

On-Line Measurement of Gaseous Iodine Species During a PWR Severe Accident

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Introduction:

• Iodine is a major contributor to the potential radiological consequences in case of a nuclear accident.

1. Due to its affinity to the thyroid.
 2. Represents a major part of the released radioactive gases.
 3. Iodine gaseous forms are less effectively trapped by the filters during an accident.
- Iodine gaseous forms consist of I₂ and CH₃I.

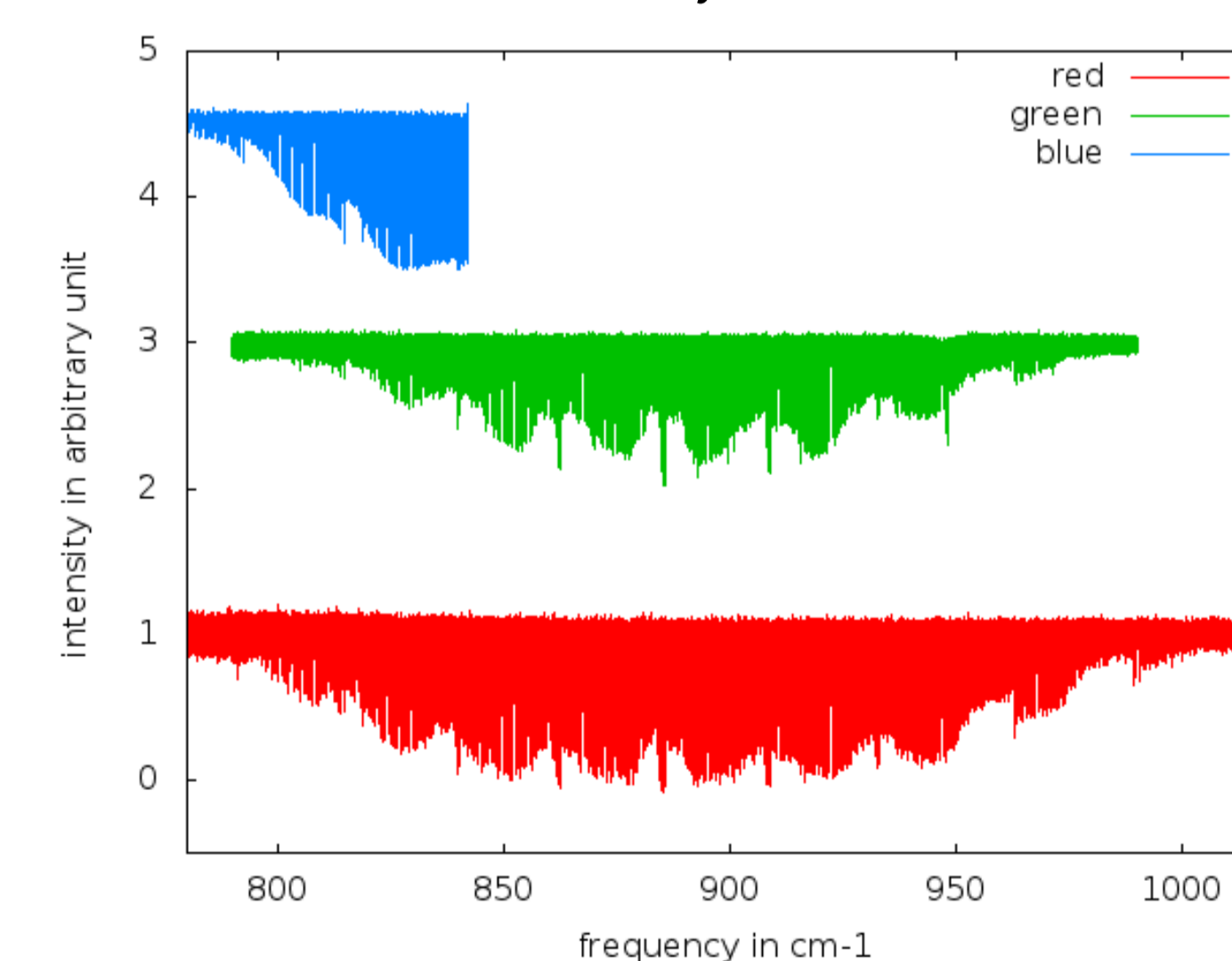
1. I₂ produced in the primary circuit.
2. CH₃I released in the reactor building and produced from the interaction, under ionizing radiation, of iodine with the paintings.

Iodine is also present under aerosol forms. Four years after Fukushima the aerosol/gaseous partition is still not clear. The uncertainty regarding chemical forms and speciation of iodine has a direct impact on prediction of iodine evolution in the containment and its subsequent release into the environment [1].

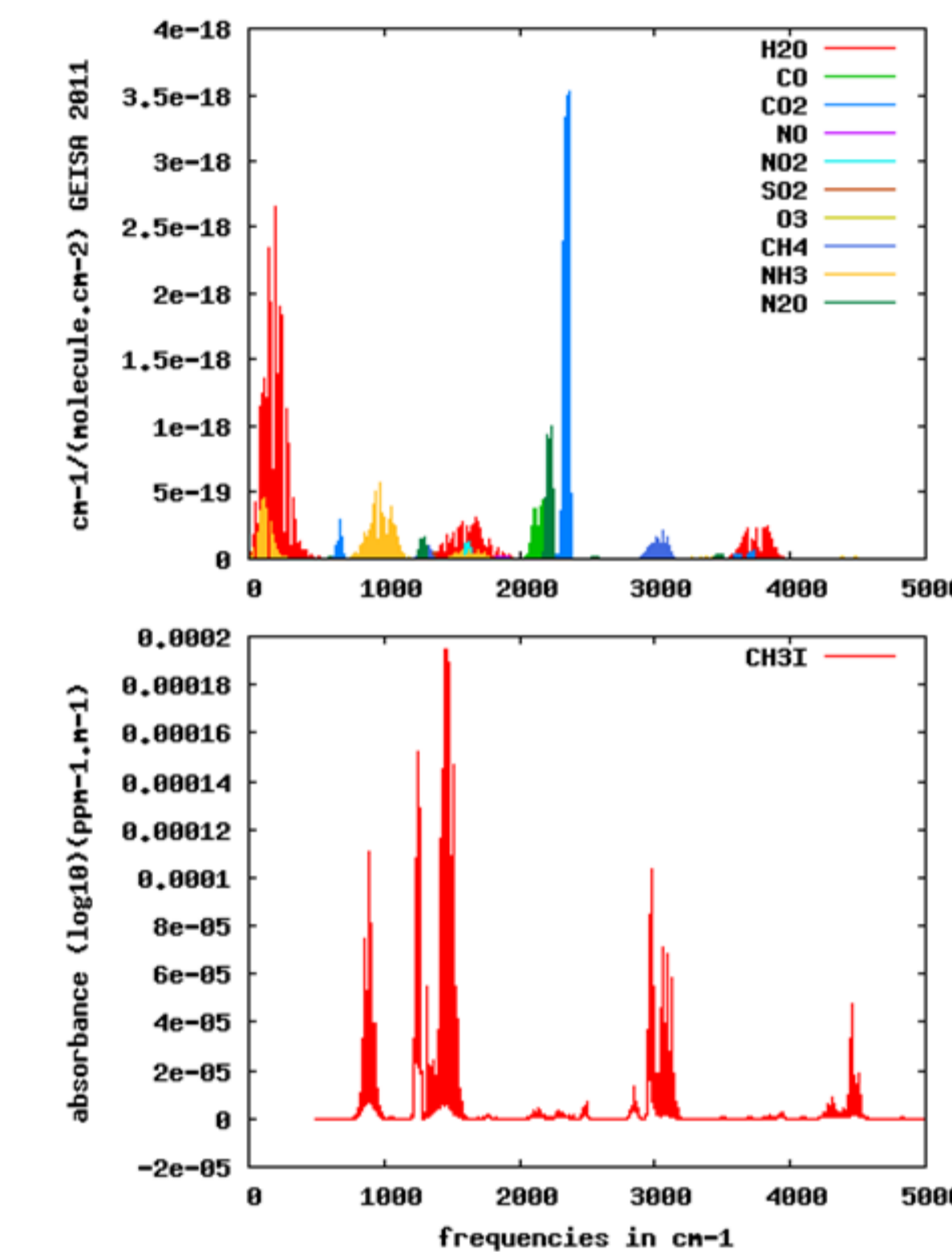
Methyl Iodide CH₃I:

Spectra's recording:

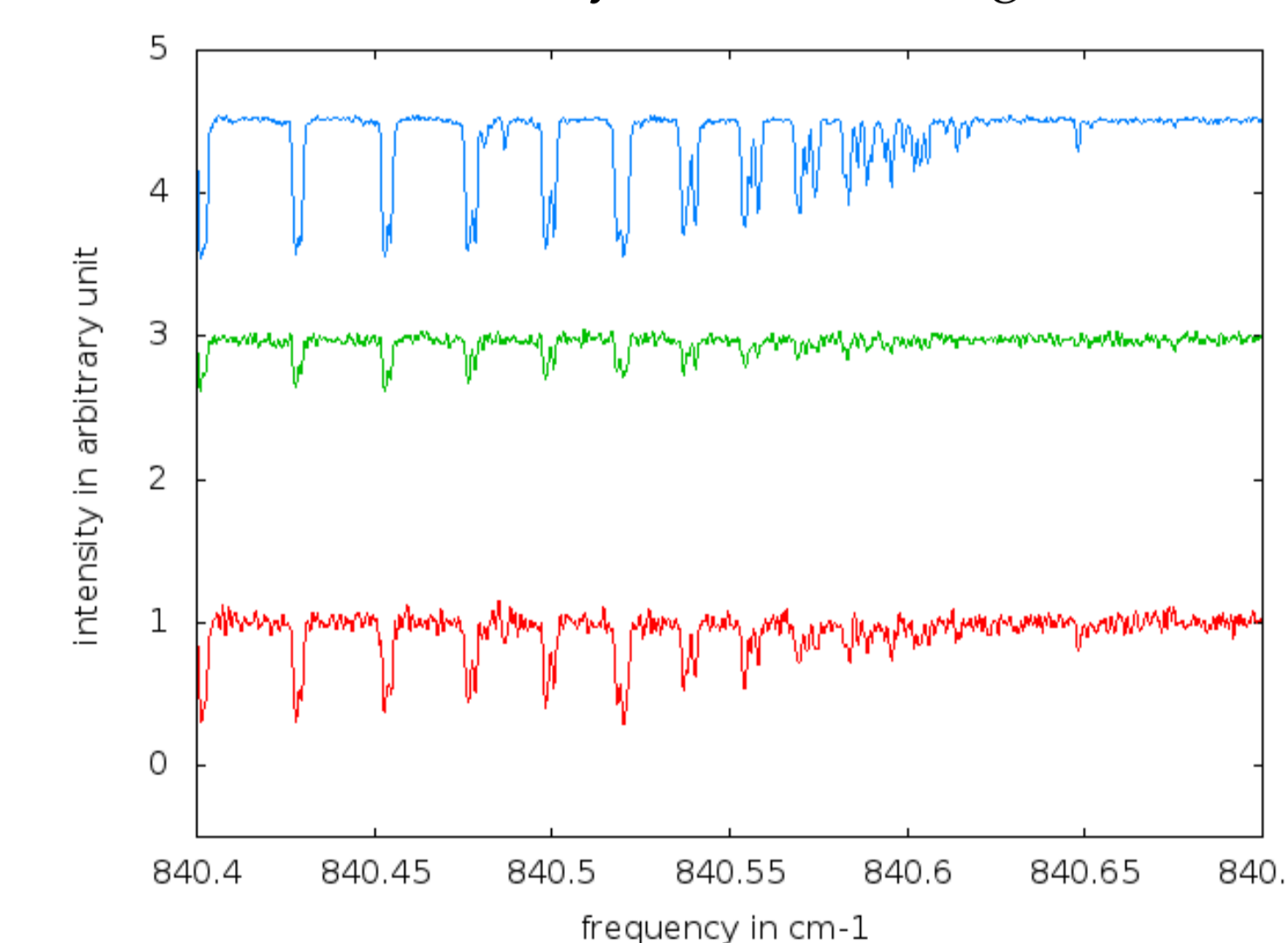
Measurements were conducted at the AILES beamline of the synchrotron SOLEIL.



The choice of the $\nu_6 = 1$ of methyl iodide band was based on the fact that it is relatively isolated from the absorption of the atmospheric species. The bottom spectrum is issued from the PNNL (Pacific Northwest National Laboratory).



The improvement of the spectral quality becomes evident by this zooming effect.



. Colour	Pressure	Temperature	Optical Length	Cell	Detector	Scans
Blue	0.255 mb	223 K	15 m	Cryogenic [2]	HgCdTe	760
Green	0.16 mb	223 K	3 m	Cryogenic [2]	MCT	872
red	0.34 mb	293 K	4 m	4 m multi pass	MCT	770

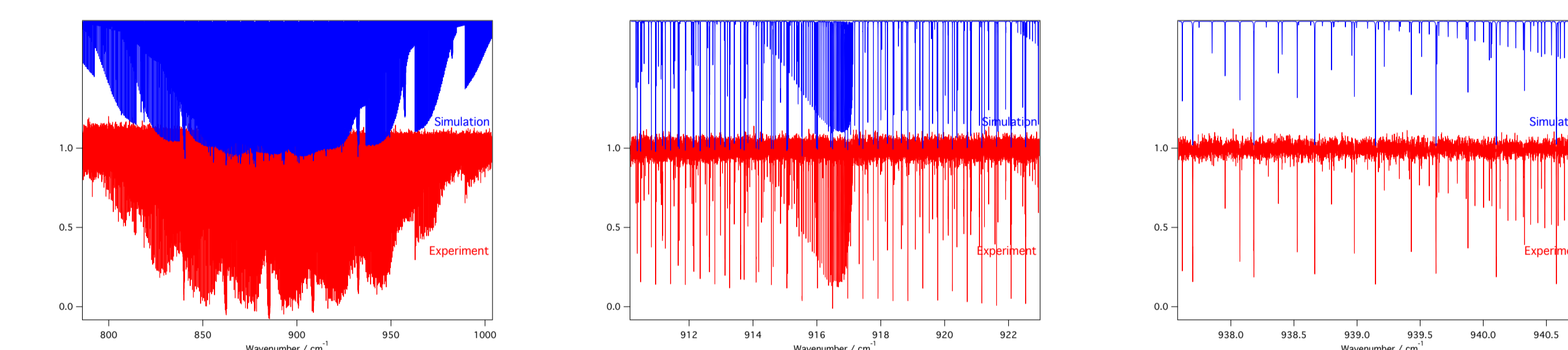
References:

- [1] Olof Johansson, Herve Mutelle, Alexander E. Parker, Sébastien Batut, Pascal Demaux, Coralie Schoemaeker, and Christa Fittschen. *Appl. Phys. B*, 114:421–432, 2014.
- [2] Fridolin Kwabia Tchana, Fabrice Willaert, Xavier Landsheere, Jean-Marie Flaud, Leatitia Lago, Mylne Chapuis, Christian Herbeaux, Pascale Roy, and Laurent Manceron. *Rev. Sci. Instrum.*, 84(9):093101(1–7), 2013.
- [3] S. Carocci, A. Di Lieto, A. De Fanis, P. Minguzzi, S. Alanko, and J. Pietilä. *J. Mol. Spec.*, 191:368–373, 1998.
- [4] A. El Hilali, Ch. Wenger, V. Boudon, and M. Loëte. *J. Quant. Spectrosc. Radiat. Transfer*, 111:1305–1315, 2010.
- [5] R. Paso and S. Alanko. *J. Mol. Spec.*, 157:122–131, 1993.

C_{3v}TDS Suite of Programs – Application to CH₃I:

- Initial assignments were based on parameters available in the literature [3].
- We used the C_{3v}TDS package of the XTDS suite of programs developed in Dijon [4].
- Watson's parameters were converted to the tensorial ones.
- The analysis was straightforward and progressive.
- As a result more than 2200 lines were assigned up to J_{MAX}=61 and an RMS of 0.327x10⁻³ cm⁻¹.

The $\nu_6 = 1$ band of methyl iodide compared to the simulation results with the C_{3v}TDS package.

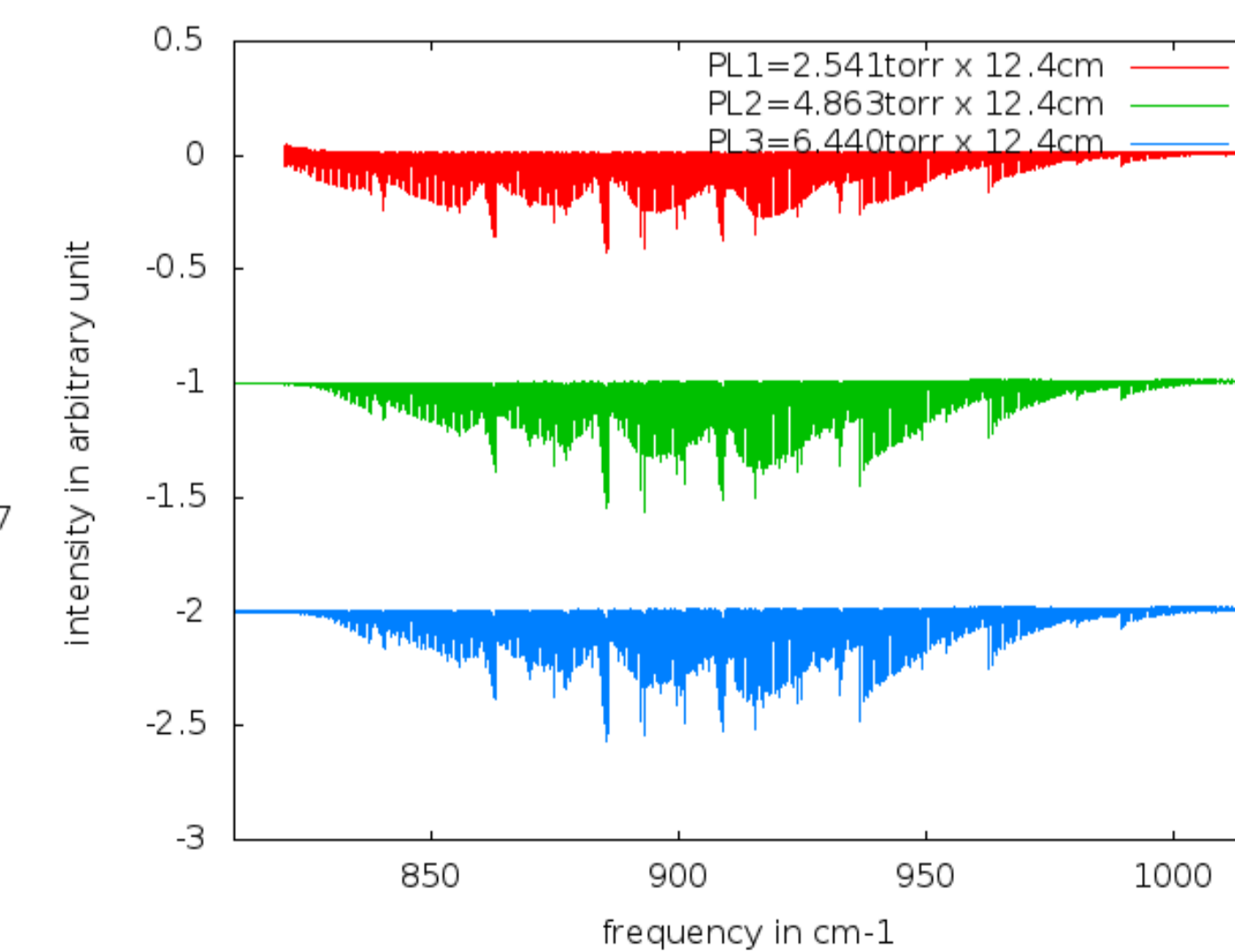


Effective Hamiltonian parameters ($t_{\{s\}}^{\Omega(K,n\Gamma)\Gamma_v\Gamma'_v}$) for the $\nu_6 = 1$ band of CH₃I:

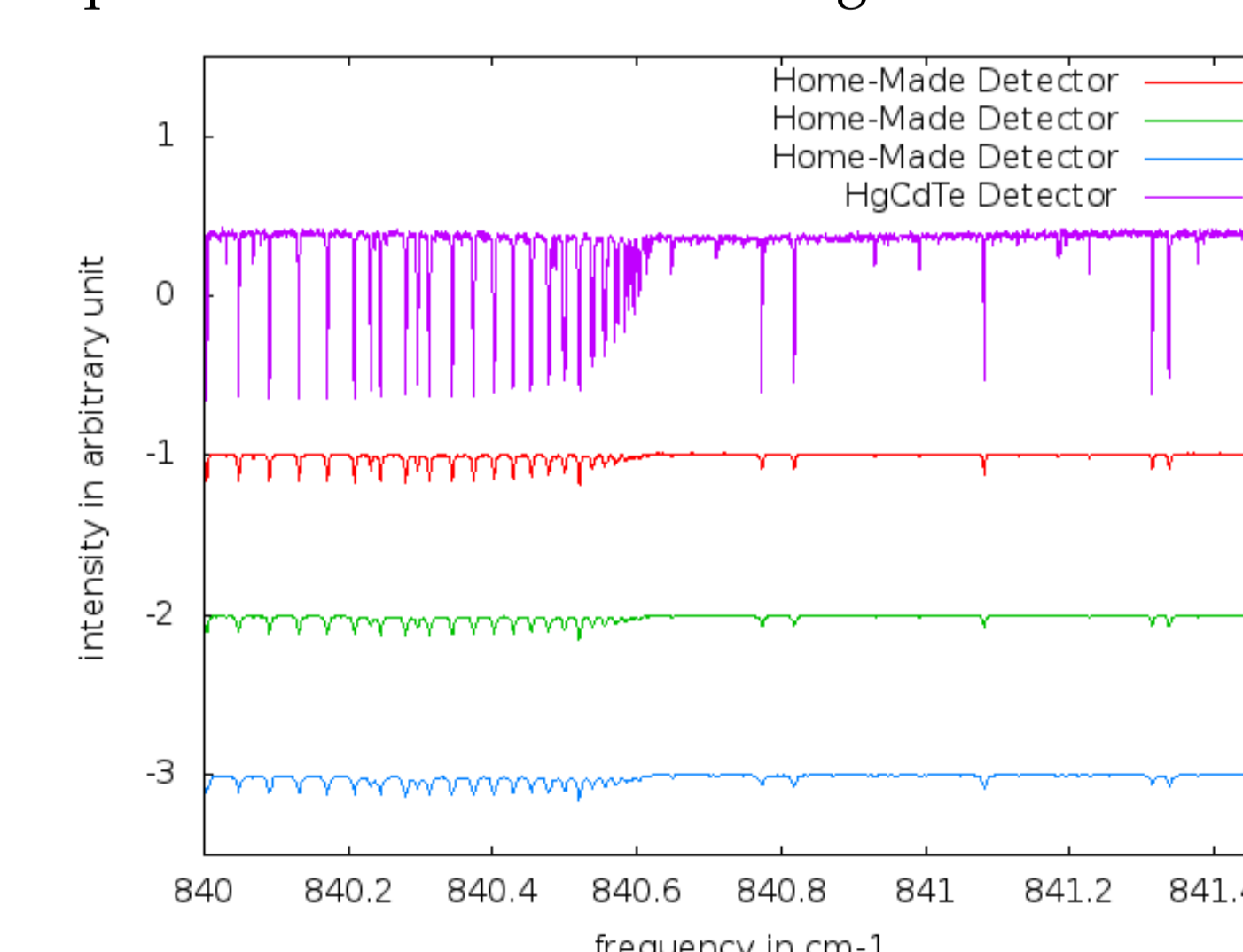
Ground State			$\nu_6 = 1$		
Order	Parameter	Value /cm ⁻¹	Order	Parameter	Value /cm ⁻¹
0	2(0, S+)	1.858(13)	0	0(0, S+)	882.819(49)
	2(2, S+)	9.846x10 ⁻¹	1	1(1, S-)	-1.408(76)
2	4(0, S+)	-1.886(37)x10 ⁻⁵	2	2(0, S+)	1.104(52)x10 ⁻²
	4(2, S+)	6.938(96)x10 ⁻⁶		2(2, S+)	1.024(44)x10 ⁻²
	4(2, S+)	-2.619x10 ⁻⁶		2(2, D)	9.876(16)x10 ⁻⁵
4	6(0, S+)	6.407(47)x10 ⁻¹¹	3	3(1, S-)	-2.752(24)x10 ⁻⁵
	6(2, S+)	1.027(75)x10 ⁻¹¹		3(3, S-)	1.550(14)x10 ⁻⁵
	6(4, S+)	-3.784(29)x10 ⁻¹²	4	4(0, S+)	-6.920(21)x10 ⁻⁷
				4(2, S+)	3.697(11)x10 ⁻⁷
				4(2, D)	1.284(25)x10 ⁻¹⁰
				4(4, S+)	-1.457(42)x10 ⁻⁷

Intensity modelling:

For the intensity modelling we recorded the $\nu_6 = 1$ band at different PxL products. For this aim we used the FTS of the LISA.



Even at high pressures this figure shows a huge signal to noise ratio highly adequate for the future modelling of intensities!

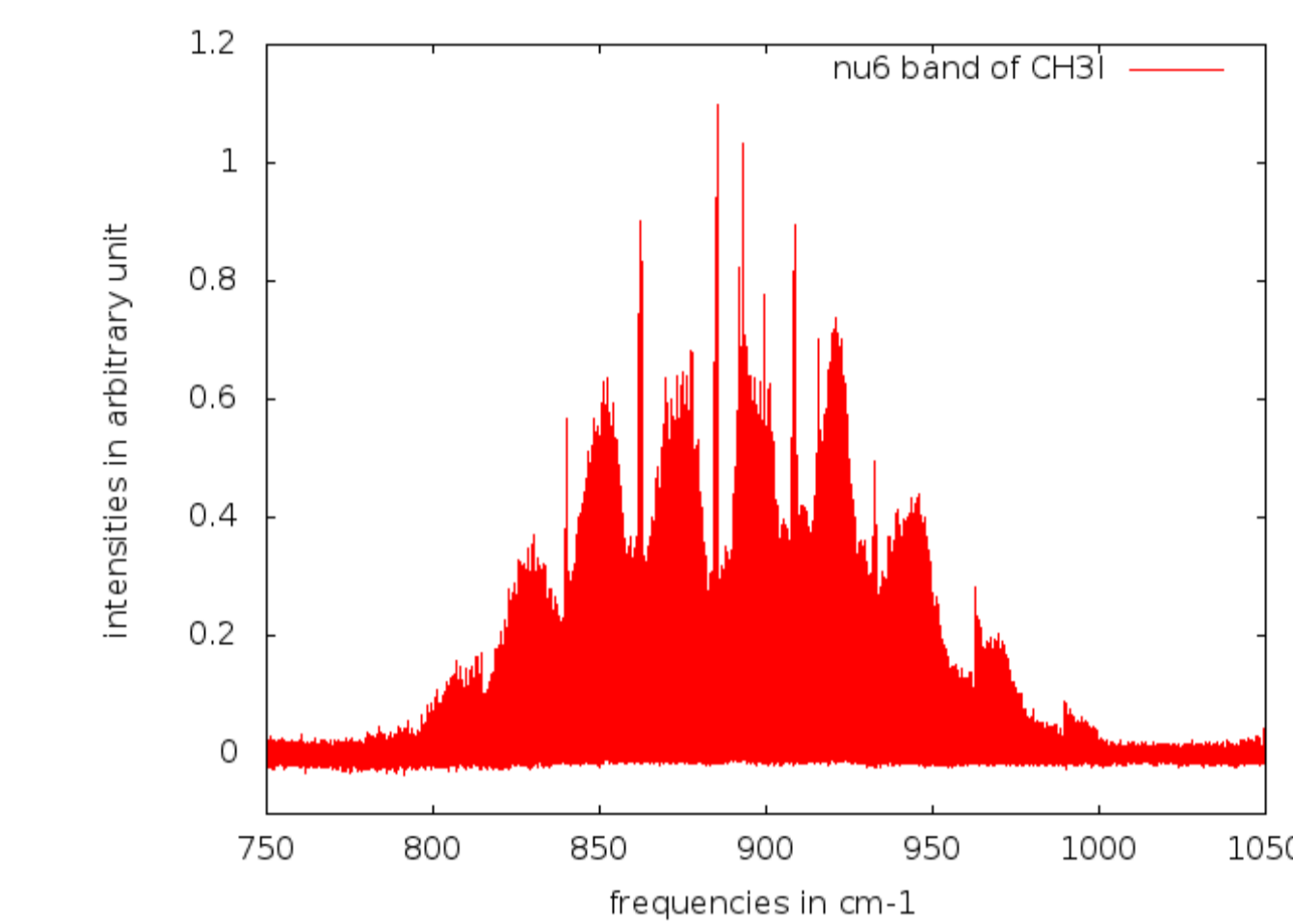


Acknowledgment:

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Line list in progress...

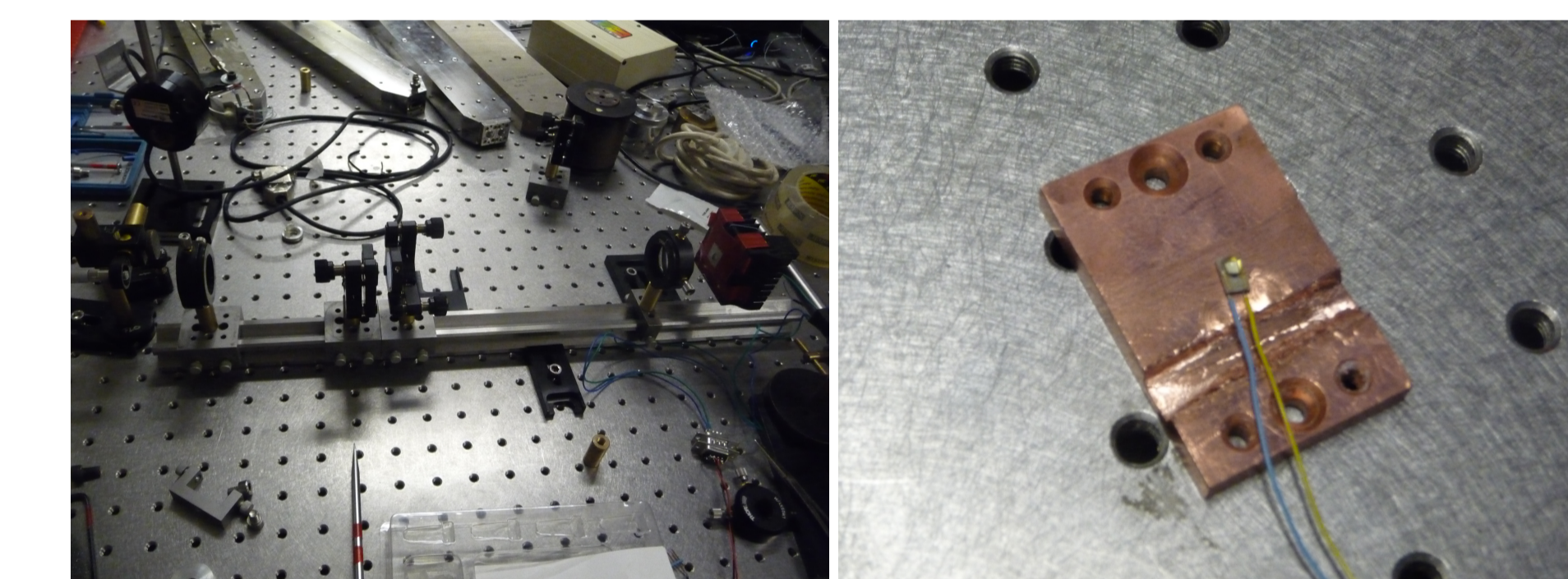
This spectrum was recorded at the AILES beamline of the synchrotron SOLEIL at room temperature. It was recorded by the combination of the synchrotron light and a home made MCT detector.



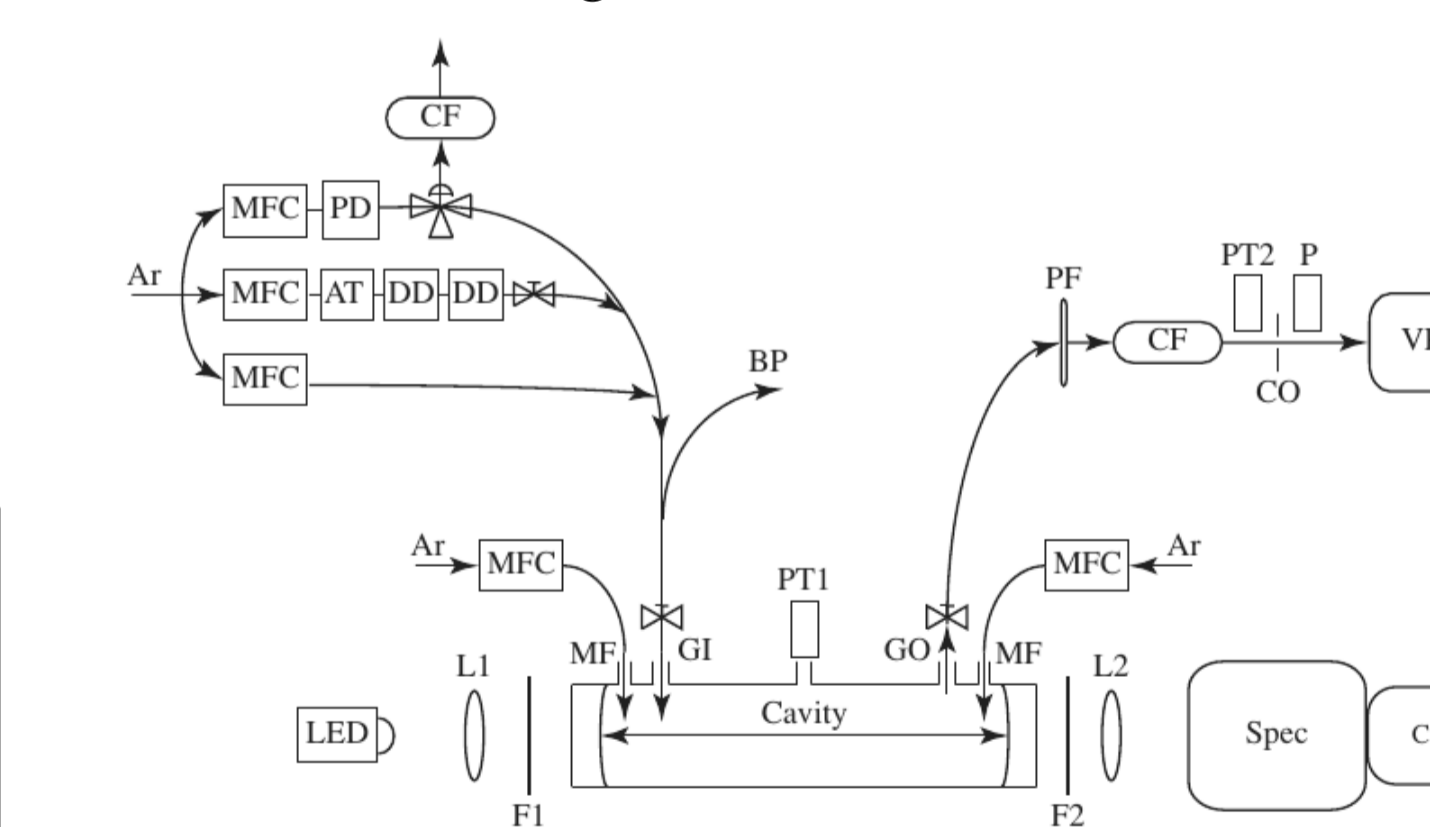
Parameter	$\nu_6 = 1$	
	Present work	[3]
E ₆ (THz)	26.46924014(24)	26.46922193(20)
A'(GHz)	156.151484(63)	156.1509369(27)
B'(Ghz)	7.47766294(31)	7.47766312(22)
Δ _K (MHz)	2.7359(31)	2.729748(55)
Δ _{JK} (kHz)	99.033(19)	98.8256(45)
Δ _J (kHz)	6.3491(16)	6.34991(18)
η(GHz)	32.851554(89)	32.852482(22)
η _K (MHz)	4.165(20)	4.16192(76)
η _J (kHz)	199.85(20)	200.933(33)
q ₀ (MHz)	-1.4798(10)	-5.92200(50)
Lines	> 9000	4000[5]
Freq(cm ⁻¹)	760-1050	760-1050 [5]
J _{MAX}	70	70[5]
K _{MAX}	14	18[5]

Molecular iodine I₂:

Incoherent BroadBand Cavity Enhanced Absorption Spectroscopy:



Schematic Diagram of the IBBCEAS [1]:



- Simple, robust and compact.
- Requires only about 10 Watts of power.
- Newest LED (Philips Lumileds 560-570 nm) technology leading to very small divergence.
- High reflective LAYERTEC mirrors (0.9999) glass.
- Very high sensitivity better than 10⁻¹⁰ cm⁻¹ implying a good detection potential for the samples with small concentrations.

Perspectives:

- Include the Hyperfine structure in the C_{3v}TDS package.
- Provide line list for the full spectral range.
- Intensity modelling is in progress.