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Application of a continuous-wave tunable erbium-doped fiber laser to molecular spectroscopy in the near infrared

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ABSTRACT Development of a continuous-wave tunable fiber laser-based spectrometer for applied spectroscopy is reported. Wide wavelength tunability of an erbium-doped fiber laser (EDFL) was investigated in the near-infrared region of 1543–1601 nm. Continuous mode-hop free fine frequency tuning has been accomplished by temperature tuning in conjunction with mechanical tuning. The overall spectroscopic performance of the EDFL was evaluated in terms of frequency tunability along with its suitability for molecular spectroscopy. High-resolution absorption spectra of acetylene (C_2H_2) were recorded near 1544 nm with a minimum measurable absorption coefficient of about $3.5 \times 10^{-7} \text{ cm}^{-1}/\text{Hz}^{1/2}$ for direct absorption spectroscopy associated with a 100-m long multi-pass cell. Detections of C_2H_2 at different concentration levels were performed as well with high dynamic detection range varying from 100% purity to sub ppmv using cavity ring down spectroscopy. A 3σ -detection-limited minimum detectable concentration (MDC) of 400 ppbv has been obtained by using the transition line $P_e(22)$ of the $\nu_1 + \nu_3 + \nu_5^1(\Pi_g) - \nu_5^1(\Pi_u)$ hot band near 1543.92 nm with a detection bandwidth of 2.3 Hz. This corresponds to a minimum detectable absorption coefficient of $6.6 \times 10^{-11} \text{ cm}^{-1}/\text{Hz}^{1/2}$. The sensitivity limit could be further improved by almost one order of magnitude (down to ~ 60 ppbv) by use of the $P_e(27)$ line of the $\nu_1 + \nu_3(\Sigma_u^+) - 0(\Sigma_g^+)$ combination band near 1543.68 nm.

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1 Introduction

During the last decade, the need for optical telecommunication, e.g., dynamic DWDM (dense wavelength division multiplexing) optical networks, has spurred the development of continuous-wave (cw) tunable lasers in the near infrared (NIR) around 1.55 μm . Today these devices are finding an increasing number of applications not only in the telecommunication domain, but also in high-resolution molecular spectroscopy and atmospheric/environmental spectroscopy. The widely tunable laser technologies available include:

temperature-tuned distributed-feedback (DFB) lasers [1–7]; current tuned sample-grating distributed Bragg reflector (SG-DBR) lasers [8, 9]; MEMS (micro-electro-mechanical system) tuned external cavity diode lasers (ECL) [10]; electro-mechanically tuned vertical cavity surface emitting lasers (VCSEL) [11] and rare earth doped fiber laser [12, 13]. Although the mid-infrared (MIR) is usually more attractive for ultra-sensitive trace gas spectroscopy (owing to the strong absorptions of fundamental bands/modes, whose strength is typically of 2–3 orders of magnitude higher than absorptions of overtone and combination bands in the near infrared) and the novel MIR quantum cascade lasers (QCL, [14]) are now commercially available, widely tunable lasers operating in the near infrared are still the best option for many field applications. This is mainly because of the ready availability of fiber-optical components from the telecommunication industry, and of low-cost cw lasers with output power in the mW-to-W range with room temperature operation.

In this paper, we describe the development of a near-infrared spectrometer based on a cw continuously tunable erbium (Er^{3+}) doped fiber laser (EDFL) for applied spectroscopy. The EDFL source used in the present work (ELT-1-CL-SF-LP, IPG LASER GmbH) is designed in a master-oscillator/power-amplifier (MOPA) configuration in order to obtain single longitudinal mode operation and narrow linewidth ($< \text{MHz}$) with high continuous-wave output power (up to 1 W). The EDFL emits in the 1543–1601 nm interval over the fluorescence spectrum of the Er^{3+} -doped active fiber. As a suitable cw spectral source for application to high-resolution laser spectroscopy, it requires: (1) single longitudinal mode operation with narrow linewidth; (2) high signal-to-background ratio (in the case of fiber laser or fiber amplifier-based laser source, the “background” is generated rather by the “Amplified spontaneous emission”, denoted as ASE); and (3) wide frequency tunability without mode hopping. Study of the spectral characteristics of the EDFL and realization of mode hop free frequency tuning (up to $2\text{--}3 \text{ cm}^{-1}$) has been performed (presented in Sect. 2 and 3). Spectroscopic application of the EDFL source was demonstrated (in Sect. 4) using high-resolution molecular spectroscopy and trace gas spectroscopy of acetylene (C_2H_2). Measurements of C_2H_2 at different concentration levels were performed with high dynamic detection range varying from 100% purity to sub ppmv using cavity ring down

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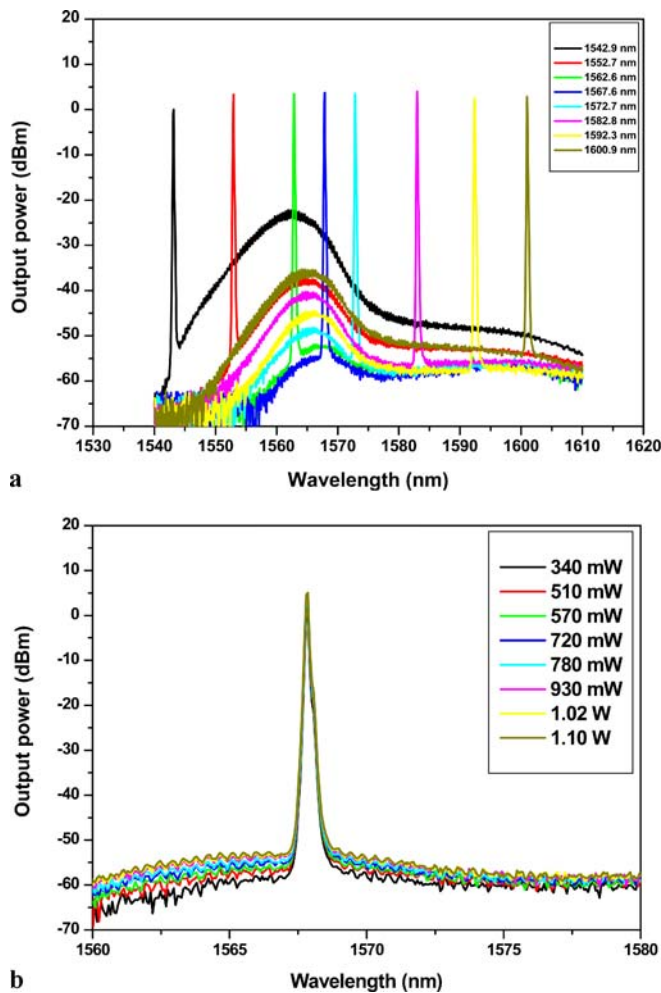


FIGURE 1 (a) Emission spectra of the used EDFL, recorded over ~ 60 nm using an optical analyzer (ANDO-AQ-6315 A) with a resolution of 0.1 nm. The pump diode current is fixed at 0.41 A, which corresponds to an output power varying from 350 mW to 700 mW depending on the laser emission channel. As shown in the recorded spectra, the ASE contribution to the signal-to-ASE ratio is different for each channel, while the signal-to-ASE ratio is almost constant and equal to about 58 dB in the whole spectral coverage. (b) Dependence of the signal-to-ASE ratio on laser output power. As can be seen, the ASE doesn't significantly affect the signal-to-ASE ratio, an almost constant value of ~ 58 dB was observed

spectroscopy. The 3σ -detection-limited minimum detectable concentration (MDC) is 400 ppbv with a detection bandwidth of 2.3 Hz.

2 Spectral characterization of the EDFL source

In order to determine the signal-to-ASE background ratio, the cw EDFL emission spectra were studied by use of eight emission channels over ~ 60 nm using an optical analyzer (ANDO-AQ-6315 A) with a resolution of 0.1 nm. In this experiment, the diode pump current was fixed at 0.41 A, which corresponded to an EDFL output power varying from 350 mW to 700 mW depending on the laser emission channel. As can be seen in Fig. 1a, the ASE contribution to the signal-to-ASE ratio is different for each channel. Maximum and minimum ASE levels have been observed at the 1542.9 nm channel and the 1567.6 nm channel, respectively. However,

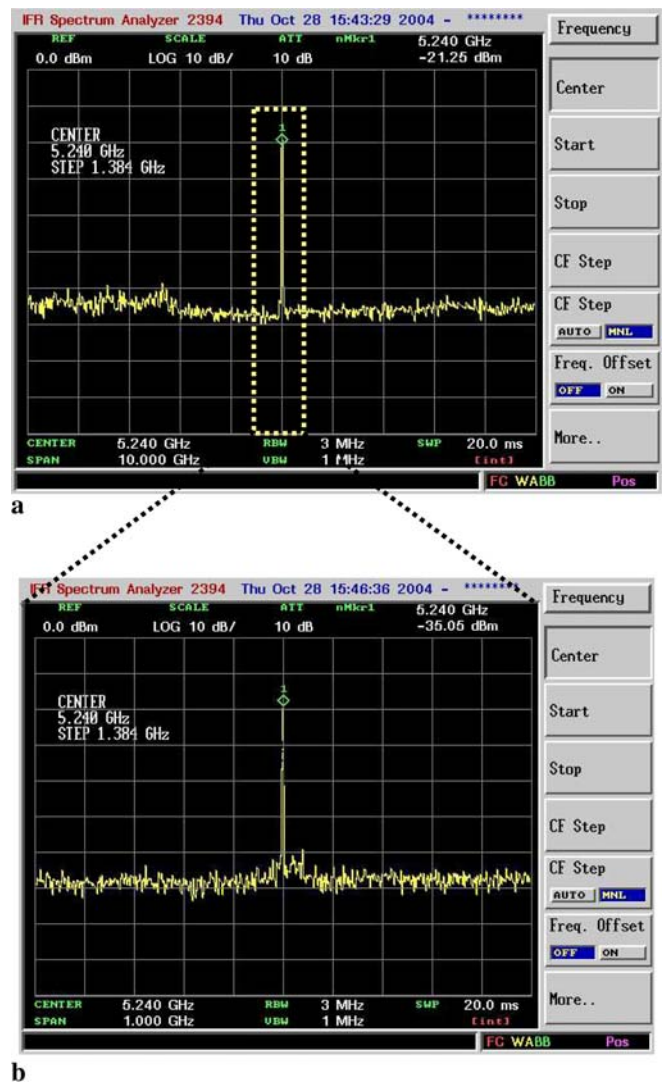


FIGURE 2 Heterodyning measurement spectra: the EDFL output was mixed with a fiber coupled external cavity telecommunication laser. The single longitudinal mode operation was confirmed. The linewidth of the beat signal was found to be ~ 10 MHz. (a) spectrum recorded over 10 GHz, and (b) over 1 GHz

the signal-to-ASE ratio is almost invariant: ~ 58 dB for each channel in the whole spectral coverage of 1543–1601 nm.

We then studied the influence of the laser output power on the signal-to-ASE ratio. In Fig. 1b, the laser output powers were increased by varying the diode pump current from 0.07 A to 0.74 A, corresponding to a power variation from 340 mW to 1.10 W. As can be seen, the ASE does not significantly affect the signal-to-ASE ratio, an almost constant signal-to-ASE level of 58 dB was observed.

Study of the spectral linewidth and single longitudinal mode operation was performed by use of a high resolution rf spectrum analyzer associated with heterodyning measurement. The EDFL output was mixed with a fiber coupled external cavity telecommunication diode laser (ANDO-AQ-4320 B) in a fast InGaAs detector (TTI-TIA-1200) with a bandwidth of 10 GHz. The beat spectrum was recorded with a rf spectrum analyzer (IFR 2394, with a pass band of 9 kHz to 13.2 GHz). Figure 2 shows a beat note signal cen-

tered at 5.240 GHz, recorded with a resolution of 1 MHz. The linewidth of the beat signal was found to be ~ 10 MHz. As the spectral purity of the mixed diode laser was not accurately known, it is not possible to precisely determine the linewidth of the EDFL, but a value lower than 10 MHz can be expected. Accurate measurements of the linewidth could be performed by heterodyning against a suitable laser source with well known and narrower linewidth, which is not available at the present time. According to the manufacturers, the spectral linewidth of the used EDFL is ~ 500 kHz (FWHM).

3 Frequency tuning of the EDFL source

One of the most important spectral features of a laser source for spectroscopic application is its frequency tunability. As the used EDFL source was originally designed for telecommunication application, study and realization of mode hop free frequency tuning with high-resolution was performed at first. In fact, the spectral coverage of 1543–1601 nm is divided into 72 channels, defined by the resonance of a tunable filter, with a spacing of 100 GHz (~ 0.8 nm) that is referenced to the ITU-WDM 100 GHz grid. A discretely tunable Bragg grating with a bandwidth of 100 GHz (~ 0.8 nm) is used for channel selection via mechanical tuning with a stepper motor. Specific channel tunability is achieved by tuning the Bragg grating so that the EDFL output frequency is coincident with a transmission peak of the tunable filter.

Fine frequency tuning of $2\text{--}3\text{ cm}^{-1}$ (~ 0.8 nm) across each channel can be made by varying the temperature of the tunable filter. Temperature tuning will modify the free spectral range of the filter across the bandwidth of the Bragg grating with a step of one arbitrary unit of filter temperature value (denoted as FTV for “filter temperature value”), which corresponds to a frequency tuning resolution of ~ 10 MHz. Fig. 3a shows a wavelength scan of ~ 0.8 nm ($\sim 3.27\text{ cm}^{-1}$) across the 1544 nm channel by varying the tunable filter temperature. The temperature adjustment was made from -1000 to $+1000$ arbitrary units of FTV. Wavelength measurements were made using a high-precision wavemeter (Burleigh WA-1500) with an absolute accuracy of ± 0.2 ppm (e.g., 3×10^{-4} nm at 1550 nm). As can be seen, temperature tuning of the tunable filter over the full channel bandwidth of ~ 100 GHz results, however, in a series of continuous portion of less than 1 GHz, separated by a mode hopping gap of about 1.5 GHz. Based on the study carried out in the present work, we found that the temperature tuning of the diode pump allowed a mode hop free frequency scan of ~ 1.5 GHz (corresponding to a temperature variation of $\sim 3^\circ\text{C}$) with a frequency tuning resolution of ~ 50 MHz (Fig. 3b), which can be used for any filter temperature value. Diode temperature tuning was thus used over the mode hopping gap between two continuous scans. Continuous frequency scan inside each channel can thus be achieved by alternation of the temperature tuning of the tunable filter and the diode. A continuous scan of $\sim 1\text{ cm}^{-1}$ around 1544 nm is shown in Fig. 3c. The fine frequency tuning was controlled with a LabView-based software.

In order to overlap two successive channels, one can adjust finely the position of the Bragg grating via a stepper motor. Figure 4a shows wavelength scans realized by vary-

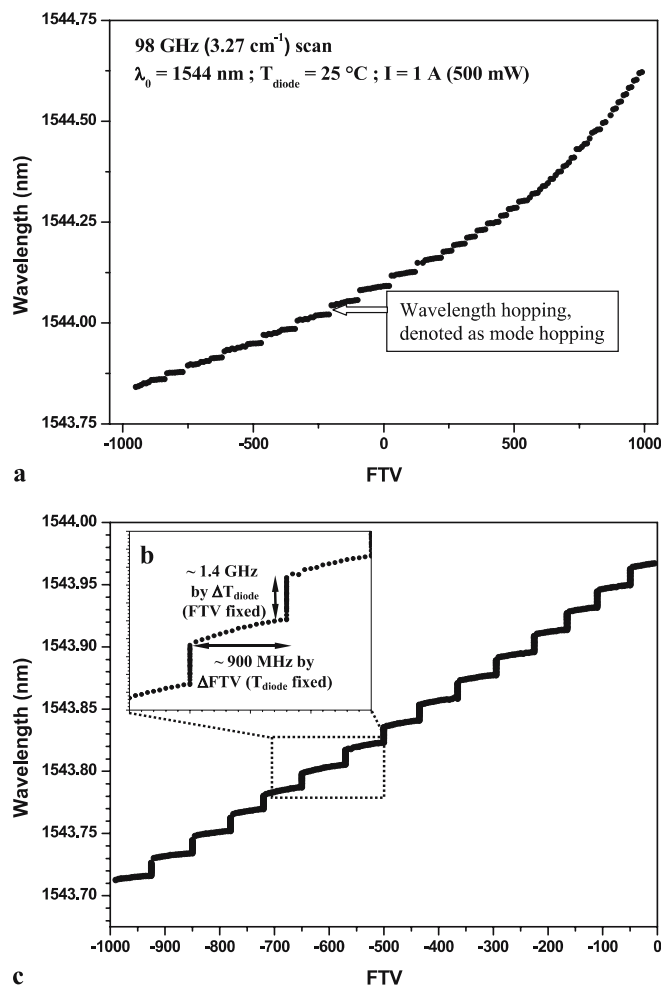


FIGURE 3 (a) Frequency scan of $\sim 3.27\text{ cm}^{-1}$ (98 GHz) across the channel bandwidth by varying the temperature of the tunable filter for the 1544 nm channel. The temperature adjustment was made from -1000 to $+1000$ arbitrary units of FTV (“filter temperature value”), which results in a series of continuous portion of less than 1 GHz, separated by a mode hopping gap of about 1.5 GHz. (b) Alternation of temperature tuning of the filter and the diode. (c) Continuous scan of $\sim 1\text{ cm}^{-1}$ by combination of the filter temperature tuning with diode temperature scanning

ing FTV from -1000 to 1000 for two successive channels (1590.6 nm and 1591.4 nm) with the Bragg grating fixed at a normal position (by default) $M = 0$. As can be seen, there is a gap of ~ 22 GHz between two successive channels. By adjusting finely the position of the Bragg grating with $M = +10$ for the 1590.6 nm channel and $M = 0$ for the 1591.4 nm channel, as shown in Fig. 4b, the two successive channels were well overlapped. In this manner, continuous frequency tuning in the full spectral range of 1543–1601 nm is realizable by using temperature tuning of the tunable filter and the diode for wavelength scan inside the channel, in combination with mechanical tuning of the Bragg grating for channel selection and channel overlapping.

4 Spectroscopic application

In the wavelength region accessible to the present EDFL, there are numerous molecular species that exhibit vibrational absorptions of overtone and combination bands,

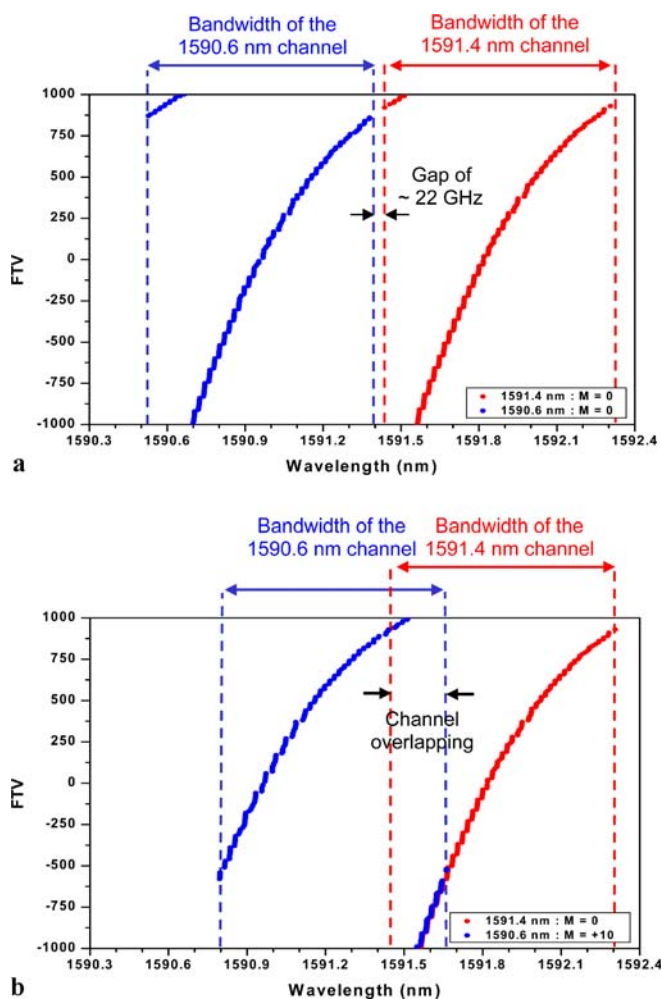


FIGURE 4 Overlapping of two successive channels by finely adjusting the position of the Bragg grating via a stepper motor. **(a)** Wavelength scans realized by varying FTV from -1000 to 1000 for two successive channels (1590.6 nm and 1591.4 nm) with the Bragg grating fixed at a normal position (by default) $M = 0$. As can be seen, there is a gap of ~ 22 GHz between two successive channels. **(b)** By adjusting finely the position of the Bragg grating with $M = +10$ for the 1590.6 nm channel and $M = 0$ for the 1591.4 nm channel, the two successive channels were well overlapped. In this manner, continuous frequency tuning in the full spectral range of 1543 – 1601 nm is realizable by using temperature tuning of the tunable filter and the diode for wavelength scan inside the channel, in combination with mechanical tuning of the Bragg grating for channel selection and channel overlapping

such as CO , CO_2 , C_2H_2 , H_2O , HI , OH , H_2S , HCN , HDO , N_2O , etc. We evaluated the spectroscopic performance of the EDFL source in terms of frequency tunability and spectroscopic measurement sensitivity by means of direct absorption spectroscopy and cavity ring down spectroscopy (CRDS) measurements of acetylene (C_2H_2).

4.1 Direct absorption spectroscopy using EDFL

As depicted in Fig. 5, a widely tunable near infrared absorption spectrometer based on EDFL has been developed in the present work. A LabView-based software was used to control the spectrometer and to process the data (for wavelength scan, data acquisition and treatment). An astigmatic Herriott multipass cell (New Focus 5612) with an optical path length of 100-meter was used for direct absorp-

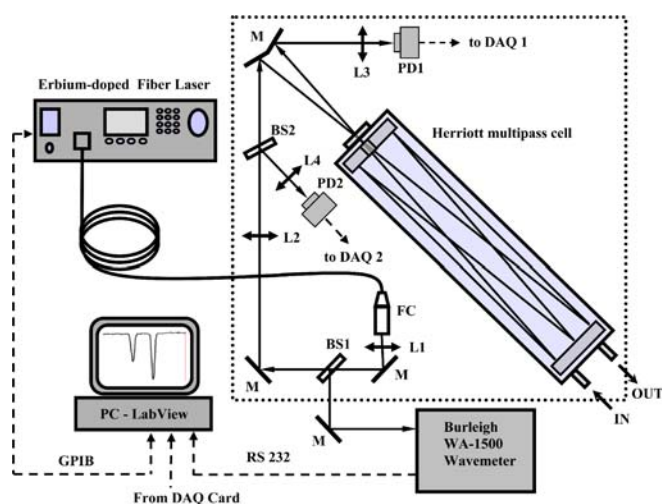


FIGURE 5 Schematic of the experimental apparatus based on near infrared EDFL direct absorption spectroscopy. FC: fiber connector, PD: InGaAs photodiode, BS: beam splitter, L: lens, M: mirror, and DAQ: data acquisition card

tion spectroscopy. The EDFL output beam from the fiber output connector was collimated at first with a short focal length lens ($L1$, $f = 25$ mm). Then the laser beam was divided into two parts with a beam-splitter (BS1): one was used for wavelength measurements by a high-precision wavemeter (Burleigh WA-1500), and the other was directed to the cell by use of a long focus lens ($L2$, $f = 500$ mm) in such a way that the laser beam was focused to a waist at roughly the center of the cell. Laser radiation detection was carried out with two high speed InGaAs photodiodes (DET410, THORLABS, with a $\text{NEP} = 10^{-14}$ $\text{W}/\text{Hz}^{1/2}$): PD1 and PD2 for detection of “spectroscopic absorption signal” and “baseline reference”, respectively. During spectrum scan, the signals were averaged over 1 second for each wavelength with a sampling rate of 100 kS/s with help of a National Instruments acquisition card (Model PCI-MIO16E1). Absorption spectra were recorded simultaneously with wavelength and filter or diode temperature values.

We recorded a portion of acetylene (C_2H_2) absorption spectrum by using the 1543.74 nm channel (ITU channel N°42). The spectrum (Fig. 6, dots), recorded with a post-detection bandwidth of 0.5 Hz over 0.25 cm^{-1} , corresponds to the $P_f(22)$ and $P_e(22)$ absorption lines of the $\nu_1 + \nu_3 + \nu_5^1(\Pi_g) - \nu_5^1(\Pi_u)$ hot band of $^{12}\text{C}_2\text{H}_2$ near 6477 cm^{-1} [15]. The spectrum is baseline corrected. The solid curve shows theoretically calculated Gaussian lineshape.

The direct absorption measurement sensitivity, usually limited by optical feedback, fringes and fluctuations in laser power, can be determined from the measurement of the rms noise feature in the background of the measured spectrum. A minimum detectable absorption coefficient of about 3.5×10^{-7} $\text{cm}^{-1}/\text{Hz}^{1/2}$ was found for the present setup.

4.2 Cavity ring down spectroscopy (CRDS) using EDFL

Measurements of C_2H_2 with different mixing ratio were performed as well using cavity ring down spectroscopy (CRDS). A fiber coupled cw cavity ring down spectrometer was used (Fig. 7), which was similar to that described in detail

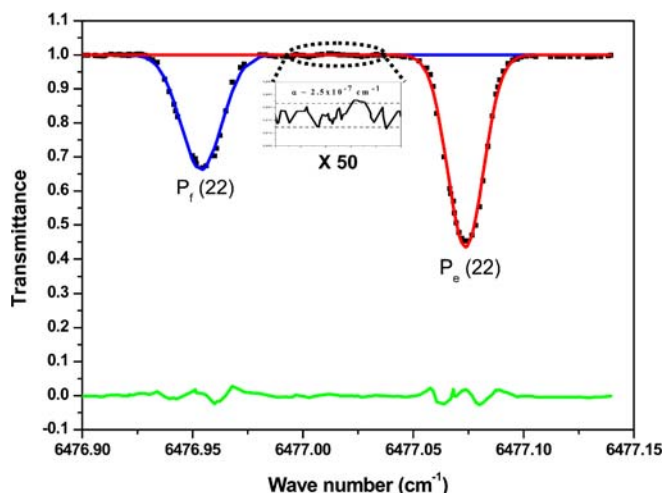


FIGURE 6 Er^+ -doped fiber laser absorption spectrum of $^{12}\text{C}_2\text{H}_2$: 1 mbar pure C_2H_2 in a 100-m long multipass cell, recorded with a post-detection bandwidth of 0.5 Hz over 0.25 cm^{-1} . Dots: experimental data, solid curve: Gaussian lineshape fit and the accompanying residual (lower line)

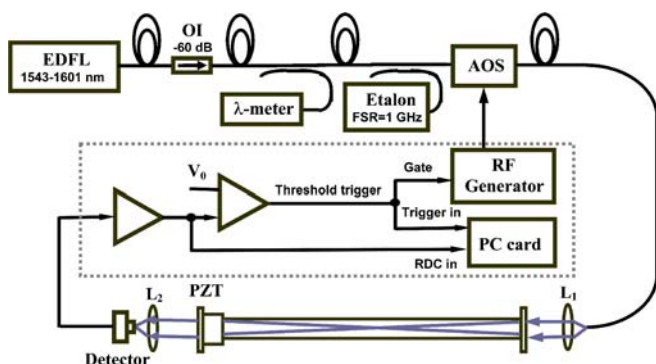
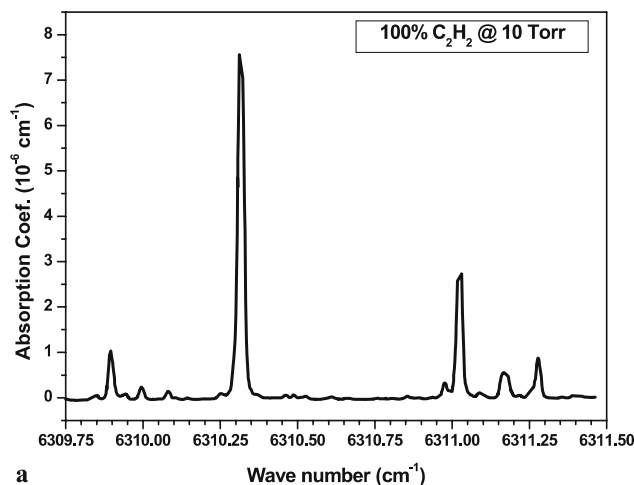


FIGURE 7 Experimental setup of the cw cavity ring down spectrometer used in the present work. AOS: acousto-optic switch, OI: optical isolator, PZT: piezo-electric actuator, L: lens

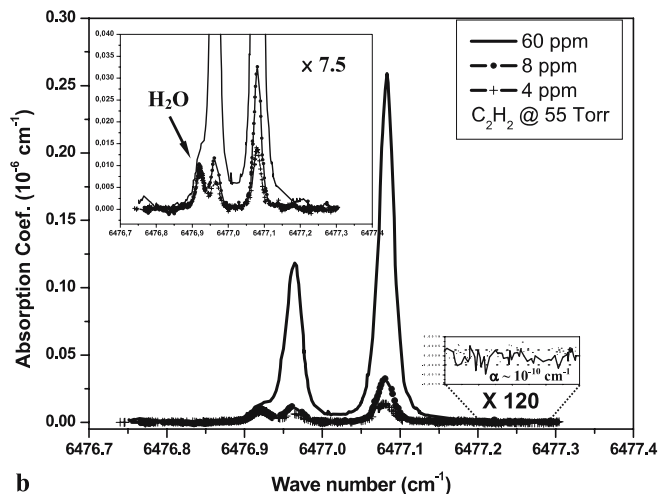
in [16]. In brief, the ring-down cavity (140-cm long) in stainless steel was fitted with a pair of high reflectivity ($\sim 99.99\%$) spherical mirrors ($R = 1\text{ m}$). One of the mirrors can be driven by a piezoelectric actuator (PZT) so that the cavity length can be modulated and so do the cavity resonance frequencies (modes). A fiber coupled acousto-optic switch (AOS) was used to generate ring-down event by shutting off the laser. The ring-down time was about $70\ \mu\text{s}$, corresponding to an effective optical path of $\sim 21\text{ km}$.

About 100 ring-down events were averaged for each point of the spectrum. High dynamic detection range varying from 100% purity to sub ppmv was obtained by use of a strong and weak absorption lines for low and high concentration measurements, respectively. The spectrometer linearity over such a large dynamic range has been checked in such a way that the spectrometer scales linearly with the absorption intensity (that is proportional to the measured gas concentration) for each absorption line used for trace measurements.

Figure 8 shows the CRDS-based measurements of C_2H_2 at different concentration levels: (a) Absorption spectrum of pure C_2H_2 over $\sim 1.75\text{ cm}^{-1}$ around 6310 cm^{-1} . (b) Trace absorption spectra near 6477 cm^{-1} . In trace gas measurements, ambient air was used to dilute C_2H_2 sample.



a



b

FIGURE 8 Wide concentration range detection of C_2H_2 by CRDS: (a) 100% pure C_2H_2 absorption spectrum over $\sim 1.75\text{ cm}^{-1}$ near 6310 cm^{-1} . (b) Trace absorption spectra. The minimum measurable absorption coefficient was about $6.6 \times 10^{-11}\text{ cm}^{-1}/\text{Hz}^{1/2}$ with a 2.3-Hz detection bandwidth, which yielded a 3σ -detection-limited minimum detectable concentration of 400 ppbv using the transition line $\text{P}_e(22)$ of the $\nu_1 + \nu_3 + \nu_5^1 - \nu_5^1$ hot band at 6477.084 cm^{-1} . As displayed in the inset, an absorption line of H_2O vapor in ambient air was detected at 6476.928 cm^{-1} ($021\ 4_{3,1} - 000\ 4_{3,2}$) with an almost constant concentration

In Fig. 8a, the strongest line observed at 6310.3166 cm^{-1} has not been spectroscopically assigned. Based on the measured spectrum shown in Fig. 8a, we deduced a line strength of $6.463 \times 10^{-25}\text{ cm}/\text{molecule}$. This value is obtained from the integrated absorbance area under the measured absorption line.

The detection limit in terms of minimum detectable absorption coefficient was experimentally determined to be about $6.6 \times 10^{-11}\text{ cm}^{-1}/\text{Hz}^{1/2}$ with a 2.3-Hz detection bandwidth. This value, comparable to those previously reported [16, 17] using CRDS approaches, yielded a 3σ -detection-limited minimum detectable concentration (MDC) of 400 ppbv for C_2H_2 using the transition line $\text{P}_e(22)$ of the $\nu_1 + \nu_3 + \nu_5^1(\Pi_g) - \nu_5^1(\Pi_u)$ hot band at 6477.084 cm^{-1} (with a line strength of $6.818 \times 10^{-23}\text{ cm}/\text{molecule}$ [15]). The MDC could be further improved by almost one order of magnitude by using the $\text{P}_e(27)$ absorption line of the $\nu_1 + \nu_3(\Sigma_u^+) - 0(\Sigma_g^+)$ combination band near 6477.942 cm^{-1}

(with line strength of 4.548×10^{-22} cm/molecule [15]) by the present setup.

As displayed in the inset in Fig. 8b, an absorption line of H₂O vapor in air was detected at ~ 6476.928 cm⁻¹ (021 4_{3,1}–000 4_{3,2}) [15] with an almost constant ambient concentration, which could be useful for frequency and even concentration calibration [18].

5 Conclusions

Wavelength tunability of a cw erbium-doped fiber laser has been investigated in the present work. Wide tunability in the whole spectral coverage ranging from 1543 to 1601 nm could be realized across 72 channels by varying the temperatures of the tunable filter and the diode pump in conjunction with mechanical tuning of the Bragg grating. The overall spectroscopic performance of the EDFL was evaluated in terms of frequency tunability along with its suitability for molecular spectroscopy by means of direct absorption spectroscopy and cavity ring down spectroscopy.

The high cw single-mode output power combined with its large frequency tunability, narrow linewidth and wide spectral coverage makes the EDFL an attractive laser source of choice for ultra-sensitive spectroscopic applications such as high-resolution molecular spectroscopy and trace gas detection of multispecies and broadband absorbers, as well as for use as a tunable and powerful pump source in nonlinear frequency conversion applications ([19]).

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