

# **Tunable Diode Laser Spectroscopy Application for Detection and Isotopes Ratio Measurements of UF<sub>6</sub> Molecules.**

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# Introduction

Beginning of Tunable Diode Laser Spectroscopy (TDLS) was related to one molecule practically in all countries. It was  $\text{UF}_6$  molecule with respect to laser uranium isotopes separation. Dramatic progress in TDLS technique achieved during last decades and success of detection of trace complex molecules again brought our intention to the same molecule because of IAEA needs related to Additional Protocol of Safeguards.

There were three objectives of present paper:

1. To investigate if it is possible to use Tunable Diode Laser Spectroscopy (TDLS) technique for gaseous uranium hexafluoride enrichment measurement.
2. To analyze possibility to detect trace  $\text{UF}_6$  presence in atmosphere.
3. To detect trace HF molecule concentration due to hydrolysis of uranium hexafluoride in atmosphere.

# UF<sub>6</sub> Absorption Bands in mid IR

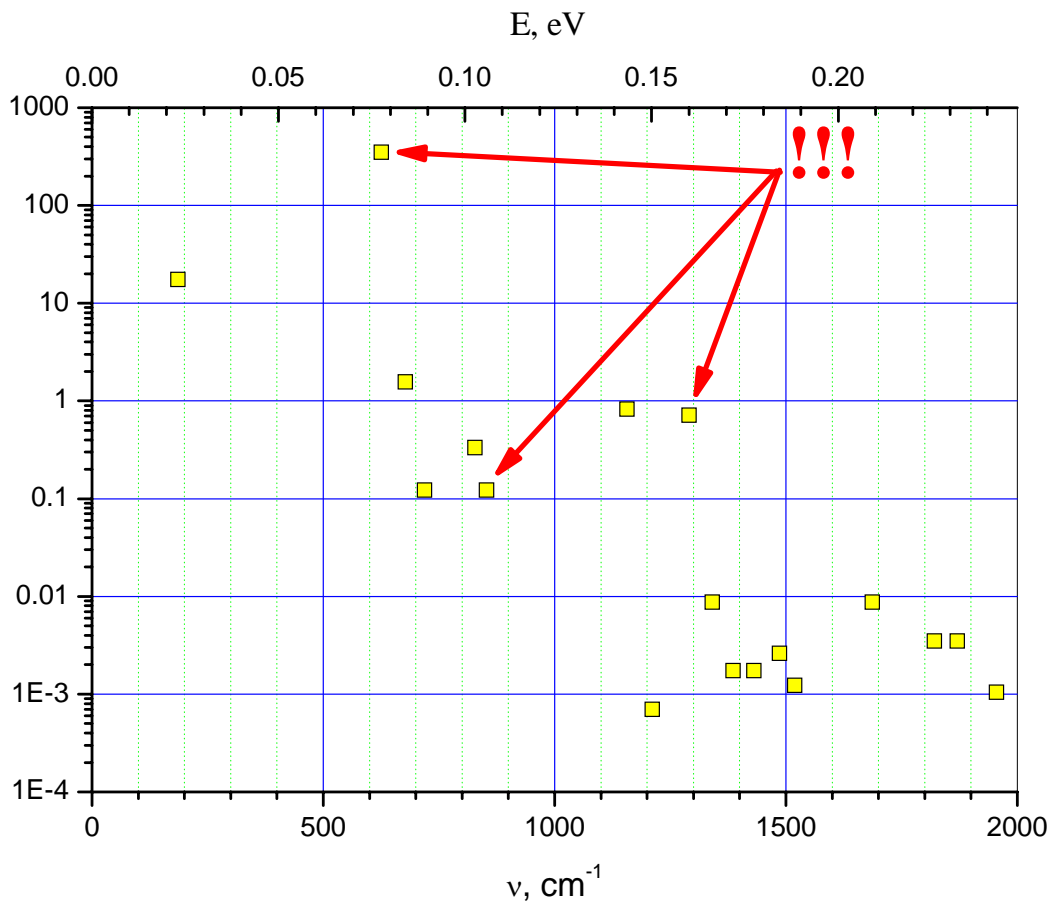
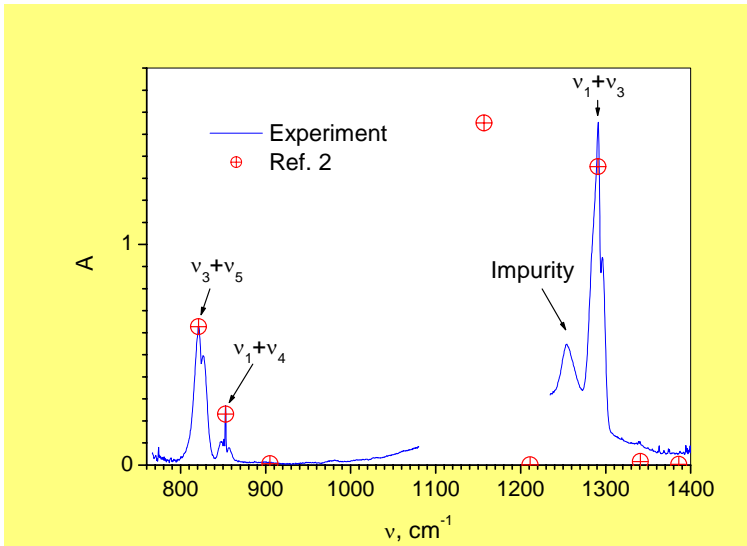


Figure presents absorption cross section of UF<sub>6</sub> molecule in mid IR.

Perspective absorption bands for enrichment measurement and trace uranium hexafluoride detection are marked by red arrows.

Assignment	$\nu$ , cm <sup>-1</sup>	$\sigma$ , cm <sup>2</sup> /atm	Application
$\nu_1+\nu_3$	1291	0.72	Enrichment measurement
$\nu_1+\nu_4$	853	0.12	Enrichment measurement
$\nu_3$	625	350	Trace detection

# Absorption bands identification



Assignment	$\nu$ , $\text{cm}^{-1}$	$\sigma$ , $\text{cm}^{-1}/\text{atm}$
$2\nu_3+\nu_6$	$1386\pm 2$	0.0018
$\nu_1+\nu_2+\nu_6$	1341	0.0088
$\nu_1+\nu_3$	$1290.9\pm 0.5$	0.72
$2\nu_2+\nu_6$	$1211\pm 2$	0.0007
$\nu_2+\nu_3$	$1156.9\pm 0.5$	0.82
$\nu_3+2\nu_6$	$905\pm 2$	0.0035
$\nu_1+\nu_4$	$852.8\pm 0.5$	0.12
$\nu_3+\nu_5$	821	0.33
$\nu_3$	625	350

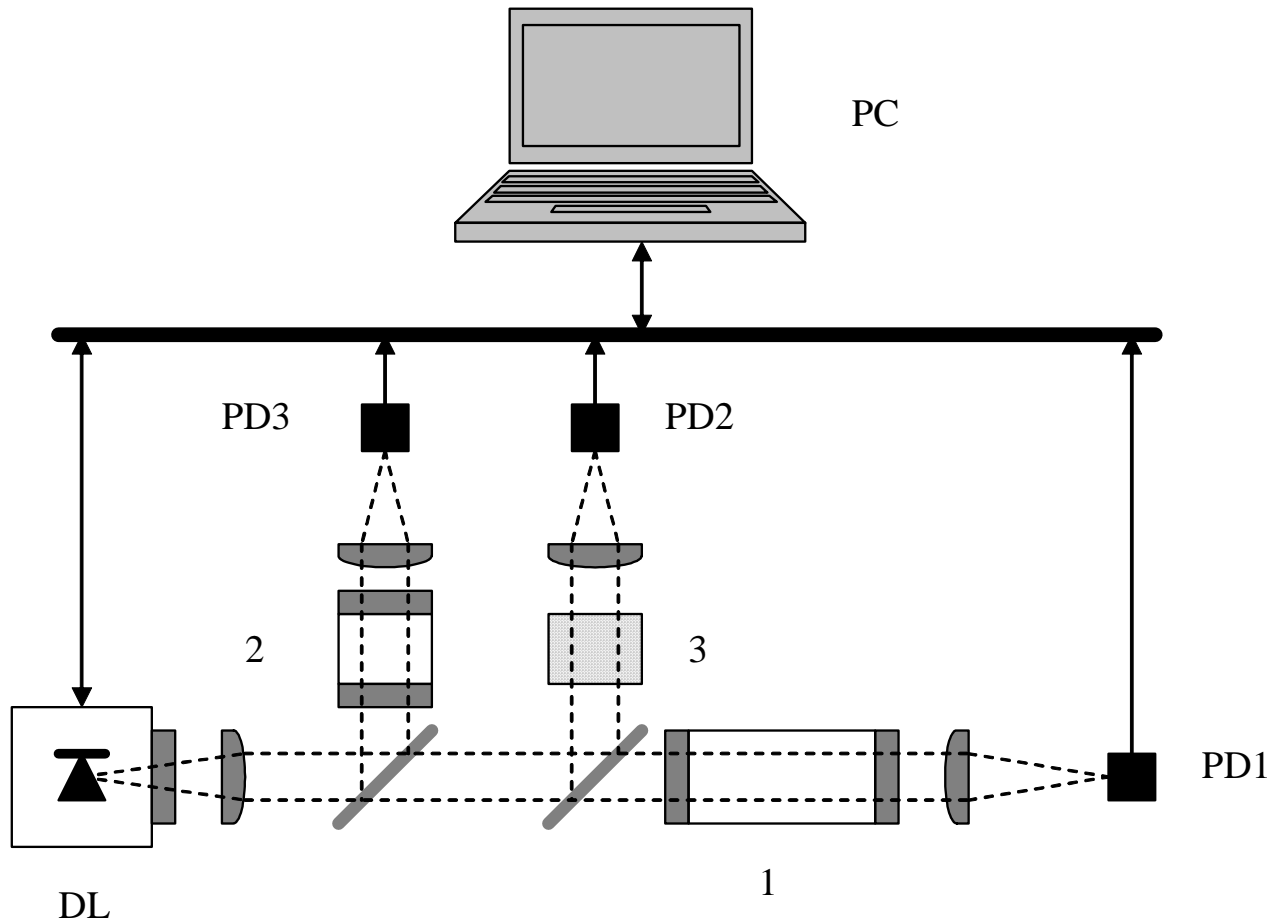
Absorption spectrum of gas mixture under investigation and observed spectral features identification.

Isotope shifts for several  $\text{UF}_6$  normal vibration modes.

Isotope shifts, $\text{cm}^{-1}$	$^{235}\text{UF}_6$ - $^{238}\text{UF}_6$
$\nu_1$	0
$\nu_3$	$0.65\pm 0.09$
$\nu_4$	$0.16\pm 0.09$

*Combination band  $\nu_1 + \nu_3$  was chosen for present experiment. Two other bands can be considered as alternative for  $\text{UF}_6$  enrichment measurements and trace detection*

# Instrument Block Scheme



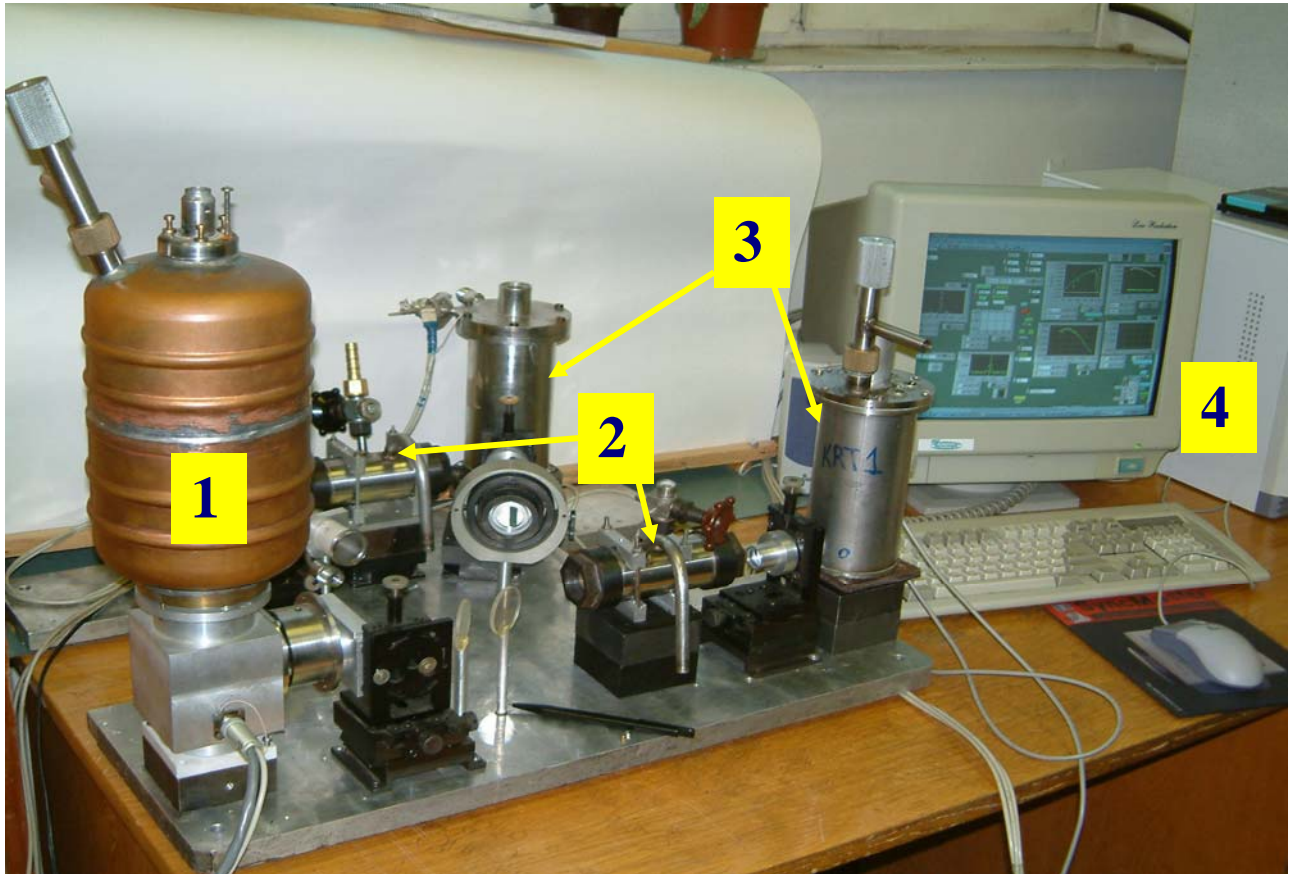
DL – diode laser; PD – photo detectors;  
PC – personal computer

Analytical channel with cell (1) containing gas under investigation.

Reference channels with cell (2) containing methane and acetylene, and with Fabry-Perot etalon (3) were used to determine laser frequency tuning.

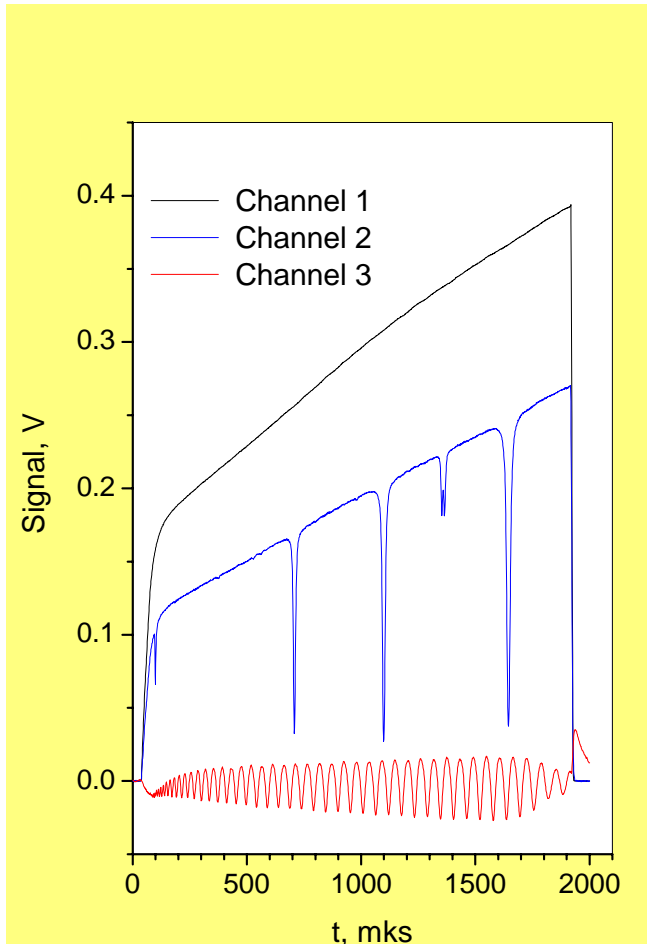
# Instrument view

Set up used to measure uranium hexafluoride enrichment



- 1 – diode laser in cryostat;
- 2 – analytical and reference channel with optical cells;
- 3 – photodetectors with preamplifiers in cryostat;
- 4 – electronic module with personal computer.

# Instrument operation mode



Diode laser was excited by trapezium current pulse provided DL frequency tuning. Signals from photodiodes in three instrument channels were recorded simultaneously. Signals from channels 2 (methane absorption) and 3 (Fabry-Perot etalon interference fringes) were used to determine DL frequency tuning curve. Final accuracy of tuning curve determination was found to be better than  $2 \times 10^{-4} \text{ cm}^{-1}$ .

Positions of reference gas lines in channel 2 signal were used as feedback signal in DL frequency tuning stabilization mode.

Using signal from channel 1  $\text{UF}_6$  absorption in sample cell was determined.

# Software development

Software was written using LabView.

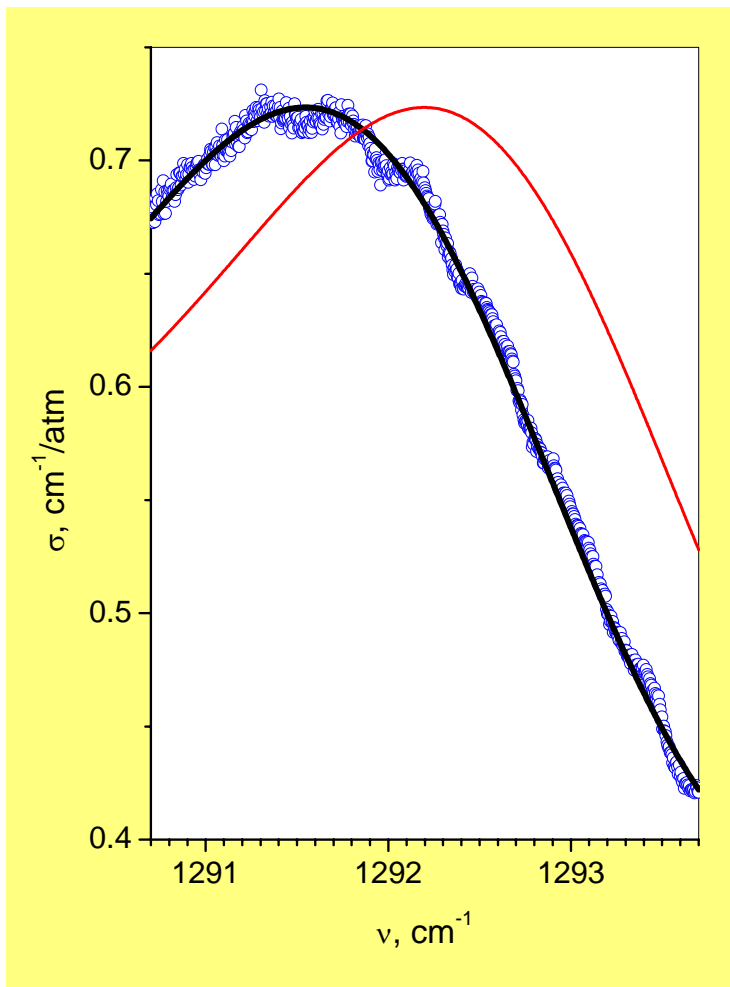
Software developed controlled all instruments options, recorded signals, performed data processing, and stored results obtained on hard disk.



Users interface of software developed to measure  $UF_6$  isotope ratio.



# Diode Laser Spectroscopy of UF<sub>6</sub>



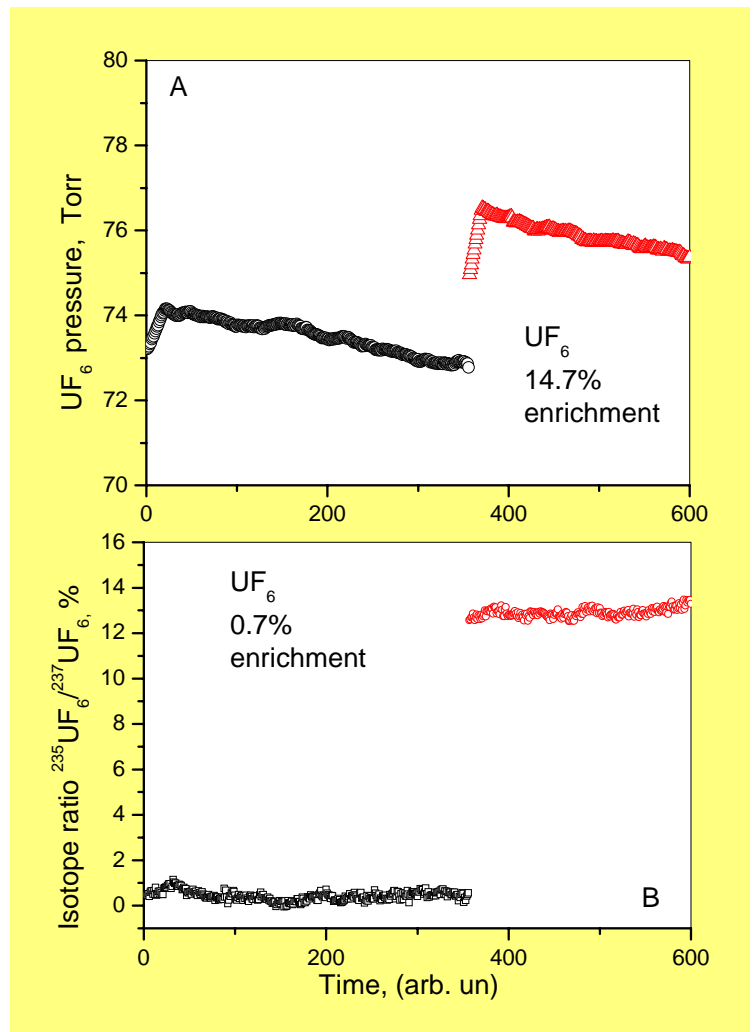
Spectra were recorded using TDL based instrument developed. Fragment of experimental absorption cross section spectrum (blue points) and model spectra for  $^{238}\text{UF}_6$  (black line) and  $^{235}\text{UF}_6$  (red line).

*Problem #2: Fine structure of UF<sub>6</sub> spectra was not included in the present model. Accurate spectrum of  $^{235}\text{UF}_6$  has to be measured.*

# Instrument Test at IAEA UF<sub>6</sub> Loop in Seibersdorf

Several UF<sub>6</sub> samples with calibrated enrichment were used. Figure shows results obtained for two calibrated samples: natural and high enriched ones. Sample cell was filled with calibrated gas and measurement procedure started.

Fig.A presents measured partial pressure, Fig.B shows enrichment.



*Accuracy in present case was limited by low quality diode laser and photo-diodes in use*

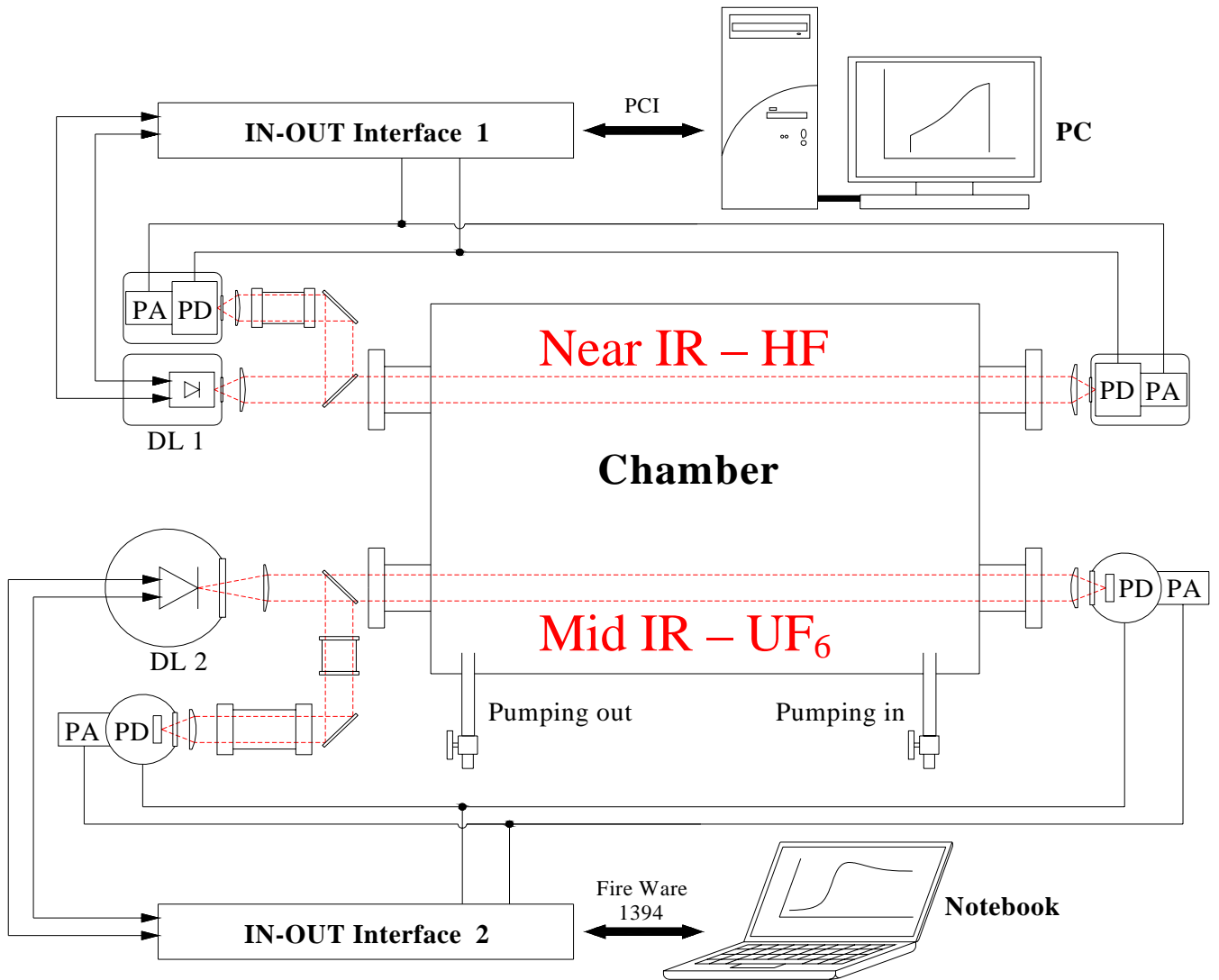
# UF<sub>6</sub> in Atmosphere

The necessity of detection of trace HF concentrations may arise in order to check the leaks of UF<sub>6</sub> from the containers or technological processes, as well as to reveal the illegal activity in the manufacturing of 235-isotope enriched uranium. When getting to the air, it reacts with atmospheric water to yield the HF:



HF is a volatile compound and can be detected with high sensitivity by diode laser based device. In this method the trace gas concentration is measured by scanning laser frequency over chosen molecular absorption line.

# Block Scheme of Experimental Setup



DL – diode lasers; PD – photo detectors;  
PA – preamplifiers; PC – personal computer

Block scheme of experimental setup developed for investigation of uranium hexafluoride behavior in atmosphere.

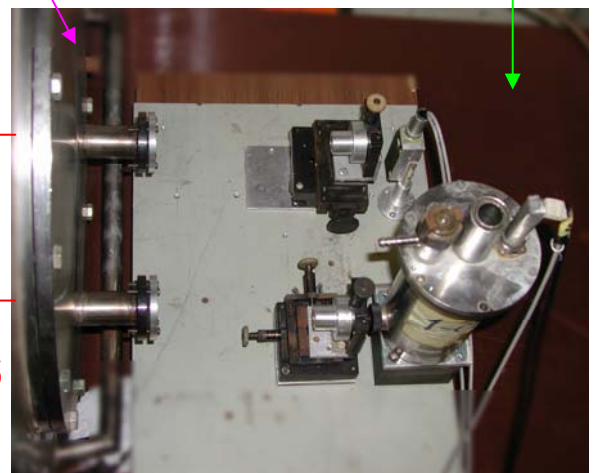
# Experimental Setup View



Diode Lasers

Photo Diodes

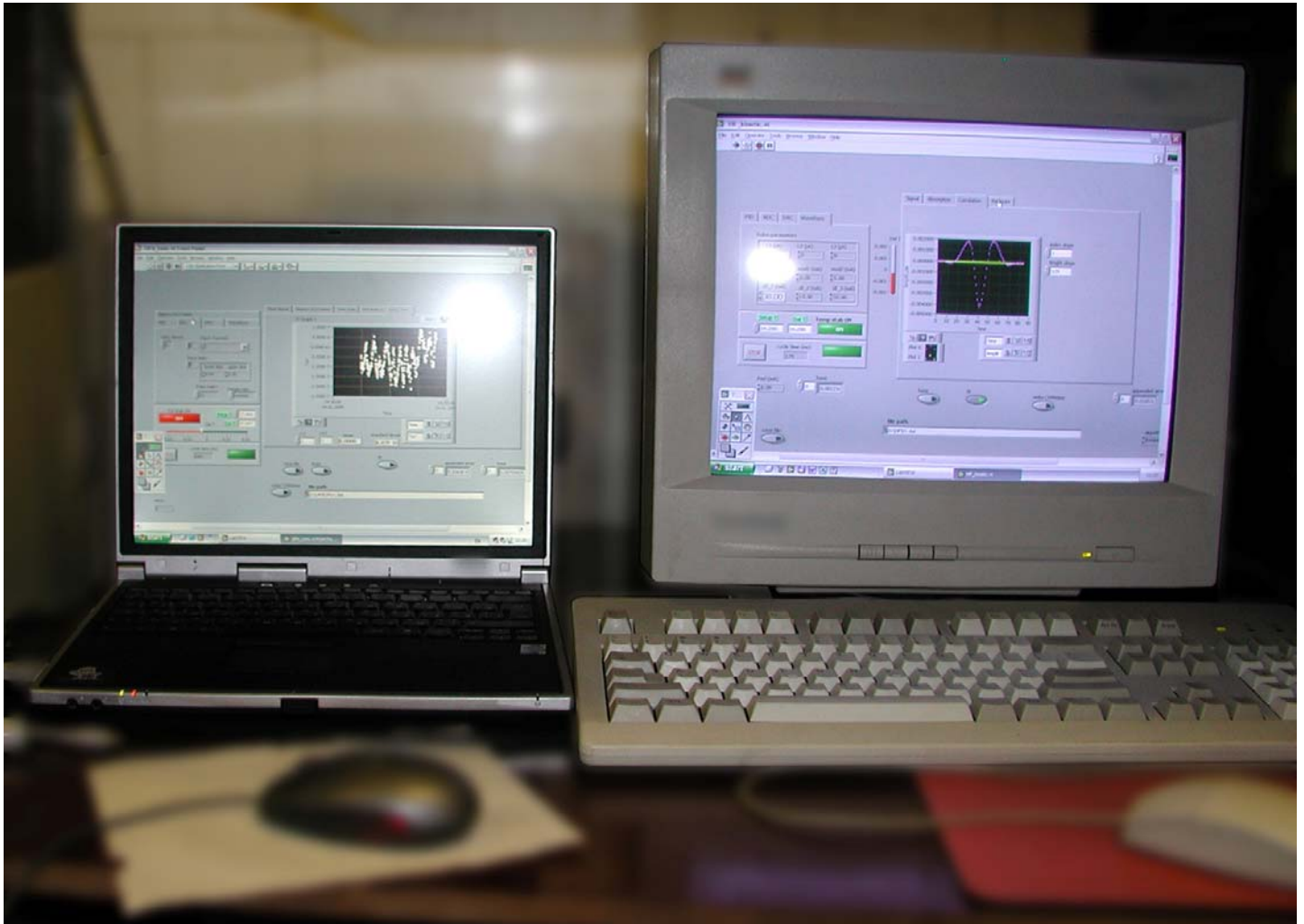
Chamber



Near IR – HF

Mid IR – UF<sub>6</sub>

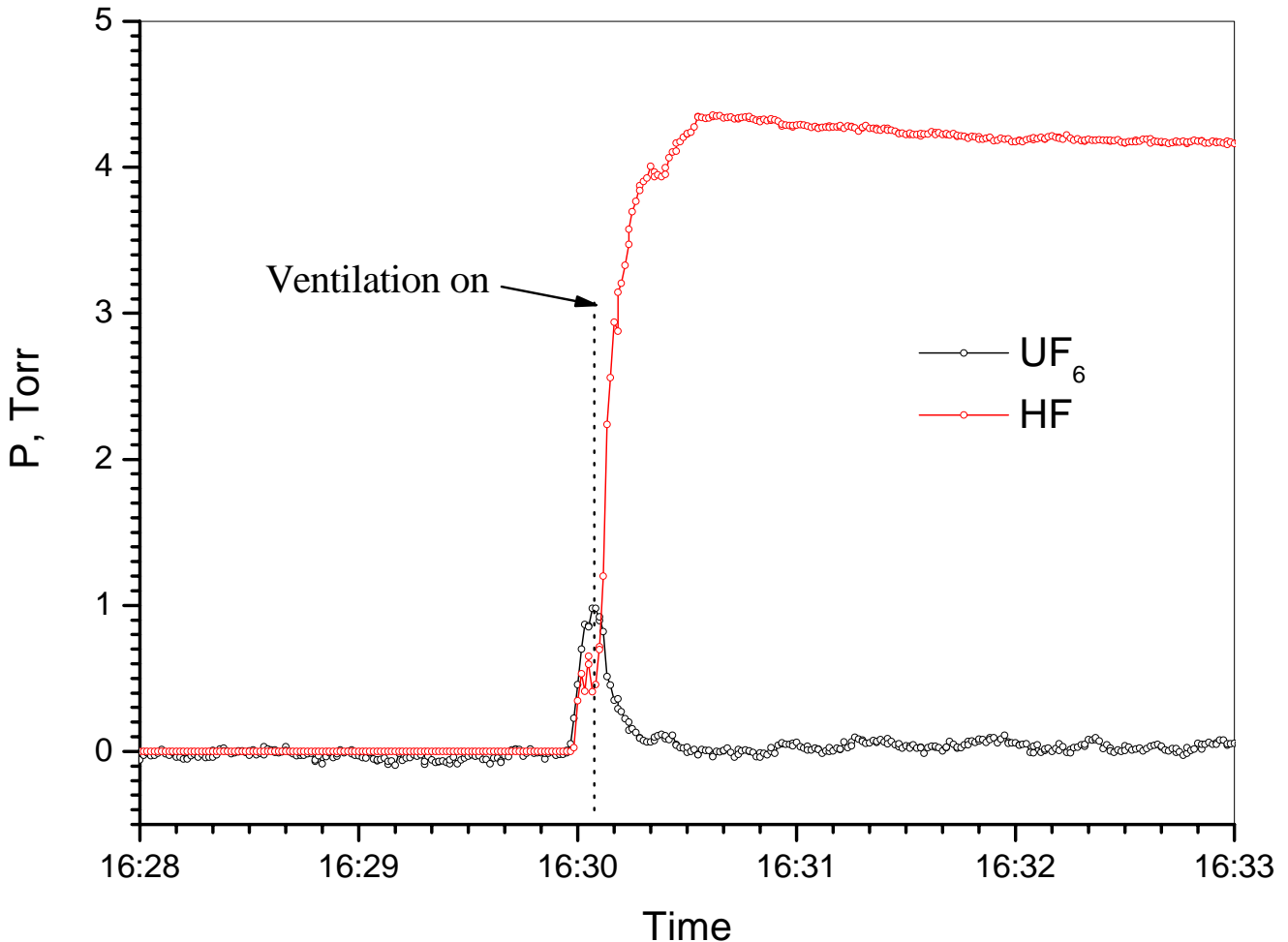
# Computers



Simultaneous measurement of  $\text{UF}_6$  (left) and HF (right) concentration

The PC was time synchronized during uranium hexafluoride hydrolyze experiment. Data concentration of  $\text{UF}_6$  and HF was wrote in data files to PC.

# Preliminary Results



Preliminary result of simultaneous measurement of trace UF<sub>6</sub> and HF concentration using experimental setup described.

*Resume: Local as well as remote monitoring of trace concentration of both UF<sub>6</sub> and HF trace concentrations can be performed using TDLS.*



# TDL based instrument modifications



**TDL based instrument for local measurements of trace molecule concentration**



**TDL based instrument for remote monitoring**