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Enrichment diagnostic by molecular spectroscopy

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MICA16 – High resolution spectroscopy for isotopic ratio measurements

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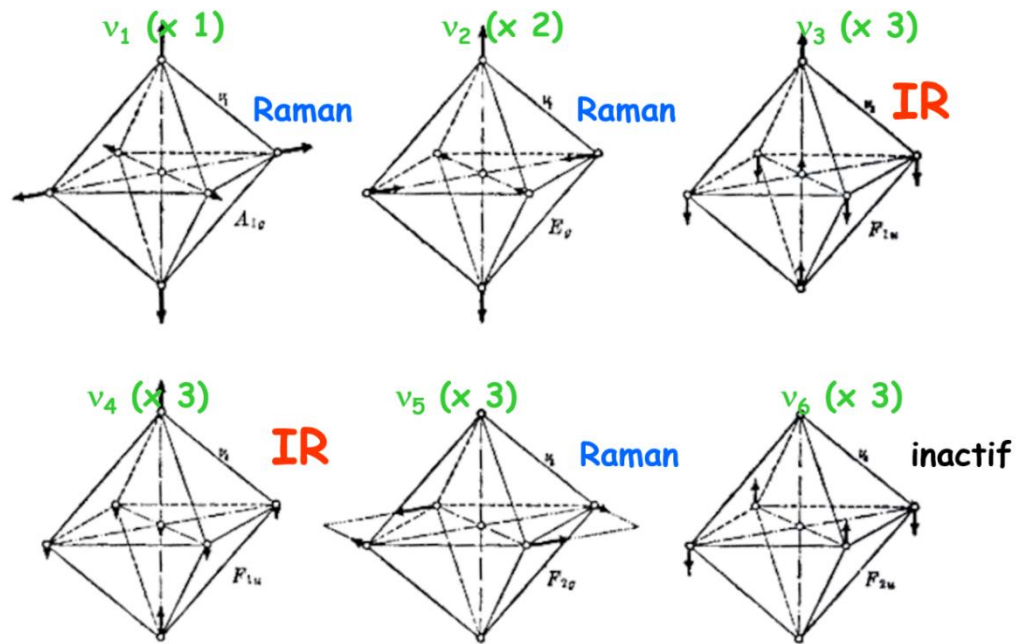


High resolution spectroscopy for isotopic ratio measurements

1. General information on optical UF₆ detection
 1. UF₆ spectroscopy / Quantum Cascade Lasers (IES – MirSense, R. Teissier)
 2. Molecular jet
2. Setup description
 1. Vacuum chamber layout and pulsed jet
 2. Optical beam folding
 3. Simulated measurements / 1f wavelength modulation
3. Measurements
 1. Temperature estimation
 2. Isotopic ratio measurement capability

Conclusion

1. Optical UF₆ detection



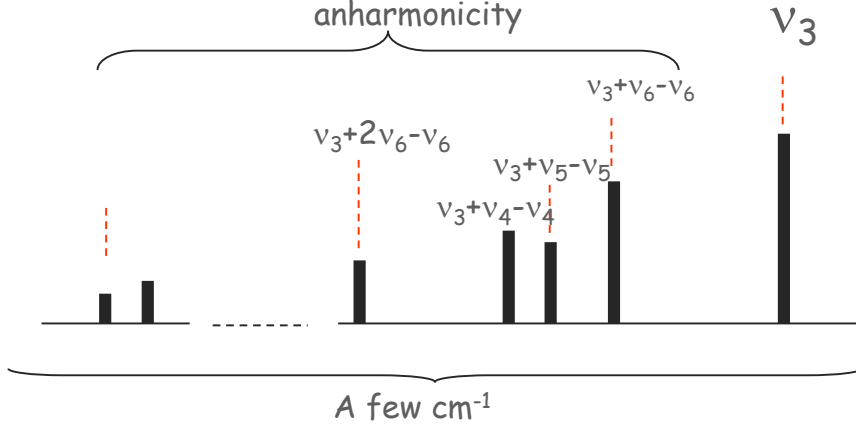
Band	Frequency (cm ⁻¹)	Isotopic shift (cm ⁻¹)
v_1	667	0
v_2	534	0
v_3	626	0.65
v_4	186	0.16
v_5	200	0
v_6	143	0

Isotopic shift maximum for v_3 band and maximal cross section coefficient

1.1 Vibrational spectra

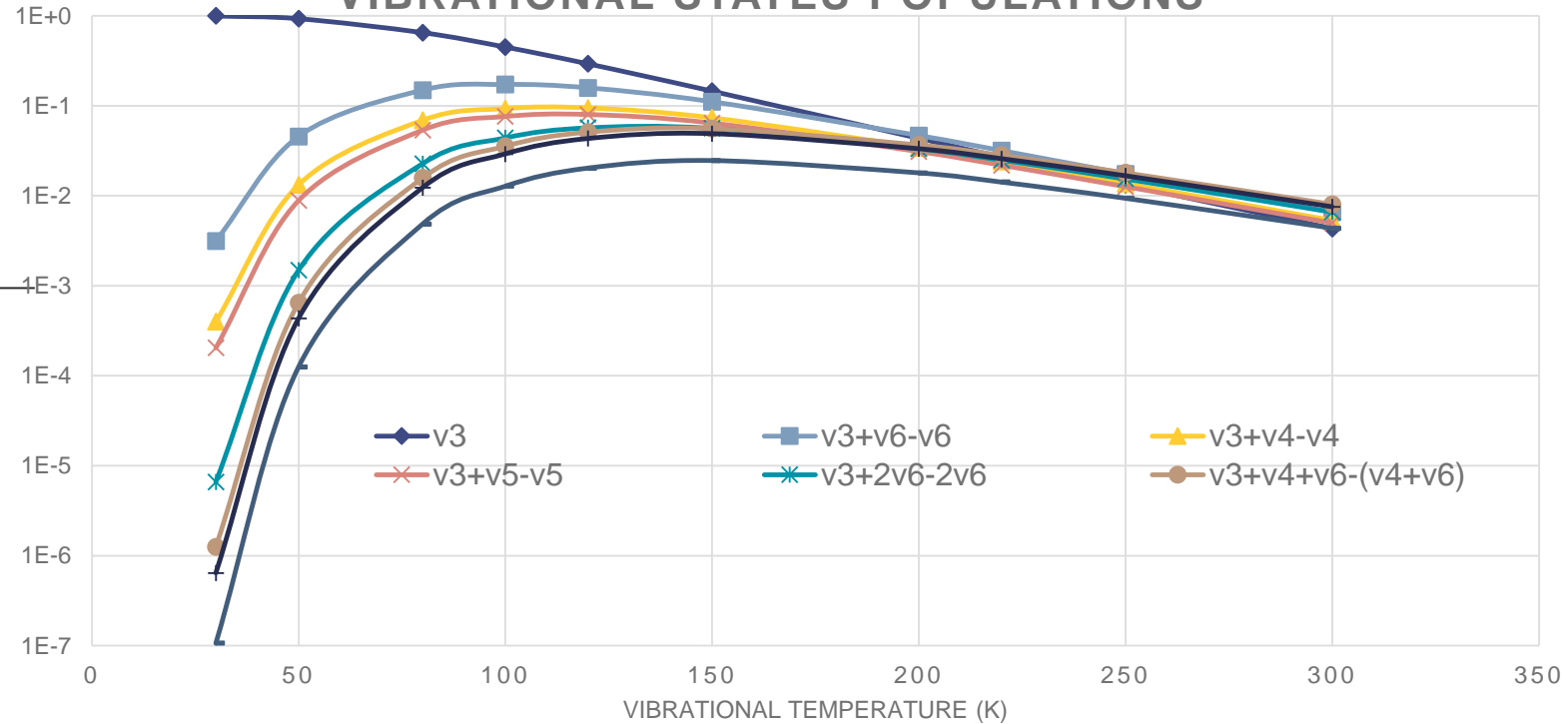
Purely vibrational spectra

Hot bands shifted by vibration anharmonicity



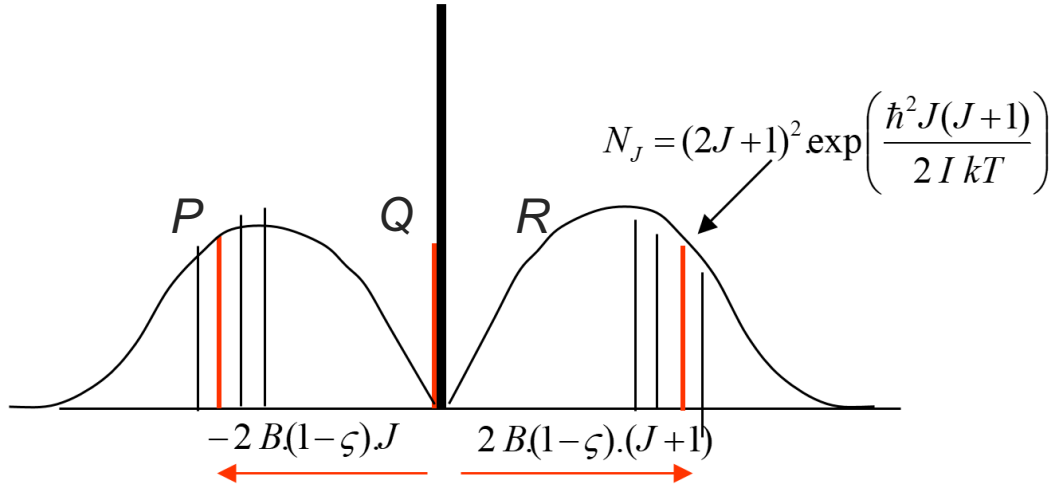
Band	Frequency (cm ⁻¹)	Isotopic shift (cm ⁻¹)
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ν_6	143	0

VIBRATIONAL STATES POPULATIONS



1.1 Rotational spectra

Purely rotational spectra



PQR branches: $P: \Delta J = -1$, $Q: \Delta J = 0$, $R: \Delta J = +1$
(J : rotational quantum number)

Energy levels spherical rotator ($I_x = I_y = I_z = I$)

$$E_J = \frac{\vec{J}^2}{2I} = \frac{\hbar^2 J(J+1)}{2I}; J=0,1,2,\dots$$

Rotational transitions : $J \rightarrow J + \Delta J$; $\Delta J = 0, \pm 1$

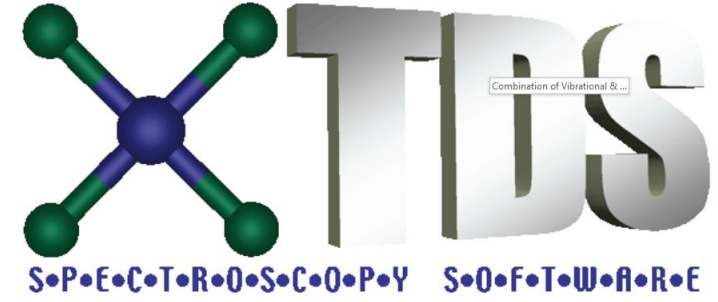
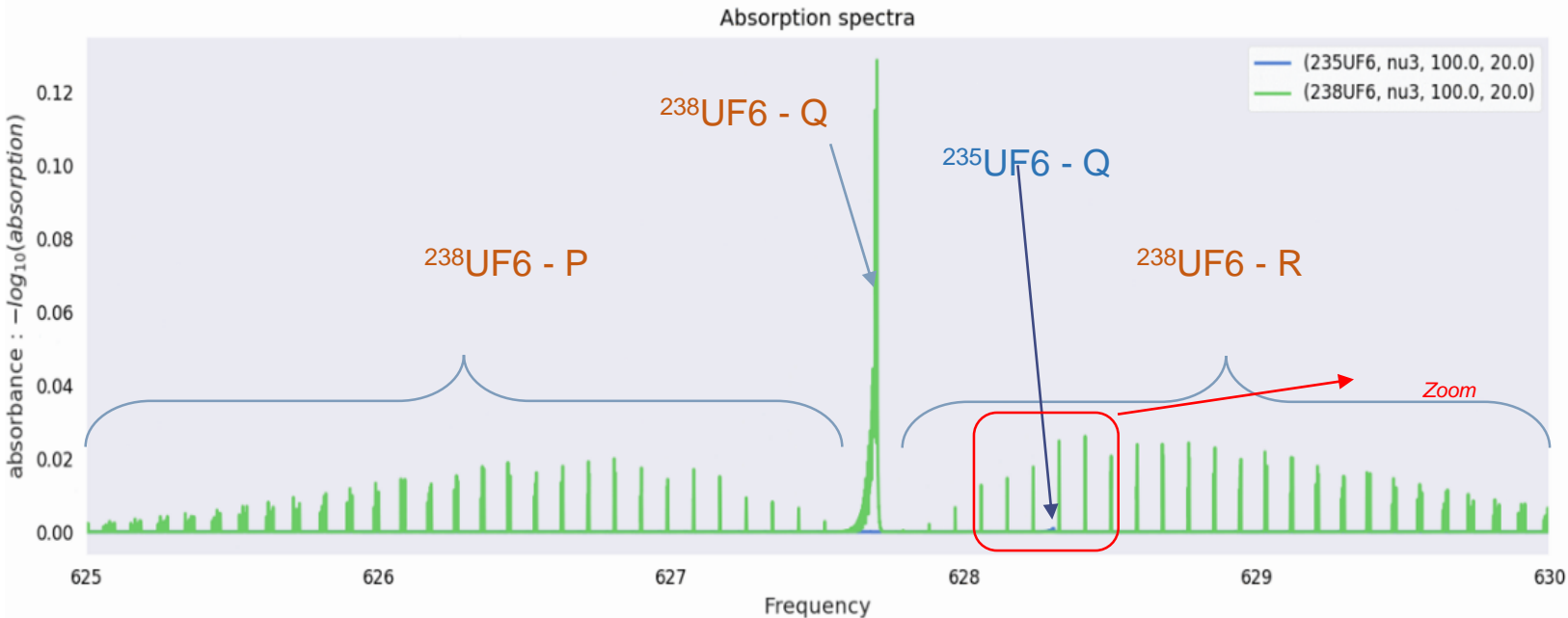
Wavenumber : $\sigma = \frac{E_{J+\Delta J} - E_J}{hc} = 2Bm$; $B = \frac{h}{8\pi^2 cI}$

$m = -J$; $\Delta J = -1$ (branche P)

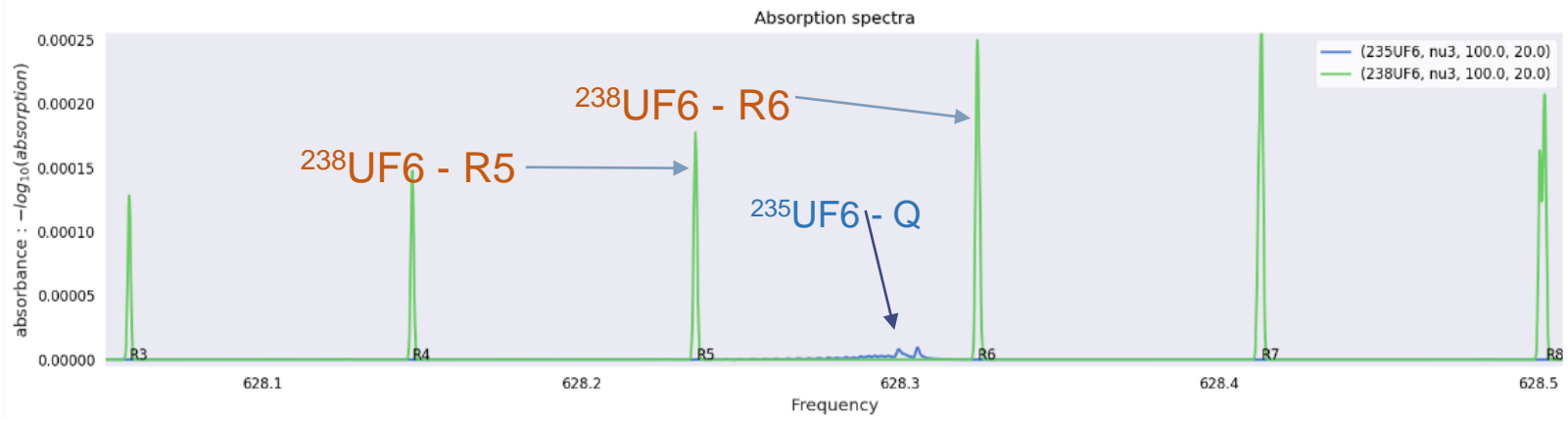
$m = 0$; $\Delta J = 0$ (branche Q)

$m = J+1$; $\Delta J = +1$ (branche R)

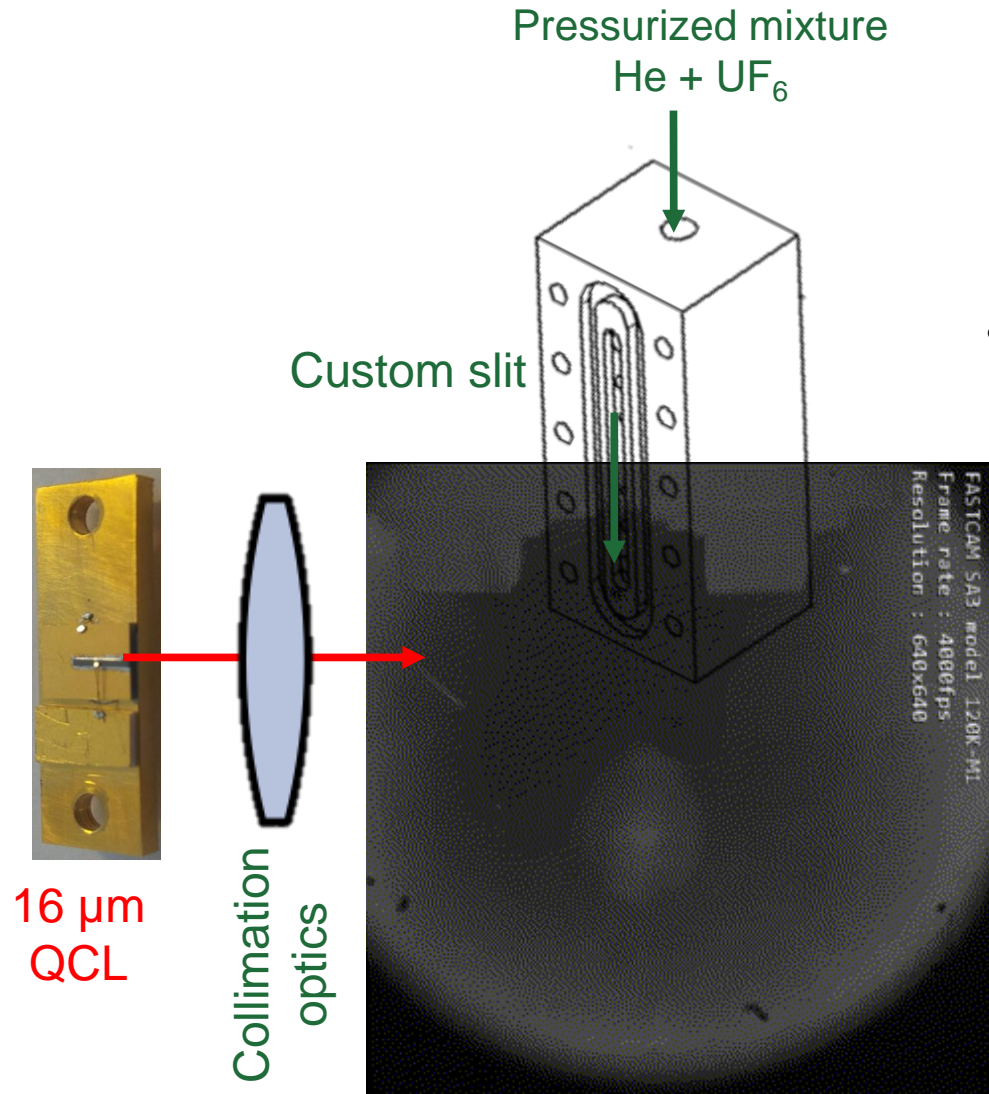
1.1 Combination of Vibrational & Rotational spectra



HTDS calculus : $^{238}\text{UF}_6 + ^{235}\text{UF}_6$



1.2 Molecular pulsed jet



*Pulsed jet symmetry
analysed in a Michelson
interferometer*

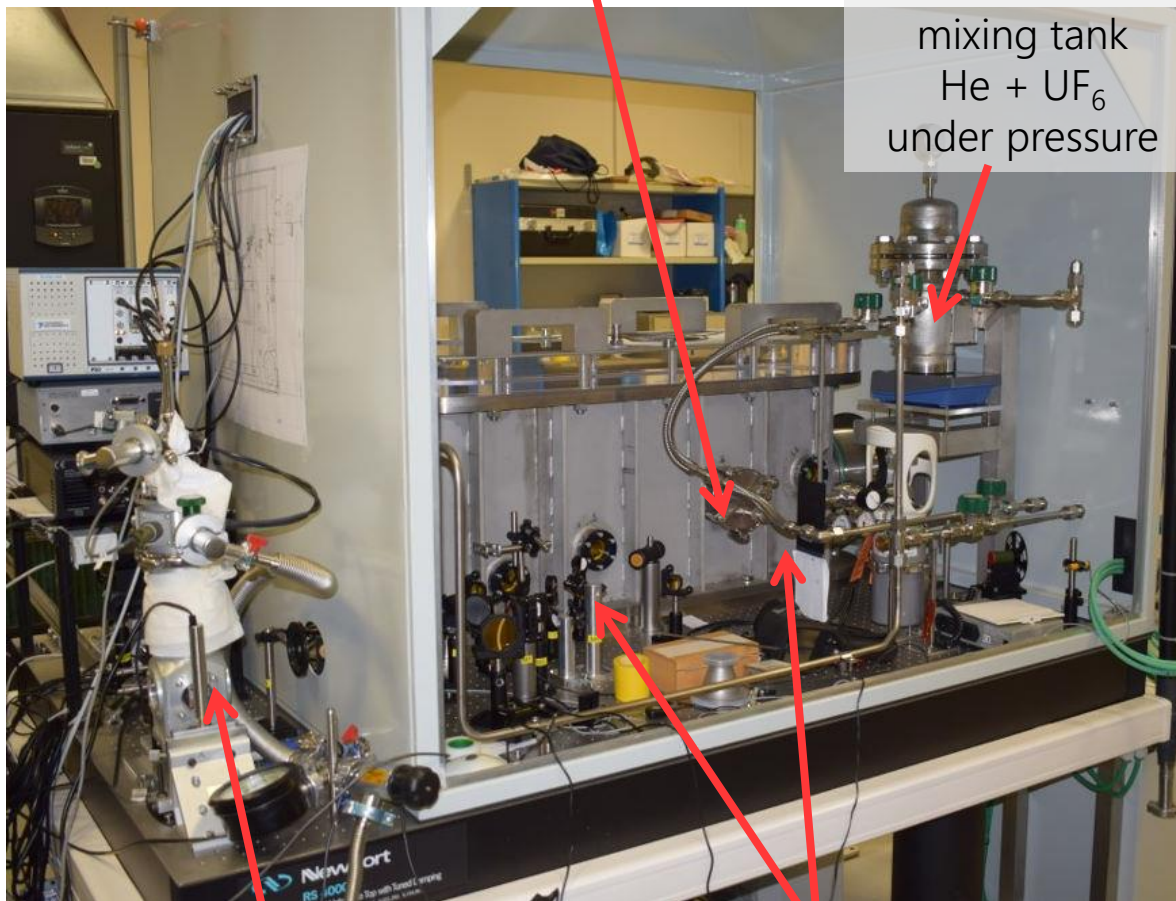
1. the mixing tank is maintained at a total pressure of a few bars
2. the vacuum chamber is maintained at a high vacuum level
3. pulsed valve on the way
4. the slit is positioned in front of the electro-mechanical needle valve



2. Key elements :

Pulsed jet
gas inlet

mixing tank
He + UF₆
under pressure



Cryostat (~100 K) for the QCL

2 detectors HgCdTe @ 16 μm

QCL laser @ 16 μm [627; 630] cm^{-1} ; bandwidth < 30 MHz
IES / Univ. Montpellier (R. Teissier)

2 IR detectors HgCdTe + cold filter: [15.490 μm , 16.429 μm]

Optical system with spherical mirrors

Pulsed valve + slit

Monaris Lab. / Sorbonne Univ. / Paris (P. Asselin)

Acquisition / Current modulation (~ > 100 kHz)
spectra simulation (HTDS code)

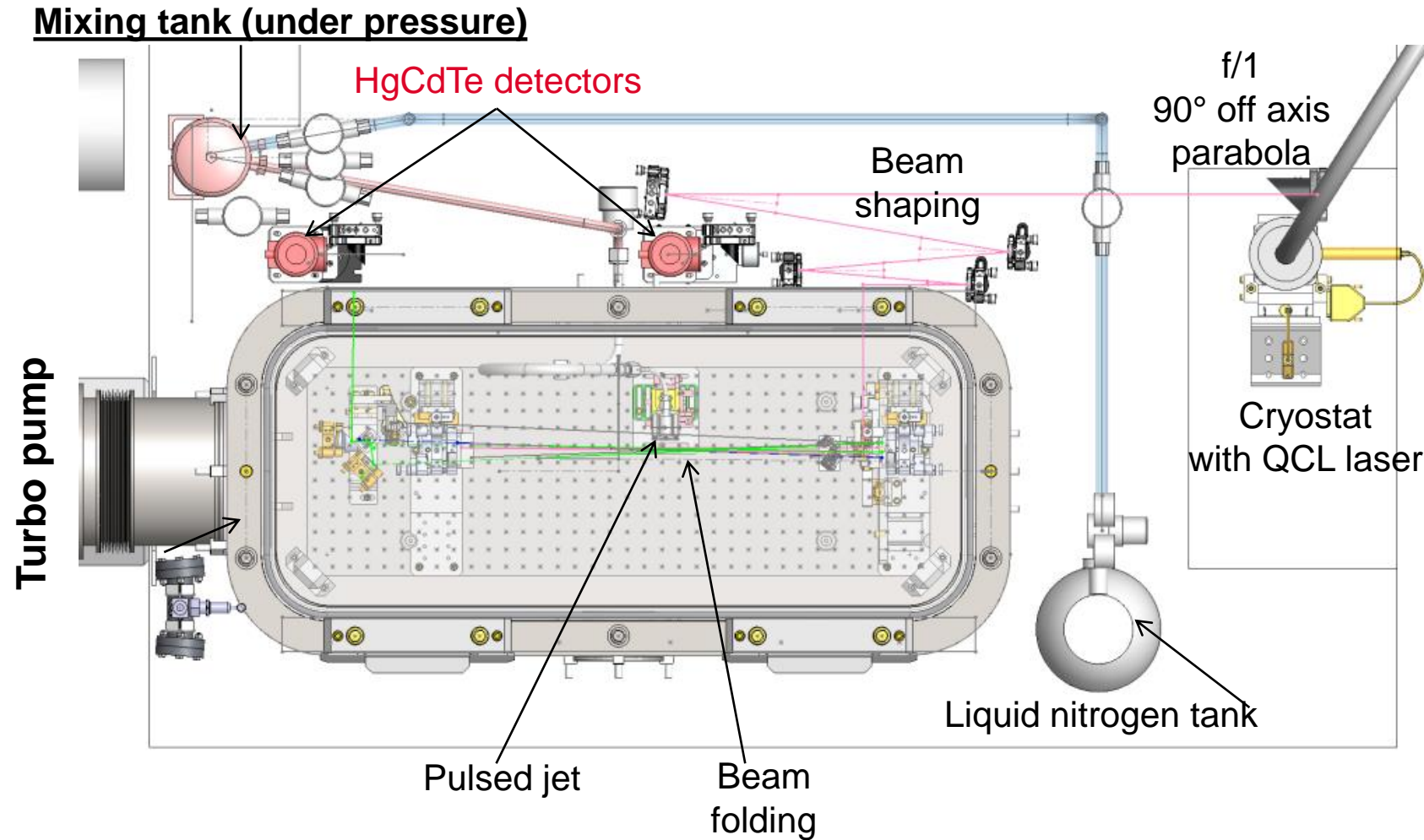
ICB: Institut Carnot de Bourgogne/Dijon (V. Boudon)

Software :

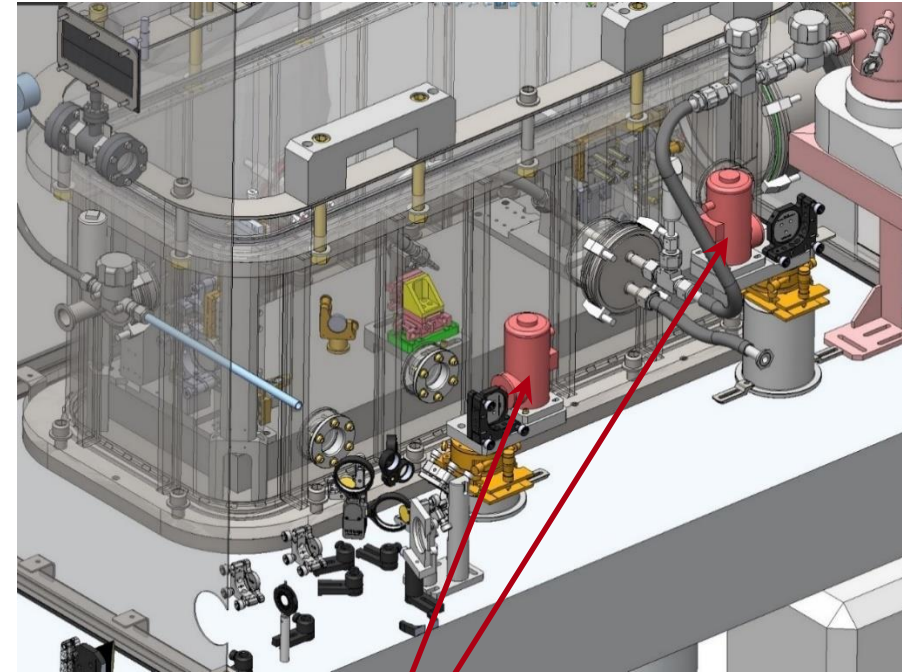
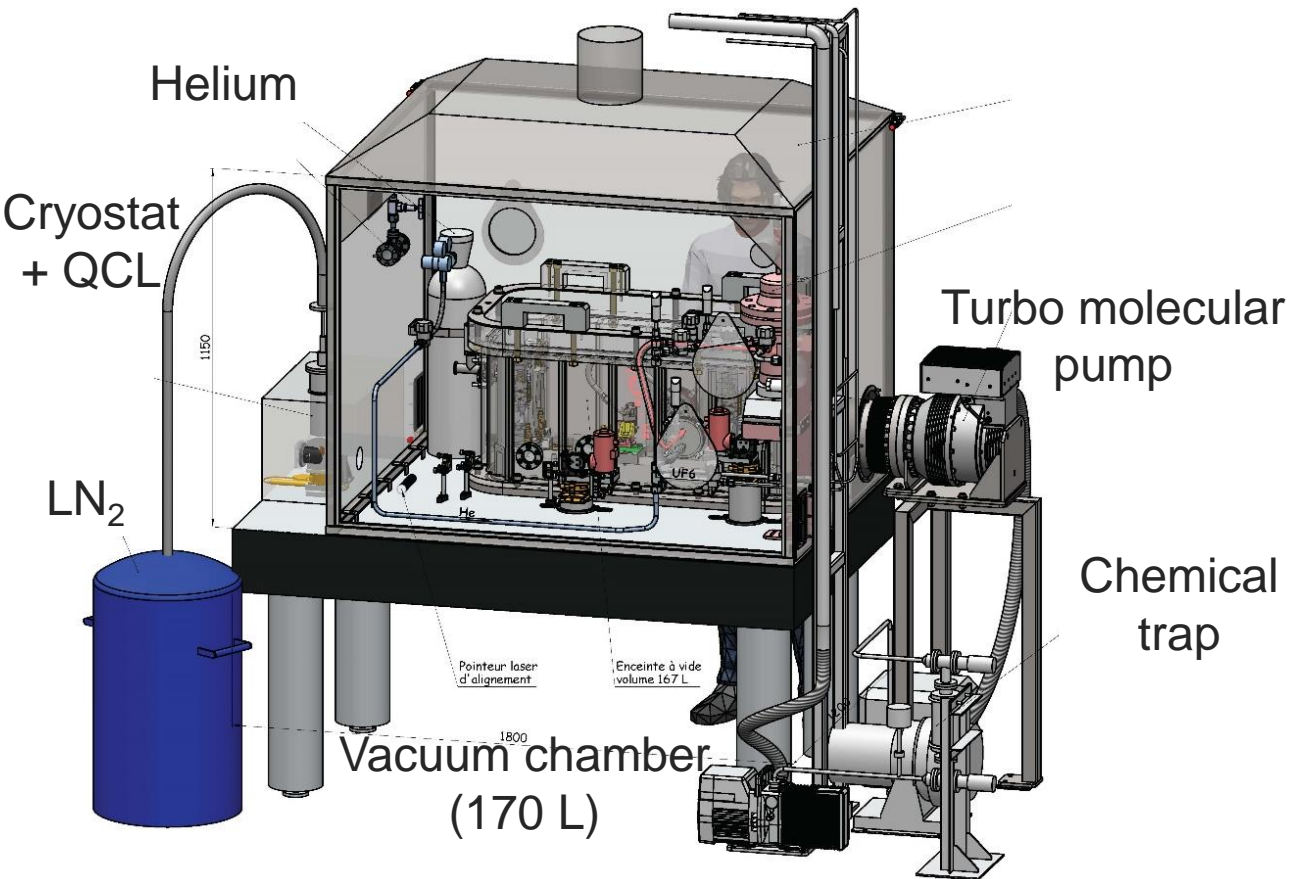
Labview for bench driving & 100 MHz acquisition

Python for demodulation & analysis
(CEA)

2.1 Optical bench layout



2.1 Optical bench



**HgCdTe Detectors
+ narrow cold filter :**
[15.490 μm , 16.429 μm]

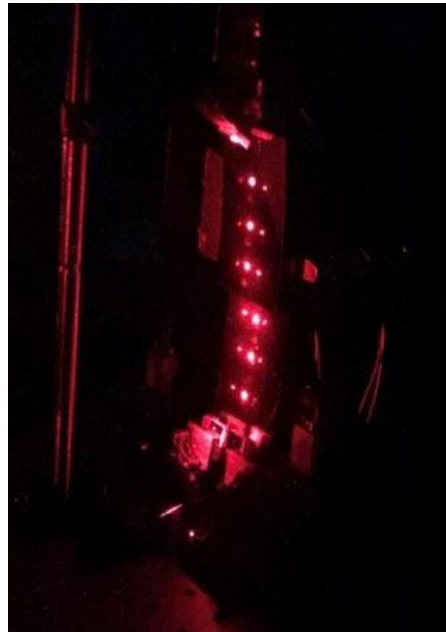
2.2 Classical multi-pass cavity



Pre-adjusting the optical cavity with a visible red beam

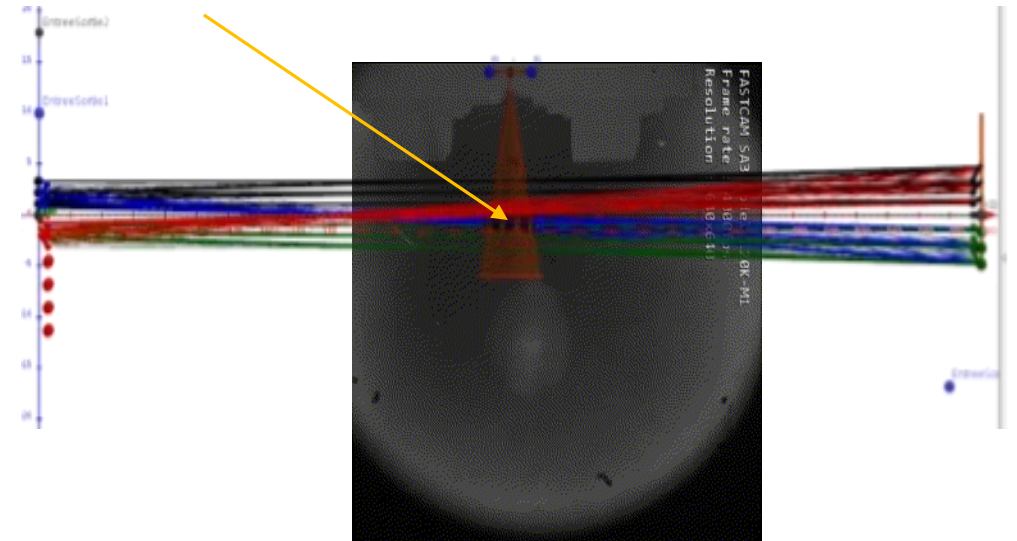


3 sheets :

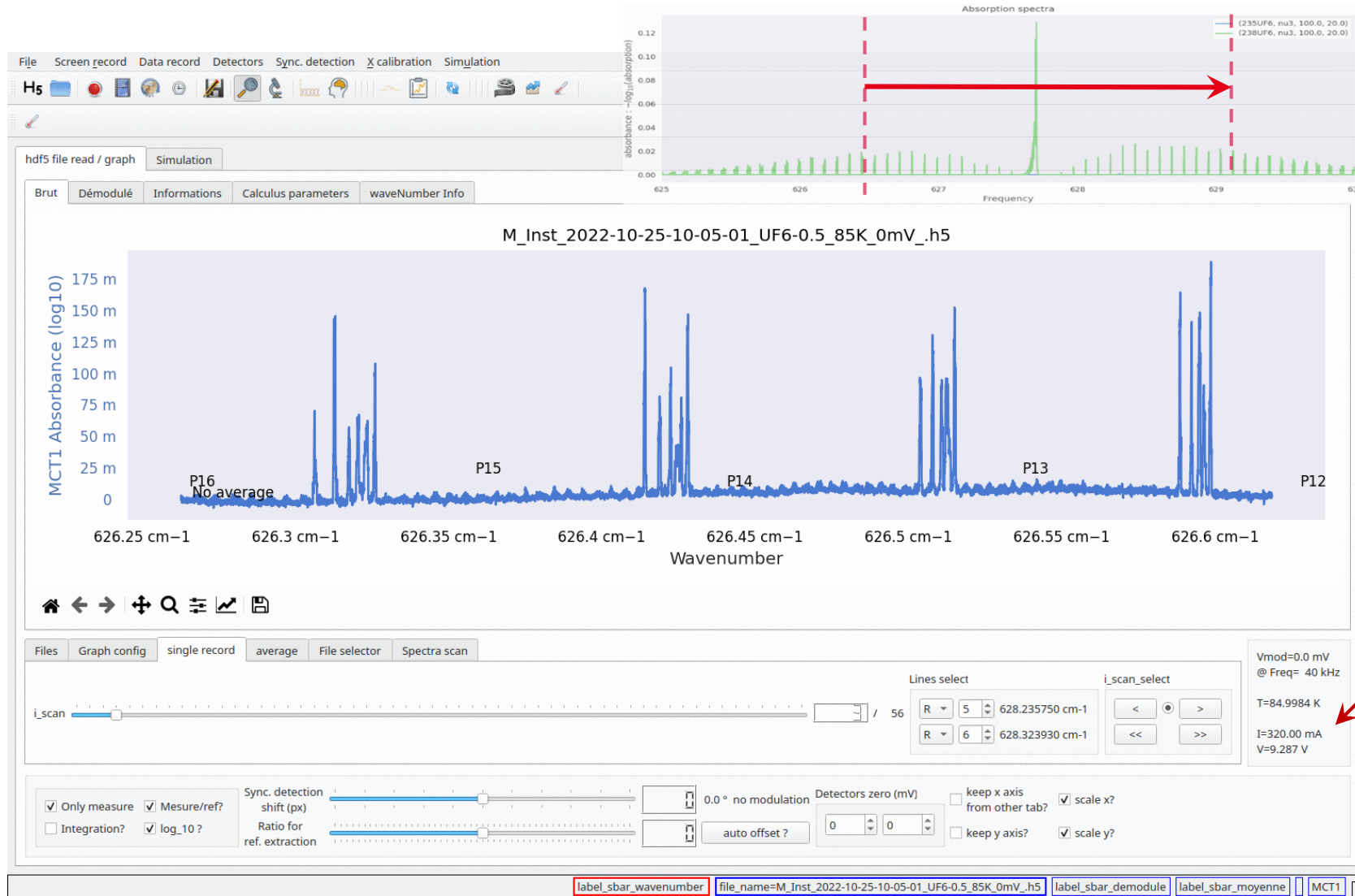


Beams are crossing the jet in a reduced volume

Pulsed jet



2.3 Wavelength Scan versus intensity variation



Intensity (mA)

3. Measurements

3.1 Raw measurement (without modulation) for LEU+, HALEU, HEU



3.1.1 : Rotational temperature measurement

Whole R branch and manual fit

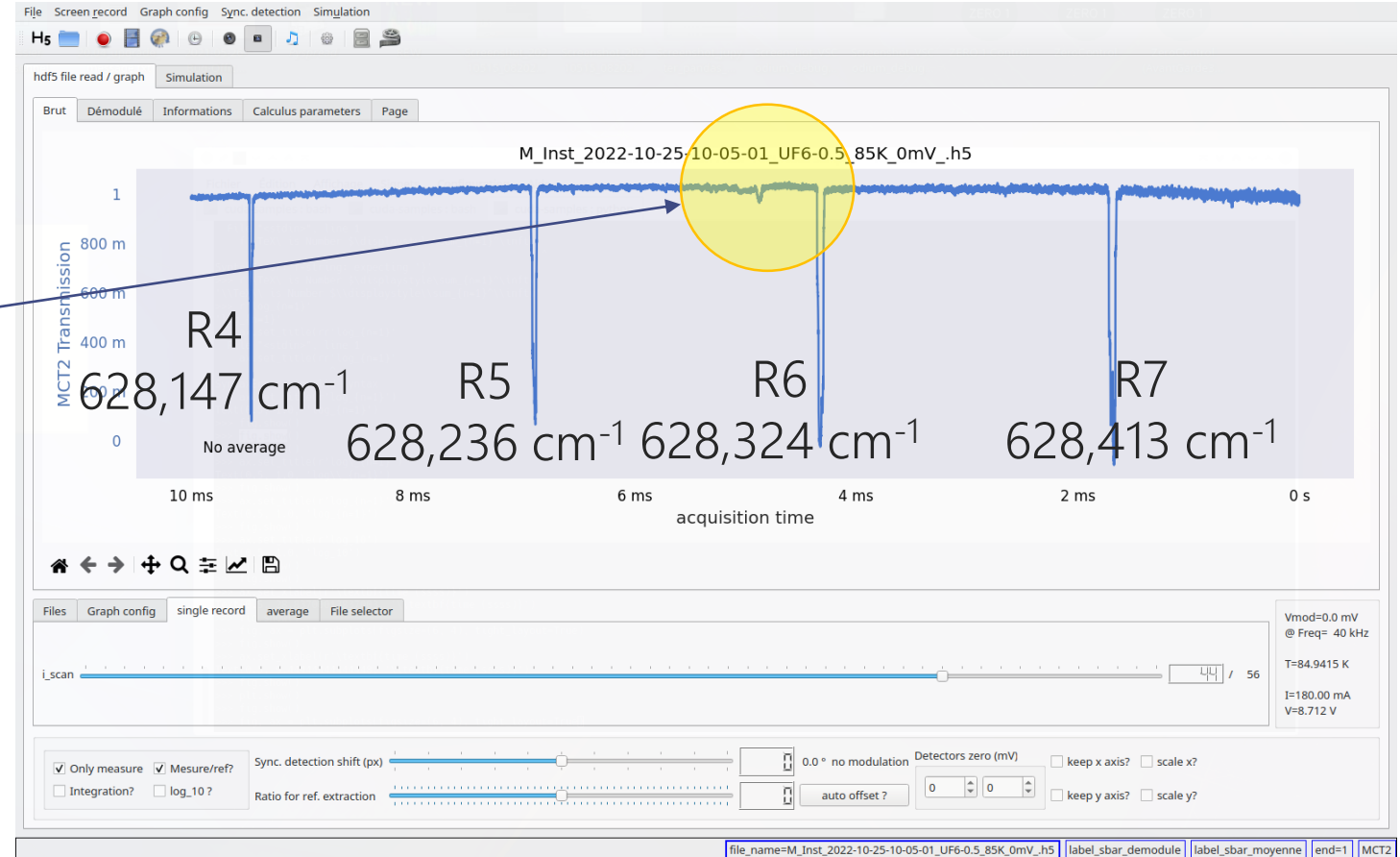
$^{235}\text{UF}_6$ branch Q signal in direct absorption

The choice of the range could be concatenated with others to cover the full spectrum

3.1.2 : Isotopic ratio

Low precision : Isotopic ratio = $\frac{\text{Mes}_{Q5}}{\text{Mes}_{Q8}}$

High precision : Isotopic ratio = $\frac{\text{Mes}_{\text{Full spectrum 5}}}{\text{Mes}_{\text{Full spectrum 8 + isotop shift}}}$



3. Measurements

3.2 for low assay => Natural up to LEU

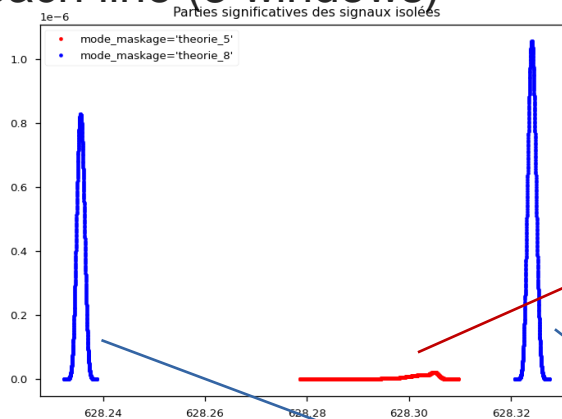
3.2.1 : Rotational temperature measurement

Fit automated by SVD analysis on noise simulation

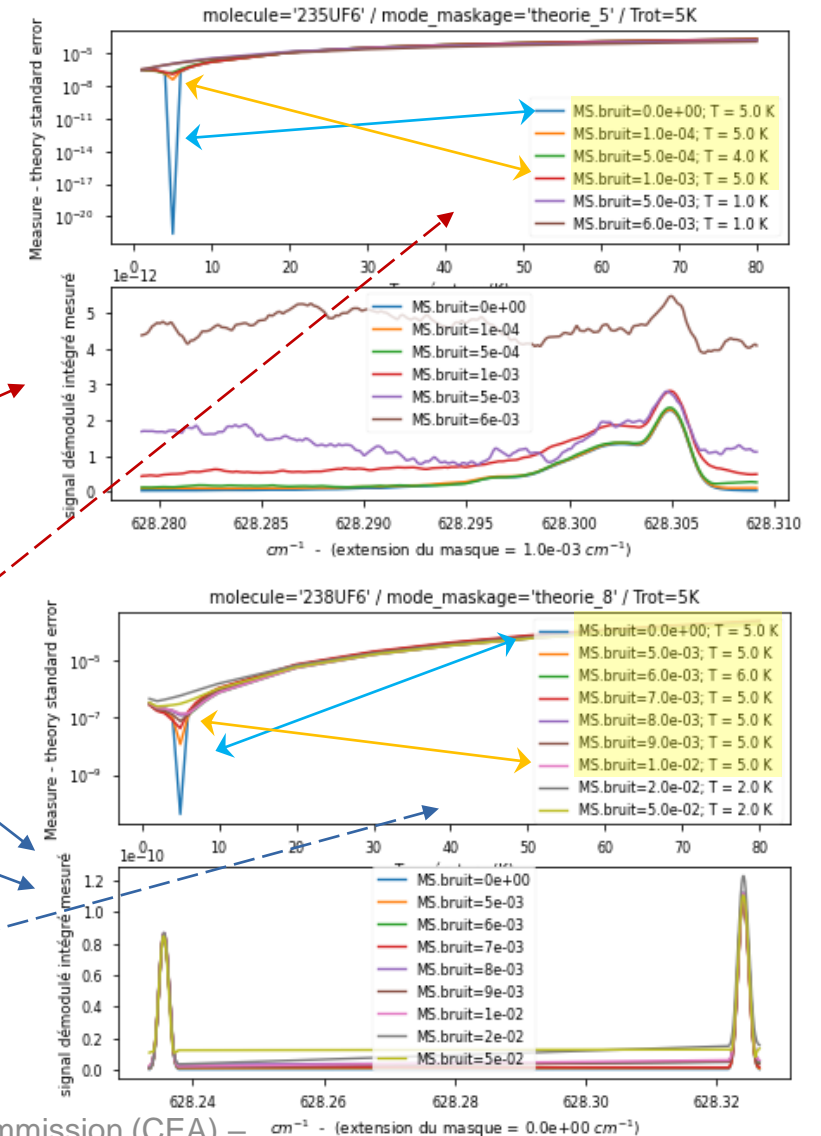
(HTDS model + noise before modulation)

=> First Choice of the window for each line (3 windows)

Use of :
235 branch Q
and
238 lines R5 R6



=> Numerical maximum noise determination
→ acceptable to assess temperature



Measurement signals with demodulation

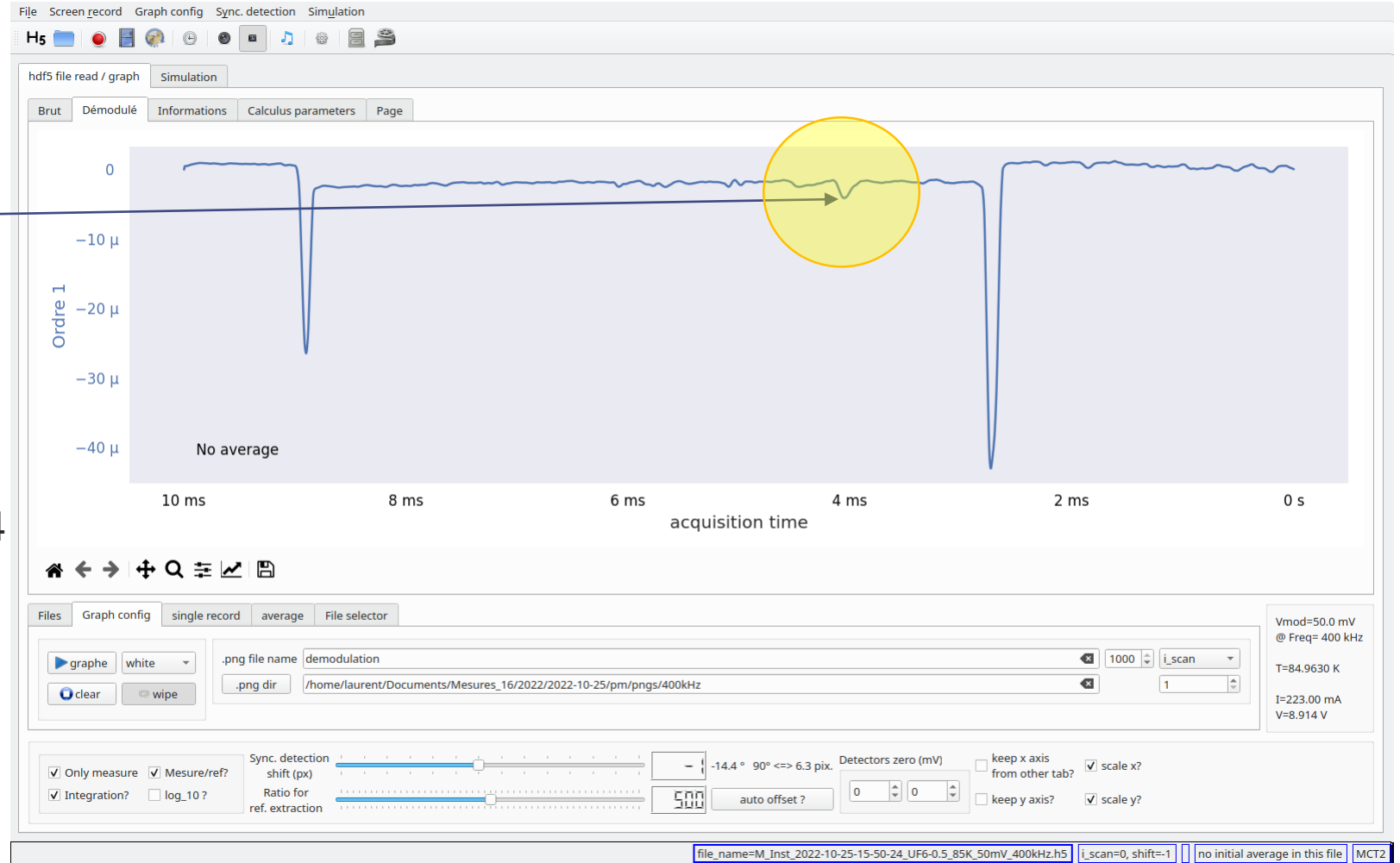
Scan with modulation (example 400kHz / 50mV) on 0,7% $^{235}\text{UF}_6$:

Cleaner $^{235}\text{UF}_6$ branch Q signal as obtained

3.2.2 : Isotopic ratio

$$I_{R5+R6} = f(T_{\text{rot}}), @5\text{ K} : \frac{I_{R5+R6}}{I_Q} = 0,214$$

$$\text{Isotopic ratio} = \frac{\text{Mes}_Q \times 0,214}{\text{Mes}_{R5+R6}}$$



Improvements over existing items



- ▶ Online automatic measurement :
 - ▶ Small tank filled with UF_6 + He mixture under pressure
 - ▶ Process execution
 - ▶ Drain tank to chemical trap
- ▶ Technical sovereignty
 - ▶ Non cryogenic QCL (MirSense)
 - ▶ No international patent
- ▶ High measurement accuracy expected **for mid assay** :
 - ▶ full spectrum analysis with HTDS(235) & HTDS(238)

Conclusion

▶ Current measurement accuracy **for low assay** :

$$n(^{235}\text{U}) / n(^{238}\text{U} + ^{235}\text{U}) = (0,7 \pm 0,3) \% \\ (=3 \cdot 10^{-3} \text{ absolute})$$

▶ Raw measurement without averaging and framing (commercial spectrometers do!)

▶ With averaging → expected gain in order of $< \sqrt{n}$

▶ Improvements :

▶ Carrier gas optimization

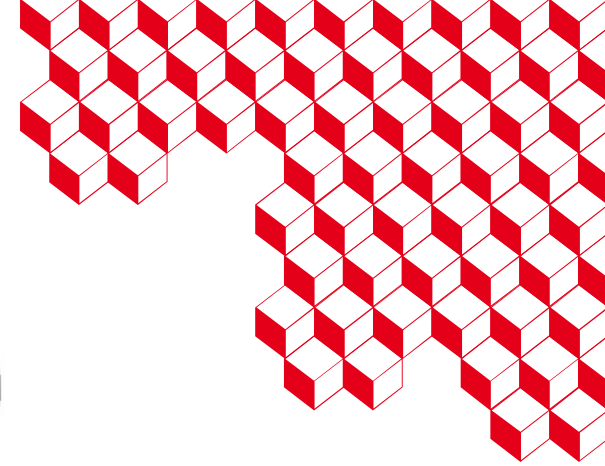
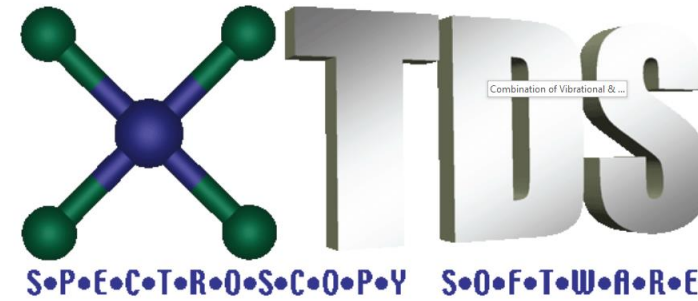
▶ Measurement averaging & calibration on reference standard

▶ No cryogenic QCL (MirSense)

▶ Volume reduction

▶ Measurement accuracy expected **for mid assay** :

▶ Much less than $3 \cdot 10^{-3}$ absolute...



Thanks

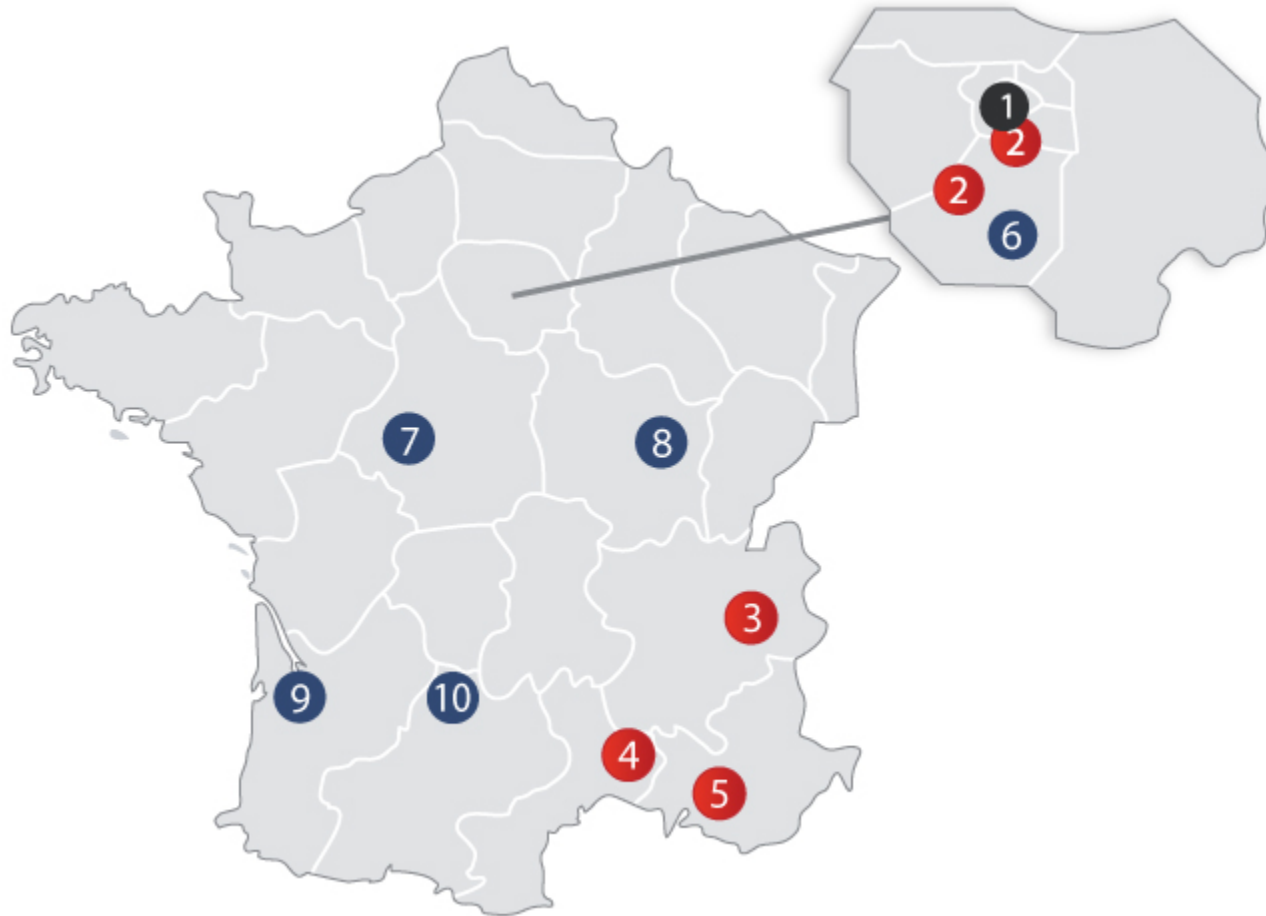
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CEA geographical locations



① SIÈGE SOCIAL

CENTRES D'ÉTUDE CIVILS

② Paris-Saclay
établissements de Fontenay-aux-Roses
et de Saclay

③ Grenoble

④ Marcoule

⑤ Cadarache

CENTRES POUR LES APPLICATIONS MILITAIRES

⑥ DAM Ile-de-France

⑦ Le Ripault

⑧ Valduc

⑨ Cesta

⑩ Gramat