

Introduction

Gain-switched frequency combs (GSFC) are tunable lasers that are simple in design and significantly cheaper than mode locked femtosecond frequency comb lasers [1]. GSFCs are generated by the direct modulation of a distributed feedback laser with a radiofrequency (RF) sine wave. External optical injection in a master-slave laser configuration is employed to generate narrow line widths and low intensity noise [2]. The approach is straightforward and enables wavelength tunability of (i) the comb position, and (ii) the line spacing (free spectral range), however, with a rather limited number of comb lines (~20+) that typically only span a small wavelength region of a few nm. GSFC lasers were developed as benchtop units for fiber coupled optical telecommunication applications in the L-band in the near infrared [1].

*The **main objective** of this study was to investigate the applicability of a GSFC laser in the near-IR for cavity enhanced absorption experiments using a medium finesse cavity ($f = 450$) without the need for cavity mode matching. The goal was to achieve sensitivities that are sufficient to detect industrial gases at and below their combustion limit.*

The target species was **hydrogen sulphide** (H₂S), a poisonous and corrosive gas that can occur as a pollutant in industrial settings, such as oil refineries, production facilities involving petro-chemistry, gas processing plants and gas fields. Since it is heavier than air it can accumulate in badly ventilated areas and constitute a fire or explosion hazard due to its flammable nature [3]. Its lower explosive limit (LEL) is 4 percent by volume. Thus the spectroscopic detection of H₂S in industrial applications is of high relevance and motivated its usage in this GSFC application.

Fourier Transform Incoherent Broadband CEAS (FT-IBBCEAS)

Before matching of frequency comb lines with absorption features of hydrogen sulfide, the near-IR spectrum of H₂S was measured using FT-IBBCEAS [4] in the region between 6000 and 6600 cm⁻¹ (see Fig. 1). These measurements utilised a super-continuum laser as a light source, a 6.44 m high finesse ($R > 0.9998$) optical cavity, and a Fourier transform spectrometer (Bruker Vertex 80) for signal detection.

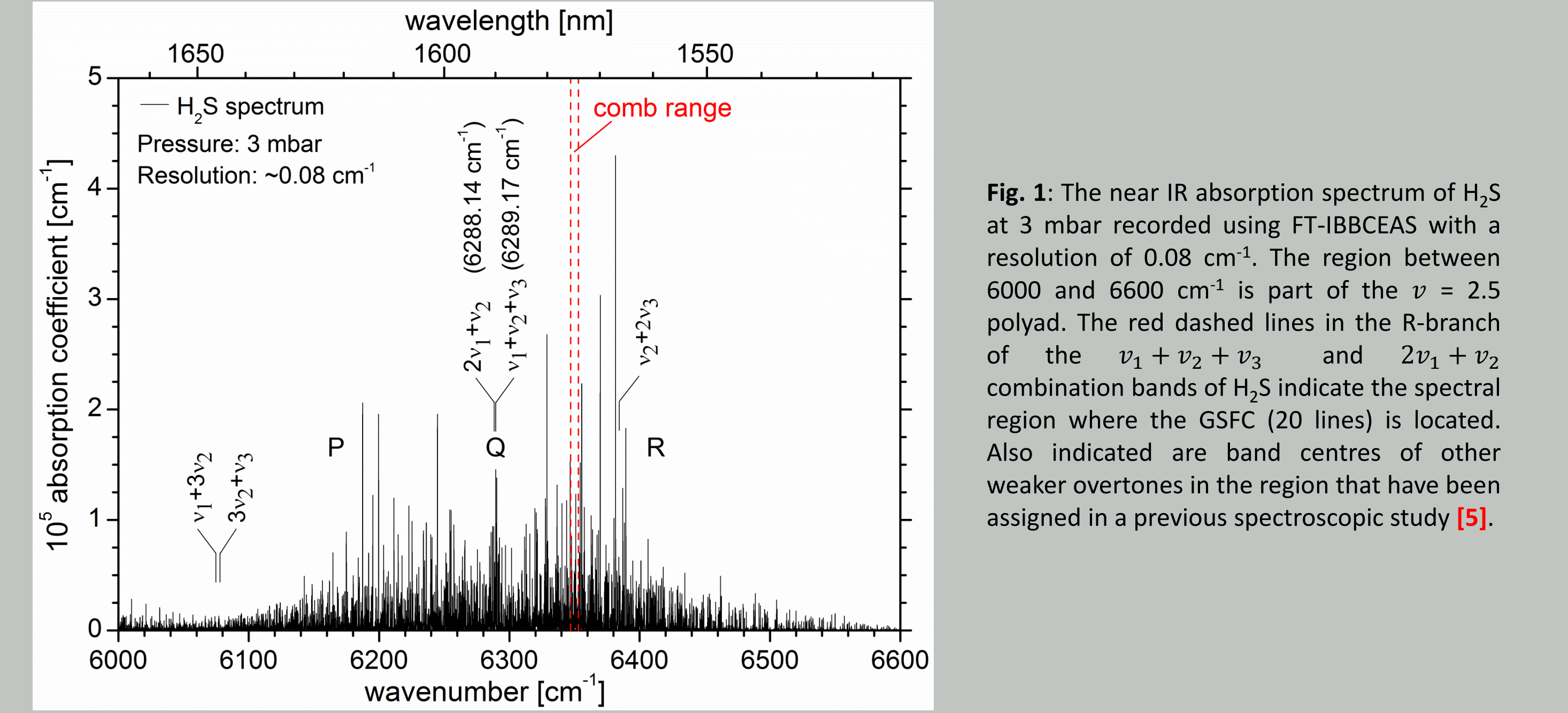


Fig. 1: The near IR absorption spectrum of H₂S at 3 mbar recorded using FT-IBBCEAS with a resolution of 0.08 cm⁻¹. The region between 6000 and 6600 cm⁻¹ is part of the $\nu = 2,5$ polyad. The red dashed lines in the R-branch of the $\nu_1 + \nu_2 + \nu_3$ and $2\nu_1 + \nu_2$ combination bands of H₂S indicate the spectral region where the GSFC (20 lines) is located. Also indicated are band centres of other weaker overtones in the region that have been assigned in a previous spectroscopic study [5].

Gain Switched Frequency Comb CEAS

Fig. 2 shows the schematic of the experimental setup used. The major components of the GFSC laser source are a tuneable master laser (Hewlett-Packard), a slave DFB laser (Thorlabs), an RF signal generator (Hewlett-Packard-83751A), a biased RF amplifier (Aldetec), a polarization controller and a circulator.

The slave laser is kept at a stable operating current of ~45.30 mA through a 10.30 k Ω resistance to achieve a stable operating temperature. The wavelength of the master laser is tuned to a frequency that results in fixed positive detuning from the lasing mode of the slave laser. The polarization controller will retain the polarization state of light injected from the master laser with the optical waveguide of the slave laser. The injected power from the master laser to the slave is optimized at ~5 dBm. Gain switching is achieved by continuously driving the slave laser with a largely amplified RF sinusoidal signal below and above threshold creating a pulse train which corresponds to an optical frequency comb in the time domain. Tuning of the output wavelength of the GSFC is provided by tuning the wavelength of the master laser. The free spectral range (FSR) of the comb is modified by varying the frequency of the RF signal generator driving the slave laser.

The GSFC laser produced 20 phase-coherent, equally spaced comb lines with an individual bandwidth of ~300 kHz covering a range from 6346 to 6354 cm⁻¹. It was tunable over ~5 cm⁻¹ in minimum steps of 0.004 cm⁻¹. The FSR of the GSFC was also tunable but for the experiments fixed at ~10 GHz. Intensity stability <1% (1hr).

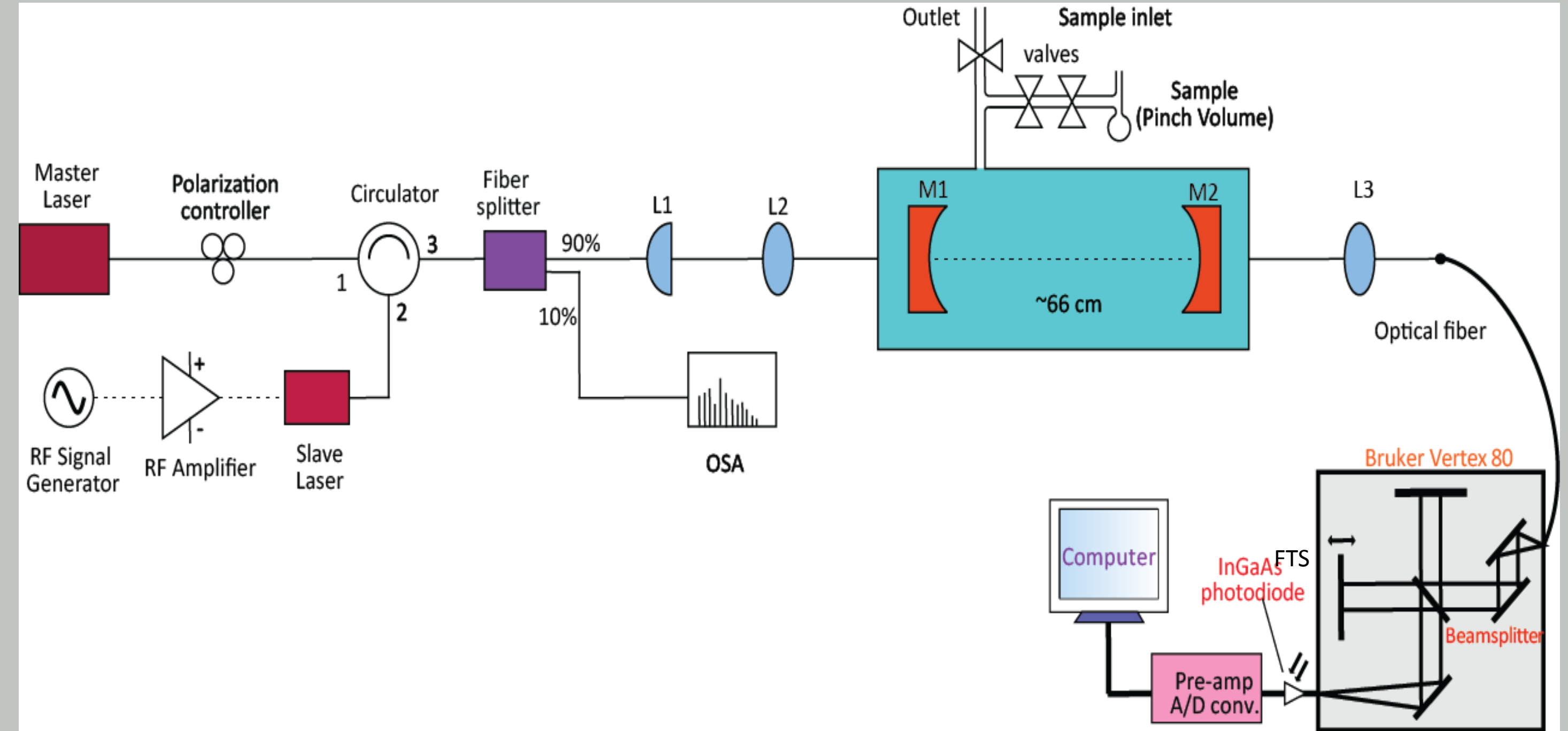


Fig. 2: Schematic of the experimental setup. M1, M2: dielectric mirrors ($R \sim 0.993$), finesse ~ 450 . L1, L2: collimation lenses. L3: achromatic doublet to focus light transmitted by the cavity into an optical fibre. The Fourier Transform Spectrometer was the same as that used in the FT-IBBCEAS measurements of H₂S (resolution 0.08 cm⁻¹). OSA: Optical Spectrum Analyser.

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Measurement Parameters, Results and Discussions

Sample preparation:

- Mixing volume of ~10 dm³ made of Pyrex glass with detachable transfer volume (~82 cm³, Pyrex).
- Mixtures of H₂S in air were prepared at a ratio of 400 mbar H₂S / 600 mbar air.
- Mixture in transfer volume released into cavity (volume 998 cm³).

Measurement conditions:

- Room temperature (296 K), static gas mixture H₂S in air at pressure ~82 mbar.
- Integration time 120 s, resolution 0.08 cm⁻¹, cavity length 66 cm, reflectivity 0.993 (Perkin-Elmer L1050)

Procedure:

- Based on HITRAN data [6] software was written that identifies the required FSR and optimum GSFC center position (arbitrarily chosen as the position of line 8 – see Fig. 3) to match comb lines with absorption features in the relevant spectral region.
- GSFC lines were matched with ro-vibrational lines of H₂S by coarse tuning over one FSR (6349.15 - 6349.45 cm⁻¹). Then fine tuning in steps of 0.004 cm⁻¹ to optimize comb line position at center (line 8) of 6349.407 cm⁻¹.
- Line 3 and 9 were matched, line 18 partially matched. Situation illustrated in Fig. 3

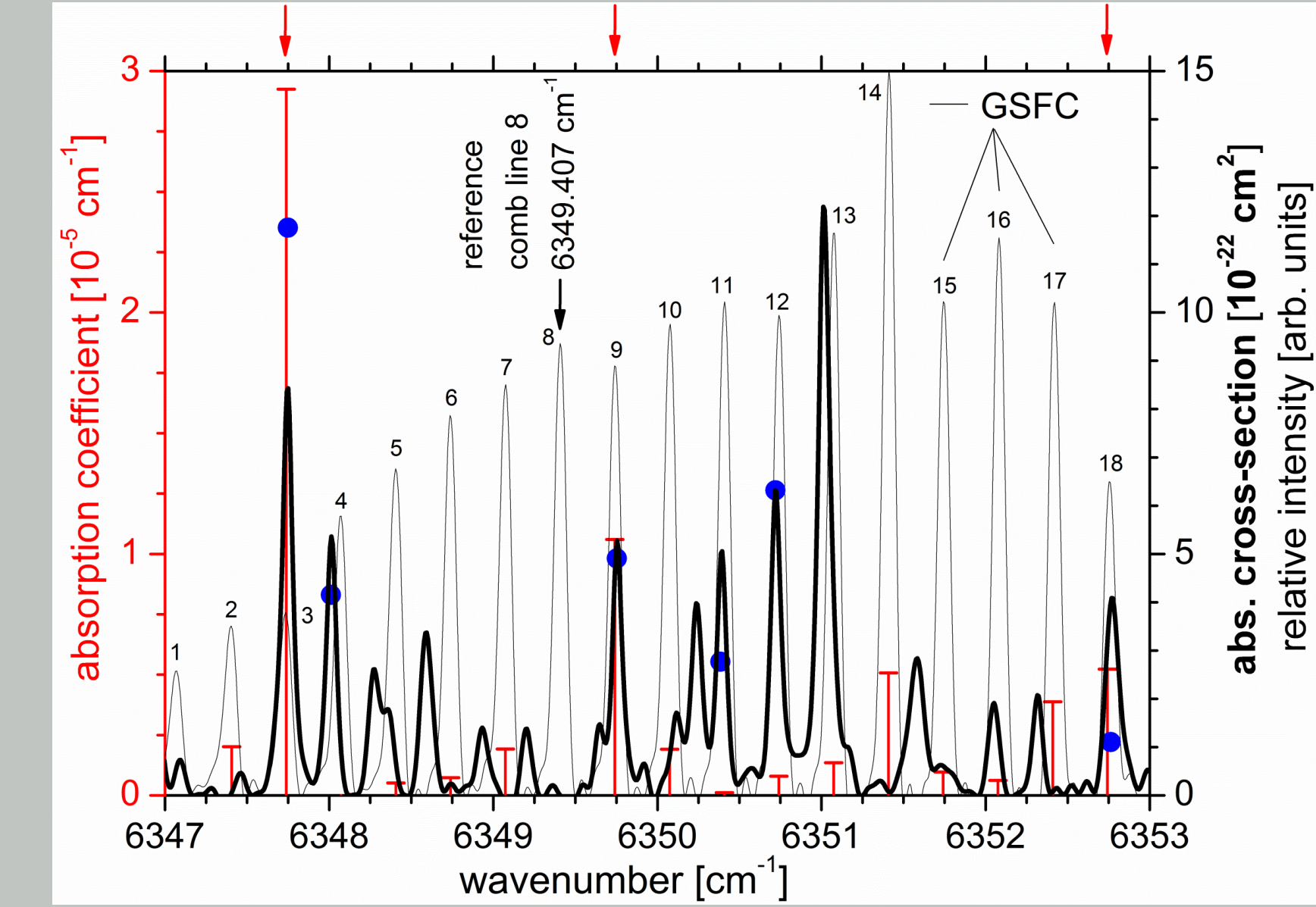


Fig. 3: The thick black trace is a small spectral segment of the H₂S reference spectrum in Figure 1 which has been scaled at 6350.71 cm⁻¹ to the HITRAN absorption cross-sections (=blue solid dots) for a spectral resolution of 0.08 cm⁻¹ (bold axis title, right axis). The thin black trace shows 18 comb lines of the GSFC laser (relative intensity; second title on the right axis). The red trace (left axis) shows the absorption coefficient measured on the 18 comb lines for a center position of the GSFC at 6349.407 cm⁻¹ and a comb line spacing of 10 GHz. The red vertical arrows indicate comb lines which overlap significantly with H₂S absorption features at 6347.74, 6349.74, and 6352.74 cm⁻¹.

From the absorption data in Fig. 3 the average mixing ratio corresponds to 0.92 ppTv. This mixing ratio was ~35 times lower than the mixing ratio of 32.4 ppTv expected based on the sample preparation procedure. This difference is attributed to the significant wall losses experienced with H₂S when in contact with the Pyrex glass mixing chamber and transfer volume. The fractional throughput of H₂S through a Pyrex glass enclosure is known to be approximately 3% [7].

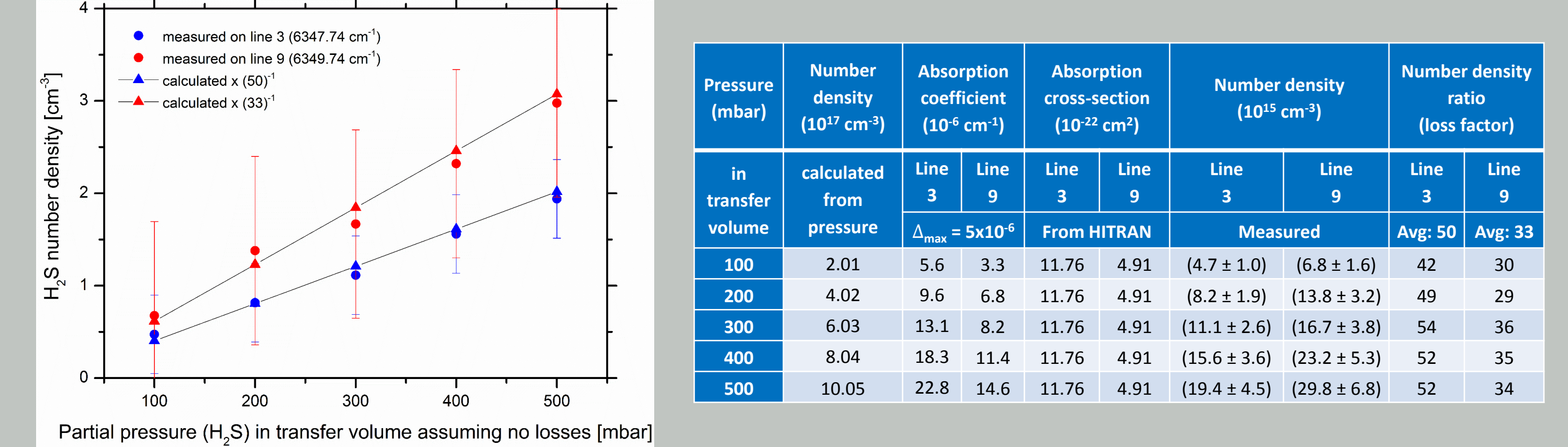


Fig. 4: The blue and red circles indicate the measured H₂S number densities for line 3 (6347.74 cm⁻¹) and line 9 (6349.74 cm⁻¹), respectively, as a function of the partial pressure in the transfer volume, assuming no losses of H₂S occurred during the transfer into the cavity. Triangles represent the number densities for the same comb lines calculated from the HITRAN absorption cross-sections scaled to account for H₂S wall losses. The deviation of the scaling factor of line 3 from the predicted value of 33 is likely a consequence of the comb line not aligning perfectly with the maximum of the H₂S absorption line. The corresponding data can be found in the adjacent table.

Zeroing the Measurement

Since line 8 of the comb (at 6349.407 cm⁻¹) does not overlap with any H₂S absorption features (see fig. 3) its peak intensity can be used as the “reference intensity”, $I_{0,8}^{ref}$, for all other comb lines, giving an intensity ratio r_k for each comb line k ; $r_k = I_{0,k}^{ref} / I_{0,8}^{ref}$. Using this ratio, and if the effective reflectivity of the cavity mirrors ($R(\nu_k)$) is known then the absorption coefficient (α_k) on a matching comb line k may be determined:

$$\alpha_k(\nu_k) = n\sigma_k(\nu_k) = \frac{1 - R(\nu_k)}{d} \left(\frac{r_k I_{0,8}}{I_k(\nu_k)} - 1 \right) \quad (1)$$

Where $I_k(\nu_k)$ is the measured intensity transmitted through the cavity, d is the cavity length over which light can interact with the sample, n is the number density of absorbing species present in the cavity, and $\sigma_k(\nu_k)$ is the absorption cross-section of the corresponding absorbers.

Summary

- Feasibility of a gain switched frequency comb laser as a light source for cavity enhanced absorption detection in the near IR has been successfully demonstrated using H₂S as a target species.
- The light from the GSFC laser was directly coupled to a medium finesse cavity without any complicated mode matching scheme.
- Even though the sensitivity is modest (~200 ppmv), it is sufficient to reach the lower explosion limit of H₂S in the near IR.
- Advantages of the GSFC are (i) the fibre coupling features, (ii) well-defined I_0 , (iii) experimental simplicity, (iv) cost effectiveness and potential future compactness.

References

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