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Spectroscopy of \textit{DCN} in a molecular beam using a \textit{YLF} laser at 1.9 \(\mu\text{m}\)

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Abstract - A newly developed tunable \textit{YLF} laser operating in the 2 \(\mu\text{m}\) wavelength region demonstrates an output power of about 300 mW in a single longitudinal mode, combined with a high spectral purity. It has been employed for measurements of the first overtone spectrum of \textit{DCN} in a molecular beam with bolometric detection, achieving a signal-to-noise ratio larger than 1000.

The development of tunable solid state lasers has provided a new class of coherent sources for the near infrared region between 1 and 3 \(\mu\text{m}\) wavelength. Very interesting is the wavelength interval around 2 \(\mu\text{m}\) where several authors studied the operation of devices based on the \(^{5}I_7\) - \(^{5}I_8\) transition of Holmium in different hosts. The laser effect was obtained both in pulsed and \textit{cw} regime and successful operation was demonstrated both at liquid nitrogen and at room temperatures [1 - 8]. The perspective applications of these devices are very varied: they are expected to be useful for lidar measurements [9] and for medical applications, as well as for fine spectroscopic research in the near infrared.

In this paper we describe the development and the first application of a tunable solid state laser of "spectroscopic quality" to measurements in the 2 \(\mu\text{m}\) region on a molecular beam. The beam environment is particularly appealing (and challenging) for high resolution molecular spectroscopy because the Doppler and the collisional broadening processes are usually negligible.

We improved the performances of a previously developed prototype of \textit{Er,Tm,Ho:YLiF}_4 laser and we obtained a tunable, narrowband, single-mode source capable of a relatively
large power emission near 2 \( \mu \text{m} \). The laser structure is based on a three-mirror astigmatically-compensated resonator. This scheme allows a tight focusing inside the active medium and a quasi plane-wave propagation in the long arm of the cavity, where optical elements can be placed for line and mode selection. In this way the insertion losses of the tuning elements can be strongly reduced.

The active medium is a 2.5 mm thick crystal of \( YLiF_4 \) (YLF) doped with 91\% of Er, 7.5\% of Tm, and 1.5\% of Ho. The crystal is kept at liquid-nitrogen temperature in a cryostat which is coupled to the resonator through two 0.1 mm thick Brewster windows. This approach allows keeping the folding and the end mirrors outside of the vacuum system, so simplifying their alignment and servicing. If compared to the more conventional solution with both mirrors inside the cryostat, this scheme is more practical, and the losses introduced by the additional optical windows are negligible.

The crystal is pumped by the 514 nm green line of an Ar\(^+\) laser, which is fed into the IR cavity through a dichroic beam splitter placed at Brewster angle (for the infrared radiation). The pump line is resonant with the \( Er^{2H_{11/2}} \rightarrow 4I_{15/2} \) transition and a fast energy-transfer \([11]\) mechanism (from Er to Tm to Ho) supports the laser emission of either Tm (near 1.92 \( \mu \text{m} \)) or Ho (near 2.06 \( \mu \text{m} \)). The tuning is achieved by a grating, placed in a first-order Littrow configuration, which terminates the long arm of the cavity, and the power is extracted from the zero-th order diffracted beam. To increase the fraction of extracted power we have mounted the grating with the grooves parallel to the polarization of the laser field inside the cavity; in this way the outcoupling is about 30\%, while in the usual configuration is about 6\%. This was possible because the optical gain of the active crystal is very high \([12]\). A 6 mm thick low-finesse solid etalon is used as a mode selector. In order to remove the hole burning mode and to obtain a single mode operation, we used a 50 mm long air spaced etalon.

All the optical elements and the cryostat are locked to a rigid frame providing good mechanical and thermal stability. This is composed of a three superinvar bars, about 1 meter long, clamped to five aluminum plates; the bar configuration forms a nearly equilateral prism.

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**Fig. 1** Experimental apparatus
We obtained about 1.4 W and 1 W multimode in the 2.06 \( \mu m \) and 1.92 \( \mu m \) regions respectively, at 3.2 W pump power and 30% extraction. In single mode operation the emitted power at the peaks of the tuning intervals is reduced to the half of these values. The estimated frequency jitter of the free-running laser (i.e. without any active frequency-stabilization feedback loop) is less than 1 \( MHz \).

Our goal is to realize a fully computer-controlled high resolution spectrometer in the 2 \( \mu m \) region, by combining the laser with a molecular beam apparatus [13] and a frequency-control data-acquisition system. The tuning can be achieved by changing the cavity length by a piezo element holding the folding mirror, and letting the computer change the alignment of the two etalons and the angle of the grating.

In order to test the laser performances before starting in with the complete frequency control devices, we wanted to make some preliminary absorption measurement on a convenient molecular transition. We decided to remove the 50-mm etalon, so allowing the presence of two hole-burning modes, and to limit our wavelength sweeps to a narrow region. In this approach the frequency-control electronics is simply a HV amplifier driving the piezo of the folding mirror.

The laser beam is split into two parts, one is for frequency measurements, while the other is fed into a multipass cell surrounding the molecular beam. A schematic diagram of the experimental set up is reported in Fig. 1.

![Diagram of experimental setup](image)

**Fig.2** Bolometer signal versus frequency offset. The vertical scale is in \( \mu V \)
In this configuration we were able to observe an absorption signal (actually an optothermal signal) from the first overtone of the C-D stretch of DCN. The transition displayed in Fig. 2 is the R(0) line at 5222.70(1) cm\(^{-1}\). The relevant experimental conditions are: chopping frequency 280 Hz, scan rate 1.5 MHz/sec, IR laser power at the molecular beam 150 mW, detection time constant 1 sec, bolometer noise 76 nV rms, maximum signal 83 pV.

**References**