D mappings of the concentration of air pollutants at highest sensitivity (ppb range) and over large distances (10km) [1-6]. Presently it is possible to monitor real-time distributions and dynamics of nitrogen oxides, sulphur dioxide and ozone. Recently, Milton has demonstrated the detection of toluene by lidar in the ppm range [7]. Routine of fully automatic operation has been severely restricted for a long time, however, by the complexity and maintenance of the usually employed Nd:YAG or Eximer-pumped dye lasers. The advent of new tunable all solid state laser systems, such as vibronic lasers, opens a new era in the domain of user-friendly and fully automatic DIAL operation. The first lidar system based on a new flash lamps-pumped Ti:Sapphire laser has been described in [8].

A challenge for lidar is now to detect hydrocarbons. Actually they are present in the atmosphere at higher and higher concentrations and are difficult to measure separately with standard methods. As all these molecules absorb light in the $3\mu\text{m}$ region (H-C stretching mode), we have developed a new infrared tunable extension: a KTP optical parametric oscillator (OPO) which can be implemented in the all solid state lidar (§ II).

And as an introduction to hydrocarbons detection, we were able to get the first vertical toluene concentration profile over the city of Leipzig in the ppb range (§ III).

II - KTP no-tracking NCPM-OPO

KTP has got large non linear coefficients, high temperature stability and high transparency up to $3.5\mu\text{m}$ [9]. Our 16mm-long x-cut crystal was pumped by a Q-switched Ti:Sapphire laser (linewidth of 0.2cm⁻¹, tunable from 700 to 950nm, pulsewidth of 50ns). The pump wavelength has been scanned to tune the output wavelengths instead of tilting the crystal angle. This is much easier and eliminates synchronisation problems for lidar applications. Theoretical phase-matching curves were computed from the Sellmeier equations and the phase-matching conditions (fig.1) [10].

The OPO was in a singly resonant configuration: two laser mirrors 2cm apart from each other allow around 250 roundtrips during the 50ns-pulse. The rear mirror was -1m curved, reflected 99.9% of the signal and transmitted 90% of the pump. The flat output mirror transmitted 5% of the signal, 75% of the idler and 95% of the pump. The wavelengths were measured with H2O Jobin-Yvon spectrometers. The signal was detected with an 88XL Photodyne infrared germanium detector.

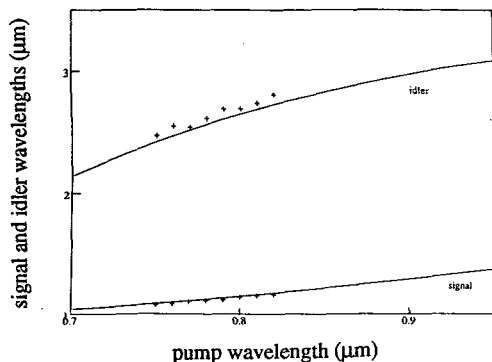


Fig.1 - Signal and idler wavelengths are plotted against pump wavelength. Solid lines are theoretical curves and crosses are experimental points.

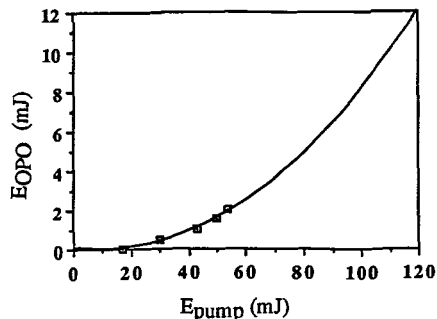


Fig.2 - OPO output energy against pump energy at 780nm.

We were able to get a signal between 1076nm and 1158nm and a corresponding idler from 2475nm to 2809nm by tuning the pump from 750 to 820nm (fig.1). The oscillator threshold was 17mJ when the pulsewidth was 50ns and the beam area was 2mm² which yields a threshold of 17MW/cm². Output energies are plotted against pump energy in fig.2 for $\lambda = 780$ nm. Efficiency at the same wavelength is given in fig.3 and is, as expected, linearly increasing above threshold. If the OPO were pump with the new lidar system [8] that is with a few hundreds millijoules, efficiencies greater than 10% and energies of a few tens of millijoules could be expected. The spectral range would also be extended from 1040 to 1380nm for the signal and from 2160 to 3100nm for the idler.

This new solid state source will allow us to do lidar in the near infrared. We will try to detect molecules such as CO, CO₂ or methane. Our final goal is to measure hydrocarbons around 3μm, maybe using of FTIR spectroscopy.

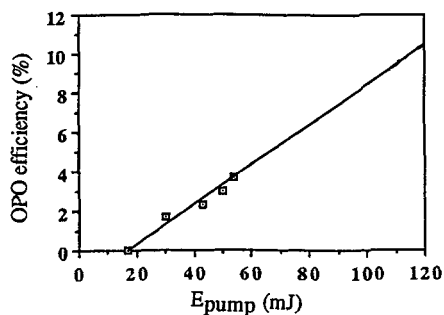


Fig.3 - OPO efficiency with respect to pump energy at 780nm. Solid line is the linear extrapolation calculated from the experimental data.

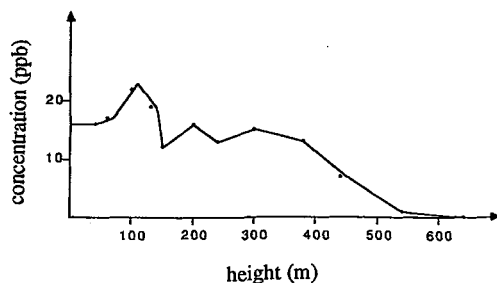


Fig.4 - Toluene vertical concentration profile over the city of Leipzig.

III - Immission toluene measurements

The aromatic hydrocarbons all absorb light in the 250nm region due to the resonance phenomena that take place in these compounds. We measured these ones before having entirely developed the OPO. We worked on the lidar station in Leipzig during summer 1993. The lidar system is in a container on the top roof of a building at a height of 110m above ground.

The system was a conventional lidar: a dye laser pumped by an Eximer laser was followed by a BBO crystal to do second harmonic generation and get near ultraviolet light around 260nm. The dye was Coumarin 307. The observation direction was fixed by the orientation of a flat mirror on the roof of the container. The backscattered light was focused on the photomultiplier by a Cassegrainian receiving telescope. Then the data were treated by a DEC 5000 workstation which gave automatically the concentrations. $\lambda_{\text{off}} = 266.1\text{nm}$ and $\lambda_{\text{on}} = 266.9\text{nm}$ gave the best differential absorption coefficient [7]. There is an interference with ozone at these wavelengths but a very low ozone concentration was measured (about 5ppb) because of cold weather and work by night. Milton mentioned a possible interference with oxygen but, however, our results don't show that.

The vertical concentration profile we obtained is presented in fig.4. These results sound very reasonable: an average value of 15ppb below 500m of height is in good agreement with values found in others works by standard methods [11]; one can also see a zero concentration above 500m, which is the general situation for tropospheric pollutants (this is the height of the boundary layer). Milton had already measured toluene concentrations in the ppm range near a tank fill area that is in emission conditions[7]. This constitutes the first toluene lidar measurements in immission conditions. These results are very promising and open a new area to lidar.

We intend to improve this results in the next year: we want to correlate the toluene concentration with traffic; results will be compared with values obtained by a standard GCMS spectrometer.

IV - Conclusion

We have developed a new Ti:Sapphire based lidar, combining the wide tunability of the laser medium with the ease of operation, characteristic for the flashlamps-pumped solid state lasers. This will allow us to do routine measurements more easily.

On the way to hydrocarbons detection, we realised the first toluene immission concentration profile and we developed a new no-tracking NCPM-OPO as a near infra-red source.

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