Sensitive broadband mid-IR Cr^{2+}:ZnS-laser-based spectrometer

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Abstract: We report a Cr^{2+}:ZnS laser for broadband ultrasensitive intracavity laser spectroscopy, with effective absorption path up to about 50 km in the 2.4 μm range. The spectrometer operates with both Er-fiber and direct diode pumping.

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Intracavity Laser Absorption Spectroscopy (ICLAS) is known since 1970 and is an extremely sensitive technique for the detection of low concentrations of atomic and molecular gases. Besides fundamental sciences such as spectroscopy, chemistry, and astronomy, it can be also applied in technology for process monitoring and in environmental control for trace gas sensing and remote control, and even in medicine for various chemical diagnostics and breath analysis. Among the existing methods of sensitive gas detection, intracavity laser absorption spectroscopy (ICLAS) offers some distinct advantages such as the simultaneous coverage of a broad spectral domain and large dynamic range[1]. Under ICLAS the absorbing medium is put inside a laser cavity with broadband gain As a result, the laser cavity acts as a multipass cell. Equivalent absorption path length of several tens of kilometers can be achieved, corresponding to the high detection sensitivities of the order of 10^{-8} cm^{-1} and better.

Only few examples of ICLAS spectrometers were demonstrated beyond 2 μm (for details see e.g. [2] and references therein), including KCl:Li Fa(II) color center laser with coverage up to 4 nm around 2638 nm, Co:MgF2, coveraging up to 30 nm around 2040 and 2245 nm, Tm:YAG with coverage up to 35 nm around 2030 nm, and pulsed Cr:ZnSe, with coverage up to 50 nm around 2500 nm. Recently we significantly broadened [3] the simultaneous coverage range (up to 125 nm around 2500 nm) by applying a tunable Cr:ZnSe laser for ICLAS measurement of weak CO_2 and C_2H_2 [5] lines. The CO_2 lines were recorded for the first time in laboratory and could be previously observed only in the Venus atmosphere.

Cr:ZnSe is representative of a whole family of similar laser materials [5], allowing central wavelength selection by compositional tuning. Another member of this family, Cr:ZnS has similar spectroscopic and laser properties. It proved to be a viable competitor to Cr^{2+}:ZnSe, demonstrating high power (0.7 W) broadband (over 700 nm tunability) CW operation under both, Er-fiber and direct diode-pumping [6]. Relatively to Cr:ZnSe, Cr:ZnS is distinguished by the blue-shifted (by 100 nm) emission peaking around 2.3 μm, making it especially attractive for ICLAS applications in the water-free window of the atmosphere.

In this paper, we report first application of a Cr^{2+}ZnS laser to spectroscopic purposes. We demonstrate an evacuated Cr^{2+}:ZnS laser for high-resolution and high-sensitivity intracavity spectroscopy in the water free region, utilizing Er-fiber and direct diode pumping. The spectral coverage was extended to 2360 nm, allowing to record very weak difference bands of N_2O with Doppler-limited resolution.

The experimental set-up is shown in Fig. 1. For pumping, we used either a Er^{3+}-fibre laser (IPG Photonics) with an acousto-optic modulator as a fast chopper, or a 100-μm wide diode array, providing up to 1 W diode at 1.85 μm. The laser crystal was a single-crystal diffusion-doped Brewster-cut Cr^{2+}:ZnS sample of 2.4 mm length, absorbing more than 90% of pump radiation at 1607 nm and more than 80% at 1.85 μm. The Cr^{2+} laser was assembled on a 60x40 cm board that has been put into a sealed chamber with windows for the pump and output radiation. The laser could be operated under secondary vacuum or filled with a gas of interest. Tuning was achieved by a micrometer-thin nitrocellulose pellicle that could be externally rotated by a stepping motor. The laser output is analyzed by the high-resolution stepping-mode time-resolved Fourier-transform interferometer, equipped with CaF_2 beam-splitter and InSb detectors.

In the present work, absorption N_2O spectra centered at 2400 nm, with pressures of 33 hPa, were recorded with an unapodized resolution equal to 0.006 cm^{-1}. The laser dynamics is sampled by 128 time-components spectra. The time between two consecutive time-components is 3.2 μs. The laser signal in each time-component here is made of 10,000 spectral elements.

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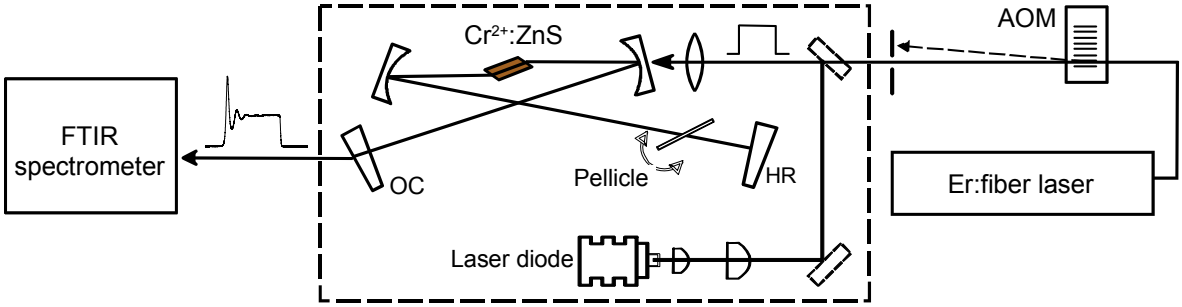

Fig. 1. Schematic diagram of the time-resolved FT spectrometer. The dashed rectangle shows the vacuum chamber. The overall length of the laser cavity was about 70 cm, allowing resolution better than 0.01 cm^{-1}.

Figure 2 shows a portion of a typical spectrum after Fourier-transform processing. The absorption is as high as 50 % for the strongest lines. Minimum detectable absorption coefficient on this spectrum is of the order of $4x10^{-8}$ cm^{-1}. It should be noted, that the explored spectral domain is the location of unidentified weak vibration-rotation bands of N_2O which are not reported in the HITRAN database [7].

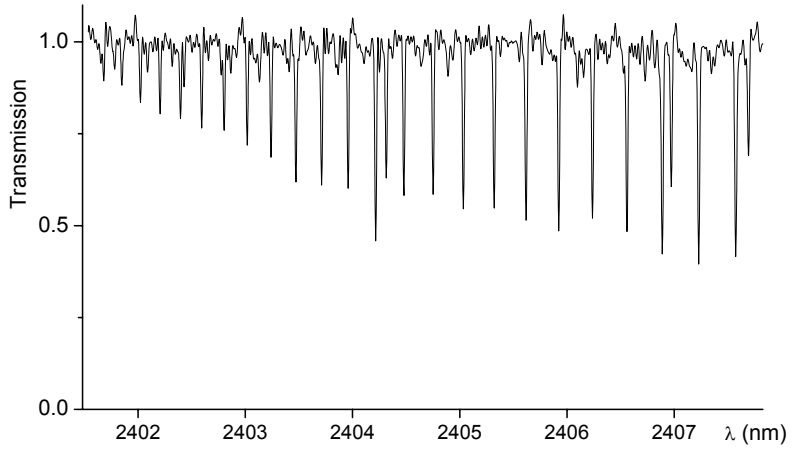

Fig. 2. Portion of a Doppler-limited spectrum of N_2O at 10.5 km effective absorption path length.

In conclusion, we demonstrated a high-resolution mid-IR intracavity spectrometer based on Cr^{2+}:ZnS laser, covering a spectral range of up to 22 nm in a single experiment. The sensitivity of $4x10^{-8}$ cm^{-1} at Doppler-limited resolution allows obtaining spectral information that was previously unreachable in laboratory conditions. We were also able to implement direct diode-pumping in this spectrometer. Such a compact and low cost spectrometer would bring the ICLAS spectroscopy closer to the real-world applications.

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