



Highly sensitive Fourier transform spectroscopy with LED sources



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ARTICLE INFO

Article history:

Received 16 April 2013

In revised form 14 June 2013

Available online 3 July 2013

Keywords:

LED

Carbon dioxide

Water vapor

Fourier transform spectroscopy

ABSTRACT

It is shown that the use of high luminance LED emitters as a light source for Fourier transform spectrometers permits to enhance their threshold sensitivity in the visible range by orders of magnitude. Using a 2.5 W Edixeon EDEI-1LS3 emitter in the range of 11,350–11,700 cm^{−1} as a light source for the spectrometer with a 60-cm multipass cell during a 24-h measurement time, we have achieved a signal-to-noise ratio of 4.5×10^4 which corresponds to the minimal detectable absorption coefficient of 1.2×10^{-8} cm^{−1}.

Such enhanced sensitivity spectrometer has been used to measure the transition frequencies of CO₂ vibrational bands 00051–00001 and 01151–01101 in the range of 11,400–11,500 cm^{−1}.

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1. Introduction

The registration of absorption spectra of atmospheric molecular gases in the visible and UV ranges is an important spectroscopic problem. Its importance arises primarily from the role this spectral range plays in the balance of incoming solar radiation and its relevance to the validation of theoretical spectroscopic models [1]. In the visible and UV ranges, the line intensities are very small (10^{-25} to 10^{-28} cm/mol) and in order to detect them, one has to use a spectrometer with a threshold sensitivity as high as 10^{-6} to 10^{-8} cm^{−1}.

Various techniques are available for recording weak spectra in the visible range to enable highly sensitive measurements of the absorption coefficient. The most sensitive laser methods such as Cavity Ring Down Spectroscopy (CRDS), Intracavity Absorption Laser Spectroscopy (ICLAS) or Cavity-enhanced absorption spectroscopy (CEAS) are capable of measuring absorption coefficients as small as 10^{-7} to 10^{-10} cm^{−1} [2–4]. Their main disadvantage is a rather narrow operating spectral range. The Fourier transform spectroscopy remains to be the most frequently used technique for the weak spectrum detection and it is characterized by a wide spectral range and high spectral resolution.

To achieve this high sensitivity, Fourier transform spectrometers employ long multipass absorption cells with a base length exceeding 4 m. Consider, for example, the spectrometers at Kitt Peak Observatory using 4 m base length cell with the total path of 434 m [5]; and in Reims and Tomsk with, respectively, 50 and 30 m bases [6,7]. Such cells are unique and expensive pieces of

equipment and require a large amount of sample gas (up to 10,000 l) to be pumped in. Large dimensions of the cell impair its thermal stabilization properties required for the long-time measurement accumulation and make it difficult to achieve a high signal-to-noise ratio. Naturally, a question arises here: whether it is mandatory to build such unique structures and whether it is possible to gain high sensitivity using smaller cells? We have shown in [8] that a high sensitivity of Fourier transform spectrometers can be reached by combining small cells with high luminance emitters. In this way we have been able to achieve a threshold sensitivity of 10^{-8} cm^{−1} using enhanced luminosity halogen lamps with a 60 cm multipass cell during a 9-day spectrum measurement time [9]. For a 24-h signal accumulation, the obtained signal-to-noise ratio (S/N) has been 16,000.

The sensitivity of a Fourier-transform spectrometer can be significantly improved by increasing the effective path length when a cavity-enhanced absorption method employing broadband light source are used in combination with FTS. The effective absorption path length was improved by a factor of 200 using Xenon lamp with FTS measurement time of 10 h and a spectral resolution of 0.05 cm^{−1} [10]. But the noise in recorded spectra caused by the strongly reduced light level does not allow achieving the maximal S/N enhancement.

The Light Emitting Diode (LED) sources are even much brighter than the halogen lamps. Earlier in 2009, one of the authors [11] demonstrated that even low-power LEDs combined with a 10 m cell enable one to obtain the sensitivity to the absorption coefficient of the order of 10^{-7} cm^{−1}. Later on in [12], we used a medium-power 0.5 W LED to measure the H₂O¹⁸ absorption spectrum with a minimal detectable absorption coefficient of 10^{-7} cm^{−1} and determined the frequencies and absorption line intensities in the region 15,000–15,800 cm^{−1}. The possibility of employing LEDs

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in the Fourier transform spectroscopy was discussed in [13] where it was reported a significant improvement of the S/N ratio in the measurements of Cr^3 lines in ruby.

In the present work we have used a light source with a greater luminance based on a LED source at $11,000\text{--}12,000\text{ cm}^{-1}$. As a result, we have succeeded to achieve a signal-to-noise ratio (S/N) of 45,000 for a 24-h signal accumulation. This S/N ratio corresponds to a Fourier transform spectrometer threshold sensitivity of $1.2 \times 10^{-8}\text{ cm}^{-1}$. To demonstrate the high-sensitivity of Fourier transform spectrometer, we have measured the water and carbon dioxide absorption spectrum in the range of $11,350\text{--}11,500\text{ cm}^{-1}$.

2. Experiment

The threshold sensitivity of a Fourier transform spectrometer (the minimal detectable absorption coefficient), K_{th} , depends on two quantities: the absorption layer path length L , and the ability of the measuring system to detect small changes of the signal ($\Delta I/I$). For example, for the absorption layer length of 1 m and the detection ability $\Delta I/I = 0.001$, the threshold sensitivity K_{th} equals 10^{-5} cm^{-1} . The absorption layer length L is usually treated as the primary enhancement factor that affects the sensitivity, disregarding the fact that a sharp fall of the radiation intensity upon

reflection from the cell mirrors makes the second factor predominant in limiting the spectrometer threshold sensitivity. The quantity $\Delta I/I$ is a reciprocal of the signal-to-noise ratio and is, in its turn, defined by two factors: the source luminance I and the amount of spectral noise. In this case the value of ΔI is the limiting signal still reliably detectable by the measuring device (usually to be taken as a double of the instrument noise $\Delta I = 2N$).

As the number of reflections n increases, so will the absorption layer length $L = nL_0$ (L_0 is the cell length), whereas the radiation intensity will decrease resulting in the change of the ratio $\Delta I/I$. The intensity of the light beam passed through the cell (in the absence of the absorbing gas) is defined by reflections losses on the mirrors and is given by the formula $I = I_0 \cdot R^n$, where I_0 is the intensity of the incident light, R is the reflection coefficient of the cell mirrors and n is the number of reflections. The expression for K_{th} then becomes

$$K_{th} = (1/nL_0) \times (\Delta I/(I_0 \cdot R^n)). \quad (1)$$

The dependence of K_{th} on the number of reflections n for different values of R (0.9, 0.95, 0.98 and 1) is plotted in Fig. 1. For the sake of example, we have chosen the following values for the parameters: the cell base length $L = 100\text{ cm}$, signal to noise ratio of input signal is 2000 and 20,000.

It can be seen from the plot that, as the number of reflections increases, the value of K_{th} will significantly differ from the threshold sensitivity of the spectrometer with a perfect mirror cell ($R = 1$). This sensitivity deterioration is the more prominent; the smaller is the mirror reflection coefficient. At a certain number of reflections, there appears a minimum in the curve depicting the spectrometer sensitivity vs. the number of reflections. This minimum becomes sharper and is observed at smaller values of n , the smaller is the mirror reflection coefficient. It means that, for a given reflection coefficient, increasing the path length by way of increasing the number of reflections, in order to reduce the minimal detectable absorption coefficient, is only expedient up to a certain limit.

It has been shown earlier [7,8] that without accounting for the photodetector sensitivity, an optimal number of reflections n_{op} , after reaching the threshold value $\Delta I/I$, is defined by the equality between the relative optical path increase and the relative intensity decrease

$$n_{op} = 1/(1 - R). \quad (2)$$

Thus, the radiation intensity, which is registered by the instrument photodetector, on par with the absorption layer length, has a significant effect on the threshold sensitivity. Small values of $\Delta I/I$ can be achieved either by increasing the mirror reflection coefficient

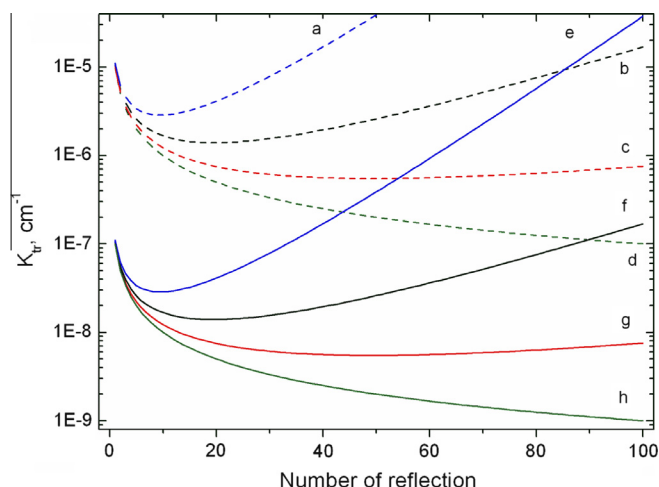


Fig. 1. The dependence of K_{th} on the number of reflections n for different R (0.9 (a), 0.95 (b), 0.98 (c) и 1 (d)) at $S/N = 2000$ and (0.9 (e), 0.95 (f), 0.98 (g) и 1 (h)) at $S/N = 20,000$.

Table 1

Water vapor absorption lines recorded in the range of $11,350\text{--}11,360.6\text{ cm}^{-1}$.

Line pos. (this work)	Intensity [15]	HITRAN 2008 [1]	Toth et al. [5]	Jenuvri et al. [6]	Kassi et al. [3]
11350.3449	3.22E-27	—	—	—	+
11350.4491	8.10E-27	—	—	—	+
11350.5636	6.44E-27	—	—	—	+
11351.4740	3.92E-28	—	—	—	+
11351.6566	2.92E-27	—	—	—	+
11351.8796	1.09E-27	—	—	—	—
11353.5144	4.41E-26	+	+	+	+
11353.7409	1.47E-26	+	—	+	+
11354.1981	3.18E-27	—	—	—	+
11354.8208	1.88E-26	—	—	+	+
11355.0692	4.50E-27	—	—	—	+
11356.1107	2.82E-25	+	+	+	+
11357.4282	1.88E-26	—	—	+	+
11357.5570	4.11E-27	—	—	—	+
11359.7475	6.04E-27	—	—	+	+
11359.8485	2.70E-27	—	—	—	+
11360.6054	1.19E-25	+	+	+	+

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