TDLS Operation Regimes and Data Processing

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The proper choice of the regimes of TDLS operation and data processing is very important in terms of maximal suppression of various noises and disturbances preventing from achieving the highest trace molecules detection sensitivity and measurement accuracy. This choice of optimal regimes depends on the type of dominant noises. DL frequency stabilization is important component of operation regimes in use.

DL frequency stabilization

DL frequency stabilization is important component of operation regimes in use.

It is combination of temperature stabilization at level 3 10⁻⁵ K and DL frequency tuning cycles stabilization using reference spectral line down to level of 1kHz



Allan plot of temperature as measured by termistor (left) and using spectral line (right). Both temperature stabilization and frequency tuning cycles stabilization were on.

TDLS operation regimes

TDLS operation regimes development was related to electronics available starting with all analog to all digital

#	Current	Signal	Electronics
1	analog	analog	Lockin, strob-integrator
2	analog	digital	Transient recorder
3	digital	digital	DAQ board

Most of recent TDLS regimes are based on DAQ boards. DL is excited by periodic excitation current with special waveform. Computer generates array for this waveform:

$$\{I(n)\}, \qquad n=1,\dots N$$

This array can be considered as vector in N – dimensional space (N is number of array elements). Hence, all approaches of vector algebra can be used. Based on this classification generation #4 of TDLS operation regime can be introduced when excitation current generated by computer can be represented as matrix:

$$\{I(n,m)\}, \qquad n = 1,...N; \qquad m = 1,...M$$

Here M is number of channels if several DL or number of waveforms involved in averaging process. Technical components for G #4 are available now (computer station).

Excitation current pulse waveform



Excitation current with white noise modulation



Excitation current waveform with modulation

PC generated excitation current can be presented as vector in N – dimensional space.





Most typical excitation current waveform in use: "0" – current below threshold value; N = 3-10000 (100 in present case). Modulation is important component. DL excitation current (solid blue line) and times of synchronal recording (vertical green dotted lines) of temperature, reference and analytical channels signals, respectively.

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Time multiplexing regime



For time multiplexing regime, excitation current generated by PC can be presented as matrix in N*M dimensional space.

$$\{I(n,m)\}, n = 1,...N; m = 1,...M$$

In a given case three DL were used to detect NH_3 (1.52 μ), CH_4 (1.65 μ), and CO_2 (1.6 μ). Part of DL radiation was directed into reference channels containing cells with reference gases and recorded by three photodiodes (three upper graphs).

Radiation from all lasers was directed onto a topographic reflector (50 m from the system). Scattered radiation was received by telescope and recorded by a PD (the lower graph). Only one laser operates at each moment of time (time multiplexing). This regime allows simultaneous concentration measurement of several molecules (up to 6 in an existing configuration).

Signal recording and averaging

DL is exciting by periodic current having one of waveforms considered above. Pulses train containing from 1 to 1000 pulses is recorded (red, 20 pulses in present case)



To reduce Flicker type noise and drifts averaging is performing over realizations: averaging of pulses in train.

Unstable external illumination



Example of unstable external illumination

Instrument was installed on helicopter and received DL light scattered by topography reflector (small pulses on the figure).

Pronounced time variations of photocurrent - i are determined by external illumination changing. Origin of this changing is Sun light albedo variation during helicopter flight. Other example of similar source is PD harmonic illumination by electrical light.

Preliminary data processing



Module "ZERO": external illumination subtraction



Allan plot of relative signal in presence of harmonic external illumination before (solid blue squares) and after (open red circles) "ZERO" module

Unstable recorded DL intensity



Example of unstable recorded DL intensity

Instrument was installed on helicopter and received DL light scattered by topography reflector (pulses on the figure).

Unstable external illumination also can be observed as baseline.

Pronounced time variations of photocurrent pulses are determined by albedo changing of area scattering DL light during helicopter flight. Other example of similar source is modulation of recorded signal due to mechanical vibrations in optical setup.

"EvenOdd" module

Recorded signal waveforms without (red solid circles) and with (open blue circles) molecular absorption



To calculate arrays "Odd" and "Even" information about three points (i-1, i, i+1) is using

$$n-1 = 2i-1 \qquad n = 2i \qquad n+1 = 2i+1$$

$$Even \quad \frac{1}{2}(S_{n-2} + S_n) \qquad S_n \qquad \frac{1}{2}(S_n + S_{n+2})$$

$$Odd \qquad S_{n-1} \qquad \frac{1}{2}(S_{n+1} + S_{n-1}) \qquad S_{n+1}$$

$$FFT_{Even}(k) = \left[1 + \cos\left(\frac{2\pi}{N}k\right)\right] \frac{FFT_s(k)}{\sqrt{2}}$$

$$FFT_{Even-Odd}(k) = \left[1 + \cos\left(\frac{2\pi}{N}k\right)\right] FFT_s(k)$$

The procedure advantages: number of arrays elements is N-2 (close to N); elements of Even and Odd arrays correspond to the same time (important for Flicker noise suppression).

Preliminary data processing

Module "EvenOdd": intensity variations and Flicker noise suppression



Harmonic modulation of relative signal (solid blue squares) due to generated mechanical vibrations in optical setup. Flicker type noise of DL radiation at 3 10⁻⁵ level can also be observed.

However, measurements of absorption or relative signal are our objective:

EvenOdd (n) =
$$\left[\frac{2S_n - (S_{n+1} + S_{n-1})}{2S_n + (S_{n+1} + S_{n-1})}\right] (-1)^n$$

Total suppression of both harmonic intensity variations and Flicker noise was achieved by EvenOdd module operation (red open circles)

Preliminary data processing Optimal filter

Theorem:

Spectrum of optimal filter is ratio of signal spectrum to noise spectrum.

Noise spectrum for Flicker noise – 1/f

For frequency domain - signal is spectral line



Hence, optimal signal filtering is cross-correlation function of recorded absorption derivative in reference channel with analytical one.

- Recorded signal in reference channel with spectral line under detection
- 2. Result of EvenOdd module operation for reference channel signal
- 3. Auto-correlation function for reference channel

Minimum detectable absorption below 10⁻⁷

Using operation regime and data processing considered above fundamental limit of absorption detection due to diode laser quantum noise was achieved: below 10^{-7} for averaging time above 1 sec (best presented in this poster result is equal <u>6 10⁻⁸</u> for 5 sec averaging time).



Frequently (in photo-acoustic and ring-down spectroscopy) minimum detectable absorption coefficient is considered. This parameter is equal to $6*10^{-12}$ cm⁻¹ for our system (Chernin multi-pass cell in use: 0.5 m, 200 passes) and it is comparable with the best known results obtained in Stark spectroscopy.

Next sensitivity parameter widely used in literature is minimum detectable molecular concentration. For example, above mentioned sensitivity for HF molecule corresponds to minimum detectable concentration <u>0.8 ppt</u>.