Single-pulse high-resolution spectroscopy on NovoFEL: methods, applications and development

V. Kubarev [1,2], A. Bragin[1], G. Sozinov[1],
E. Chesnokov [3], P. Koshlyakov [3]

[1] Budker Institute of Nuclear Physics, Novosibirsk, Russia,
[2] Novosibirsk State University, Novosibirsk, Russia,
[3] Institute of Chemical Kinetics and Combustion, Novosibirsk, Russia

Outline

- **Introduction:**
  - – motivation of ultrafast single-shot spectroscopy
  - – key elements of the spectroscopy
- **Free induction decay as basic of the spectroscopy**:
  - – exotic forms of FID signal
- **Different types of the spectroscopy:**
  - – Simple analytical spectroscopy a priory known spectra
  - – Common spectroscopy a priory unknown spectra
  - – Spectroscopy in magnetic field

Motivation and key elements

Motivation:

Ultrafast real-time spectroscopy is necessary in investigation of unrepeatable or single-pulse processes where classical well known methods spectroscopy based on sampling technology can't be applied (loss information because of averaging).

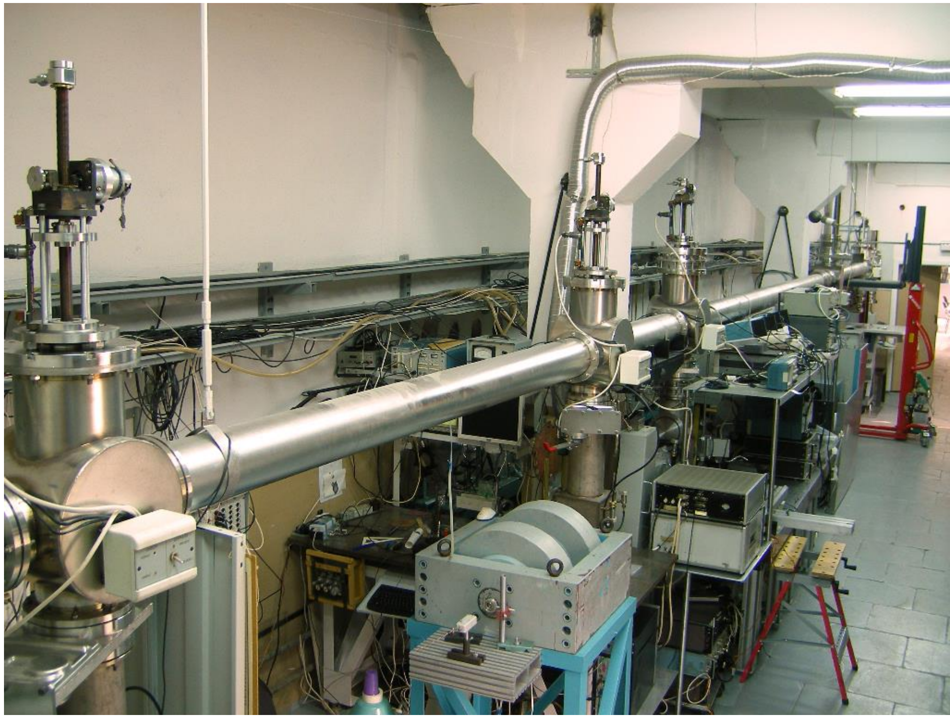
Key elements:

- Powerful THz pulse source with qualitative beam (linear polarized gauss beams) – THz NovoFEL
- Ultrafast detector – special Schottky diodes
- Ultrafast direct oscilloscope – LeCroy 30 GHz (300 k$)

Novosibirsk terahertz free electron laser (THz NovoFEL)

Accelerator hall

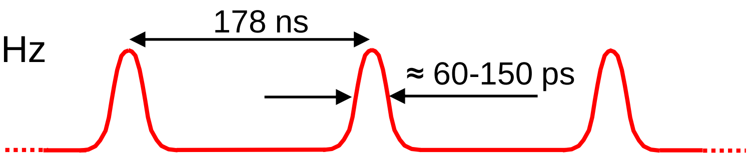
User's hall



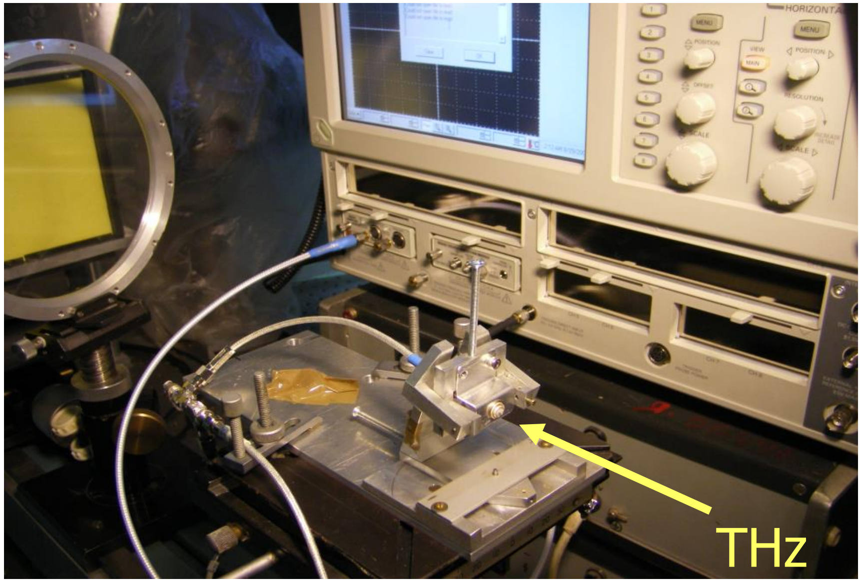
Radiation parameters of THz NovoFEL:

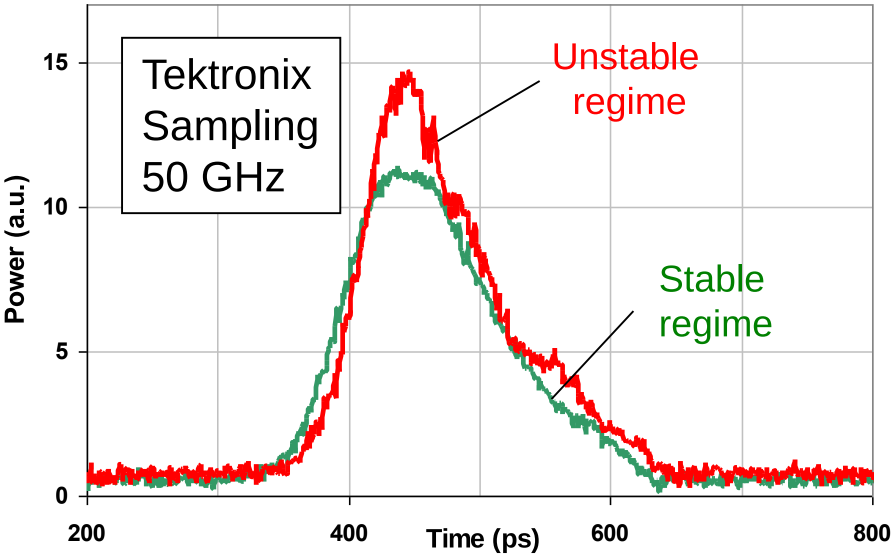
λ = 90 – 240 μm, $P_{average} \leq$ 500 W, $P_{pulse} \leq$ 0.9 MW, $(\Delta\lambda/\lambda)_{min} = 2\cdot10^{-3}$, f $\leq$ 22.4 MHz

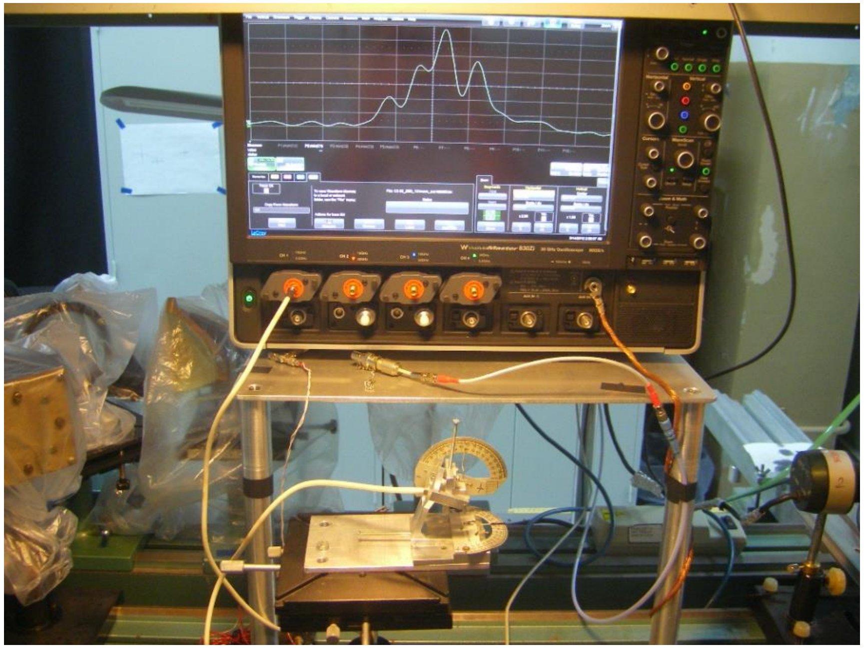
Routine regime of THz NovoFEL: f = 5.6 MHz

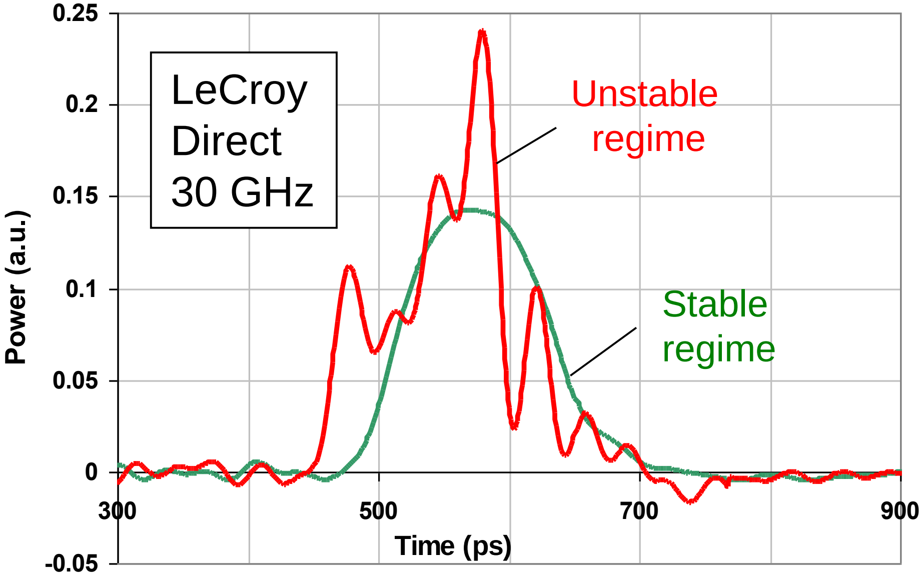


Ultra-fast THz Schottky diode detector and oscilloscopes

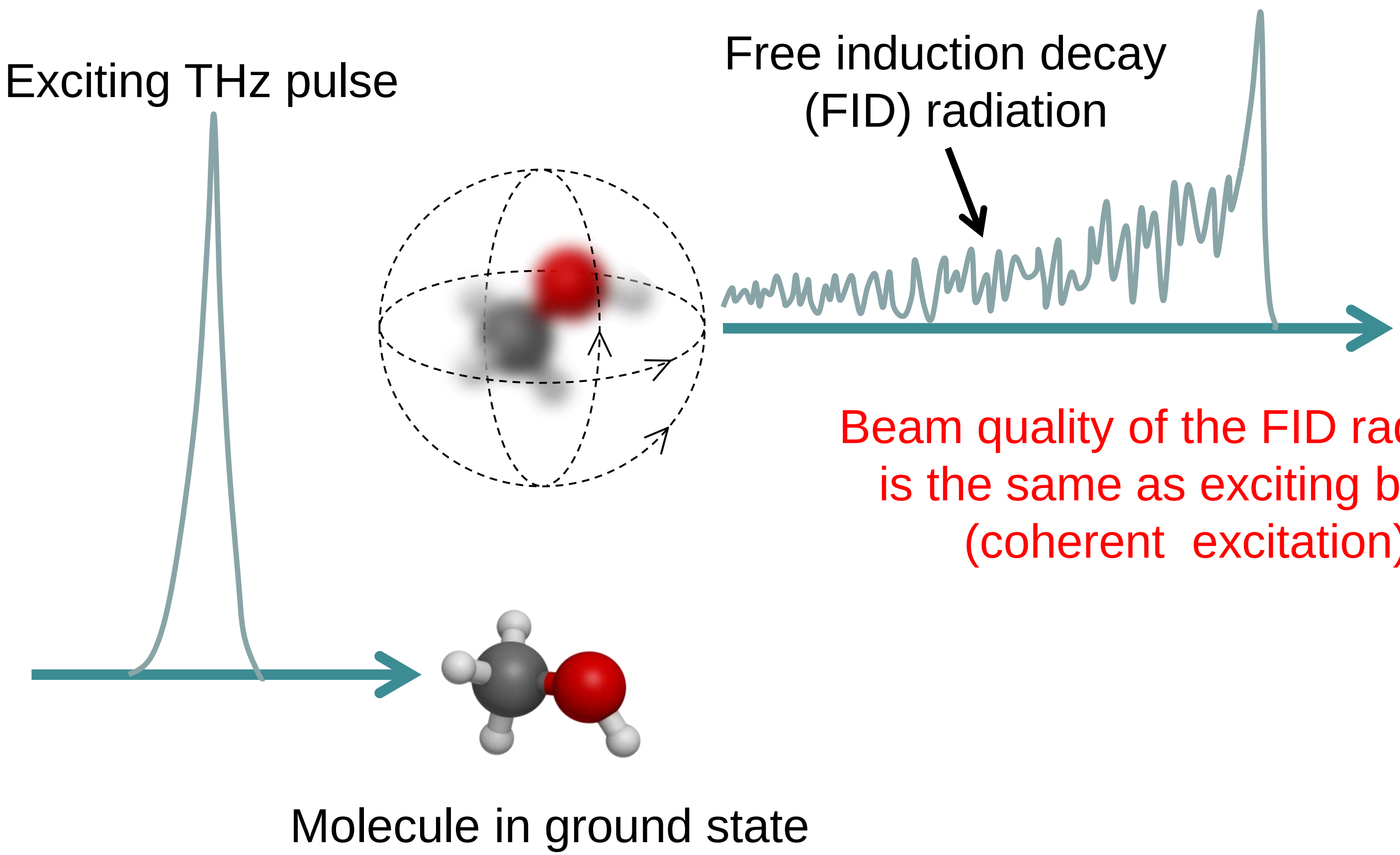








Scheme of free induction decay radiation (FID) of molecules



Beam quality of the FID radiation
is the same as exciting beam
(coherent excitation)

Chesnokov E.N., Kubarev V.V., Koshlyakov P.V., and Kulipanov G.N.
"*Direct observation of the terahertz optical free induction decay of molecular rotation absorption lines in the sub-nanosecond time scale*",
Appl Phys Lett 101 (2012) 131109-(1-4).

Theoretical model of the free induction decay

Basis: Lorentz dispersion theory of gases and Fourier transform:

$$n(\omega) = n_r(\omega) - i n_i(\omega) = 1 + \sum_m A_m \frac{(\omega_m - \omega)\gamma_m - i\gamma_m^2}{(\omega_m - \omega)^2 + \gamma_m^2}$$

$$\alpha(\omega) = \frac{\omega n_i}{c} = \frac{\omega}{c} \sum_m A_m \frac{\gamma_m^2}{(\omega_m - \omega)^2 + \gamma_m^2}$$

$$\Delta k(\omega) = \frac{\omega}{c}(n_r - 1) = \frac{\omega}{c} \sum_m A_m \frac{(\omega_m - \omega)\gamma_m}{(\omega_m - \omega)^2 + \gamma_m^2}$$

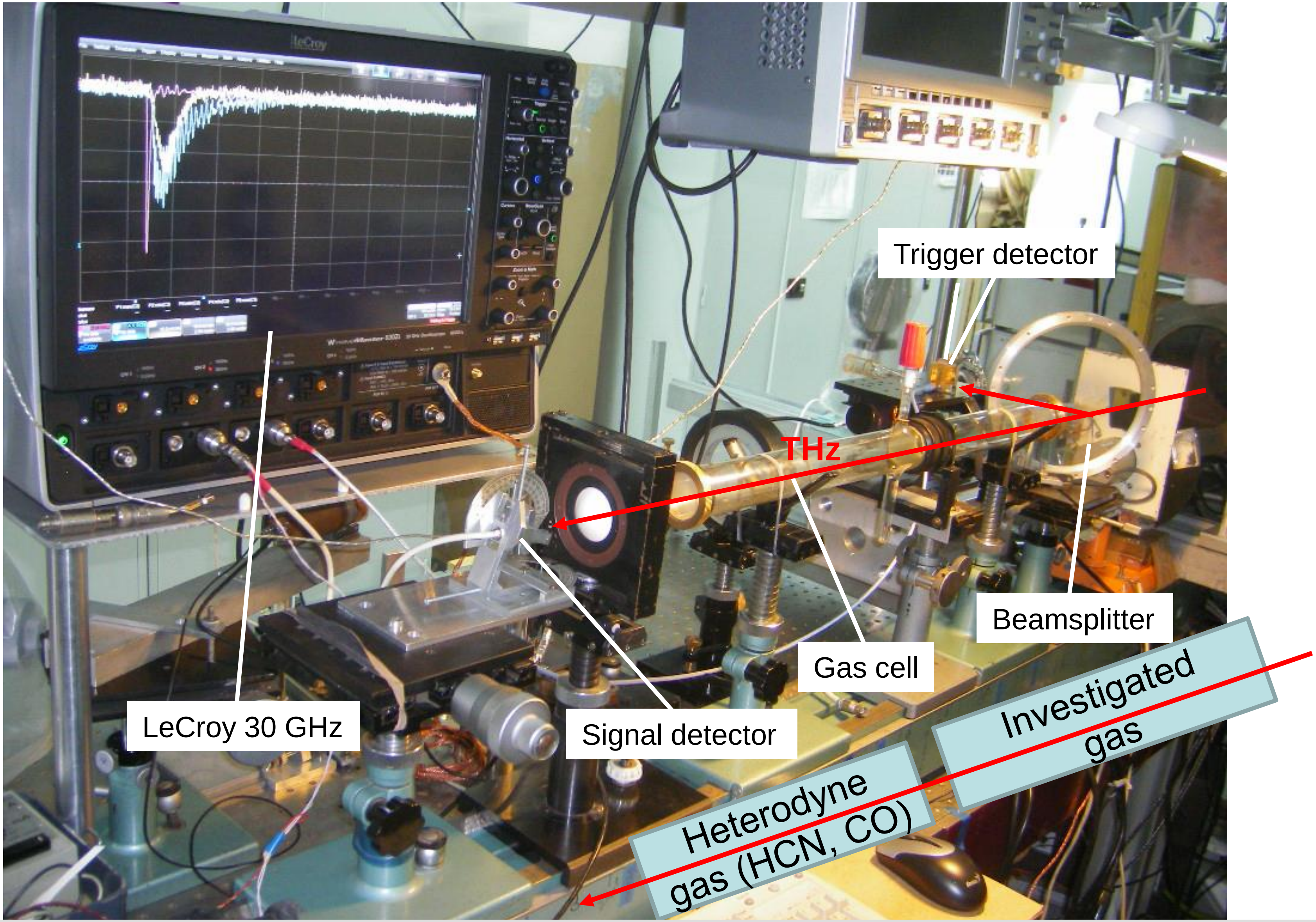
$$E(\omega) = E_0 \cdot \exp\left[-\frac{(\omega - \omega_0)^2 \tau^2}{8}\right]$$

$$\tilde{E}(\omega) = E(\omega) \cdot \exp\left[-\alpha(\omega) L\right] \cdot \exp\left[i \Delta k(\omega) L\right]$$

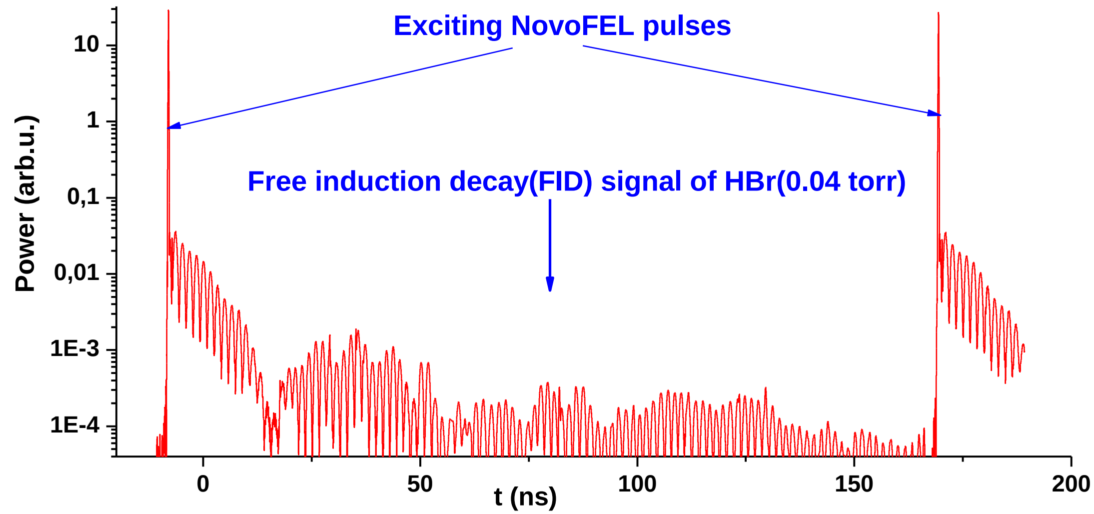
$$E(t) = \frac{1}{2\pi} \int_{-\infty}^{\infty} E(\omega) \cdot e^{-\alpha(\omega)L + i\Delta k(\omega)L} \cdot e^{-i\omega t} d\omega$$

Free induction decay of rotational transitions in molecules

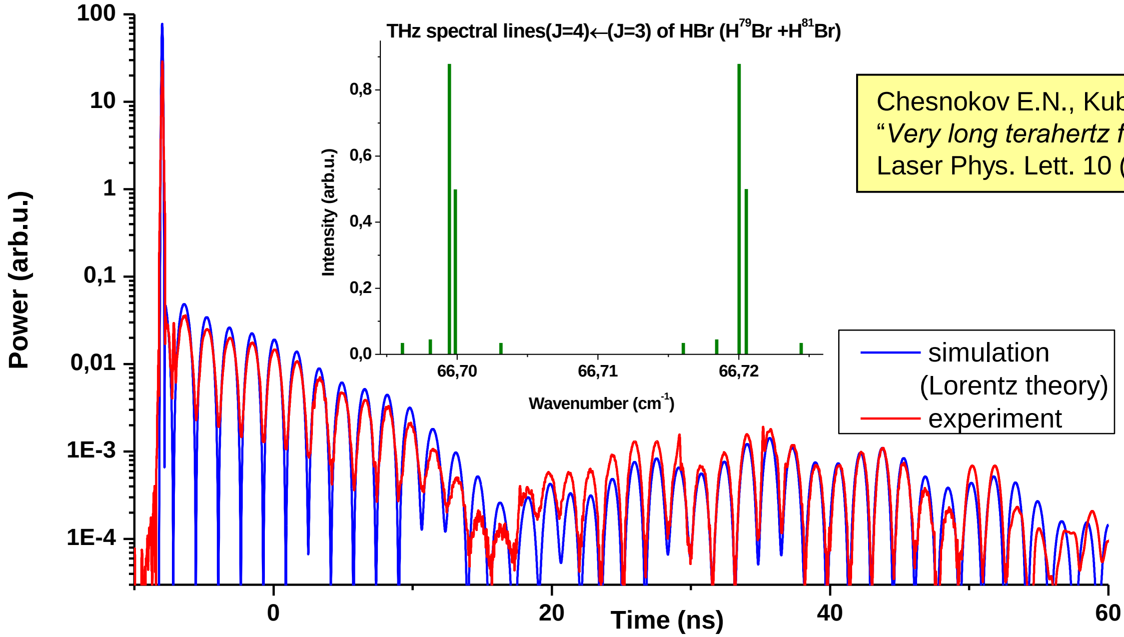
Experimental setup:



Very long free induction decay of HBr molecules

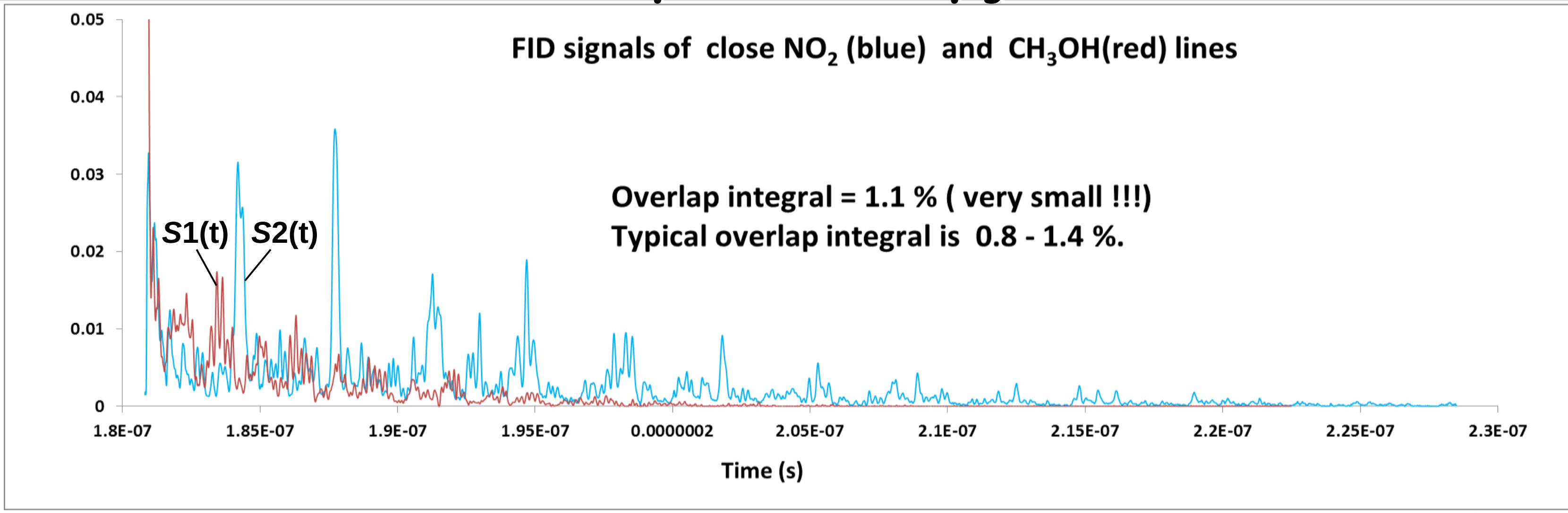


$(\Delta f / f)_{min} = (2\text{-}4) \cdot 10^{-6}$

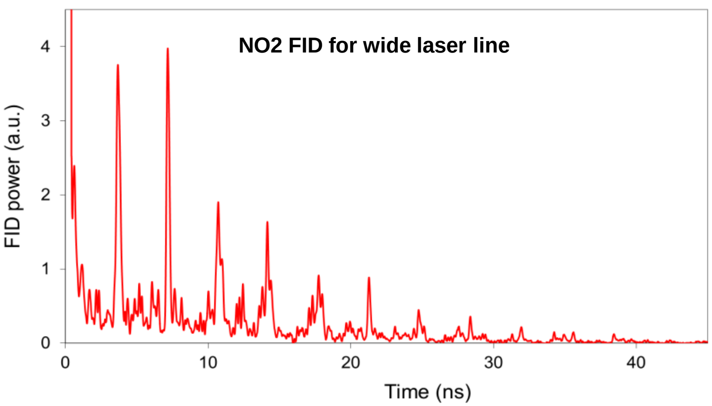


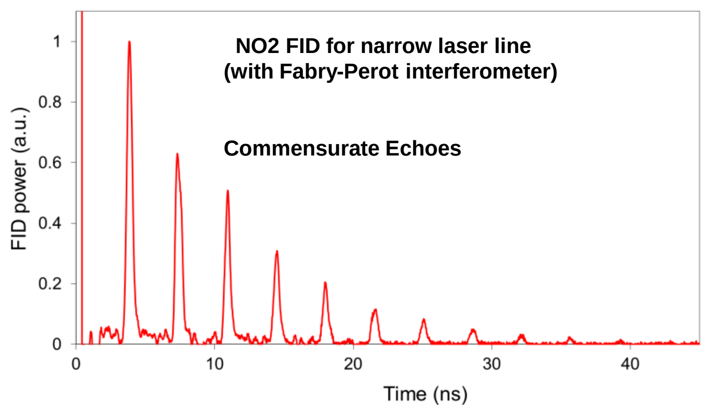
Chesnokov E.N., Kubarev V.V., Koshlyakov P.V., Kulipanov G.N., "*Very long terahertz free induction decay in gaseous hydrogen bromide*", Laser Phys. Lett. 10 (2013) 055701.

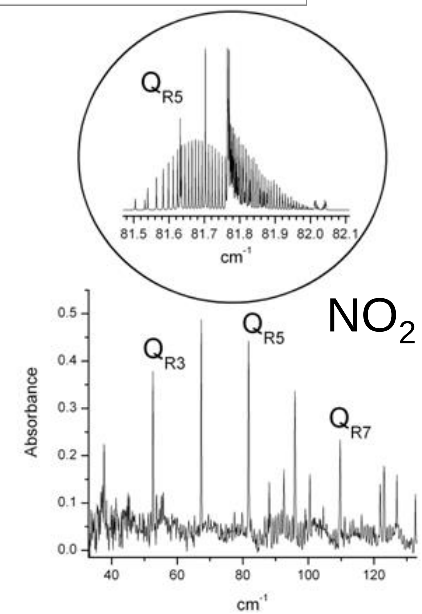
Commensurate frequencies and simple analytical spectroscopy



$$\textit{Overlap integral} \equiv \frac{\int S1(t)\cdot S2(t)dt}{(\int S1(t)^2 dt)^{1/2}\cdot(\int S2(t)^2 dt)^{1/2}}$$

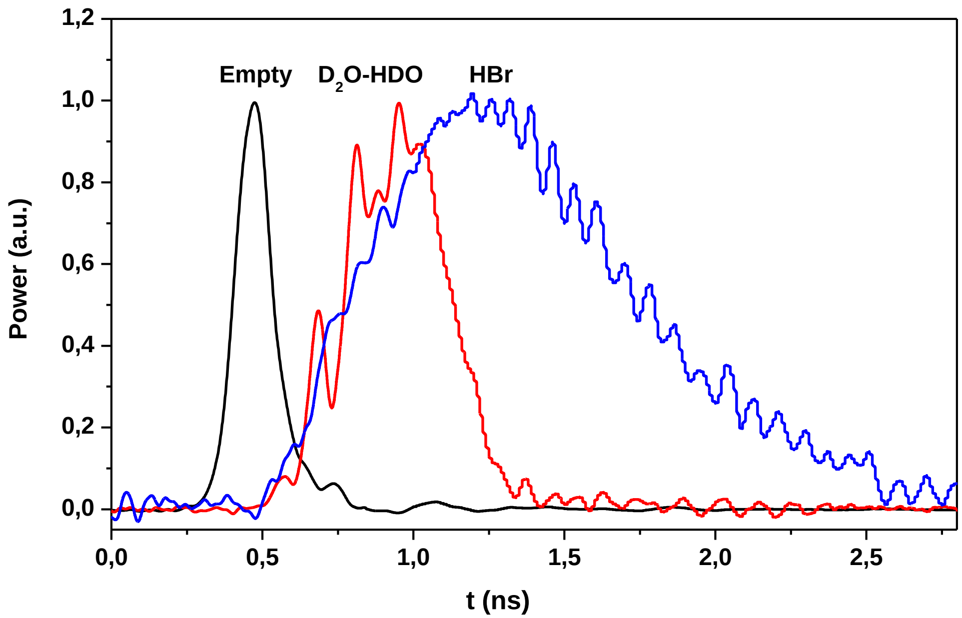


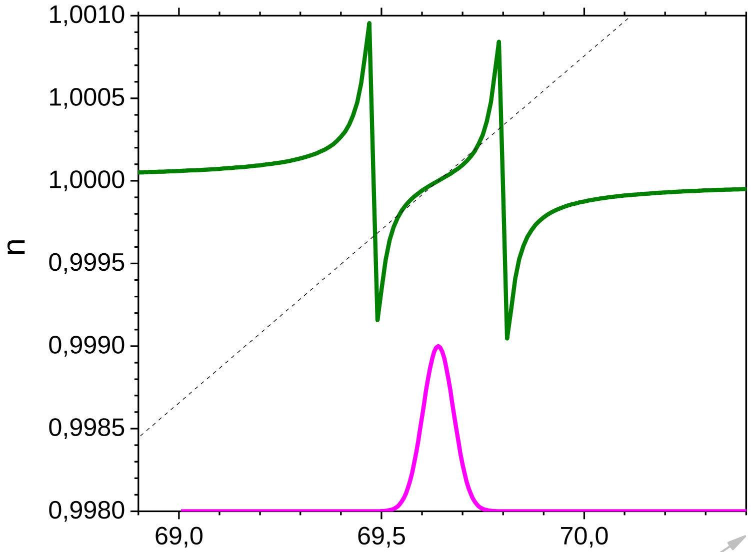




Chesnokov E.N., Kubarev V.V., and Koshlyakov P.V.
"*Rotation commensurate echo of asymmetric molecules - Molecular fingerprints in the time domain*", .Applied Physics Letters 105 (2014) 261107-(1-4).

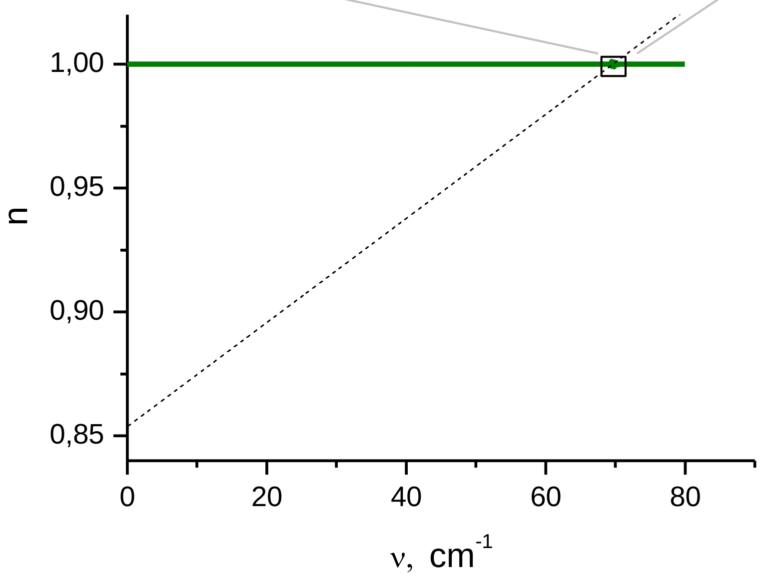
Giant light speed reduction in high-dispersion gas medium





Group light speed:

$$V_g = \frac{d\omega}{dk} = \frac{c}{n + \omega \dfrac{dn}{d\omega}} = (0.79 - 0.87)c$$



L.V. Hau, S. E. Harris, Z. Dutton, and C.H. Behroozi, "*Light speed reduction to 17metres per second in an ultracold atomic gas*", *Nature*, v. 397, pp. 594-598, 1998.

Sample: T = 450 nK, L = 229 μm

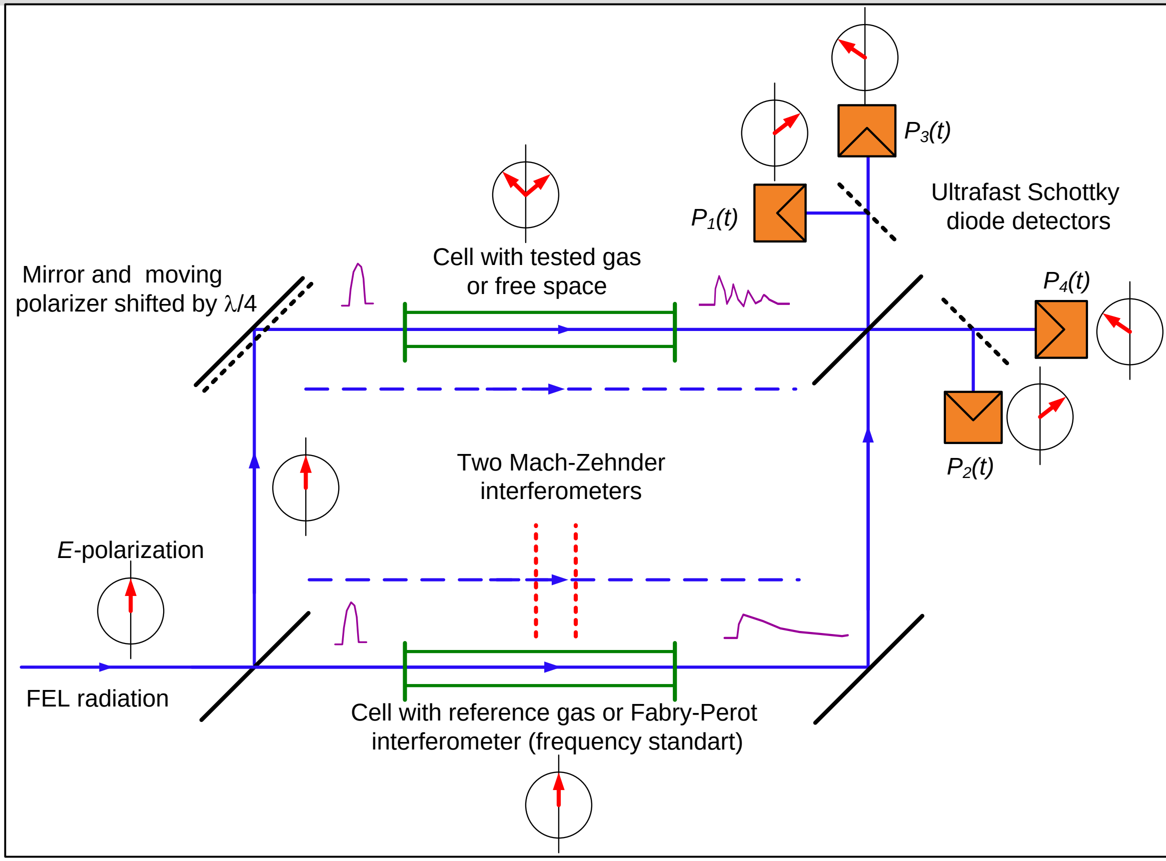
Scheme of the ultrafast time-domain spectrometer

$$E(t,\varphi_0) = \frac{1}{2\pi}\int_{\omega} E(\omega)e^{i(\omega t+\varphi(\omega)+\varphi_0)}d\omega$$

$\mathrm{Re}(E(t,\varphi_0))$ $\mathrm{Im}(E(t,\varphi_0))$

$\mathrm{Im}(E(t,0)) = \mathrm{Re}(E(t,\pi/2))$

$\mathrm{Re}(E(t,0))$ $\mathrm{Re}(E(t,\pi/2))$



Four-channel scheme:

$$E_x(t) = E(t)\cos\varphi(t) \sim \frac{P_1^{(0)}(t) - P_2^{(\pi)}(t)}{\sqrt{P_{ref}(t)}};$$

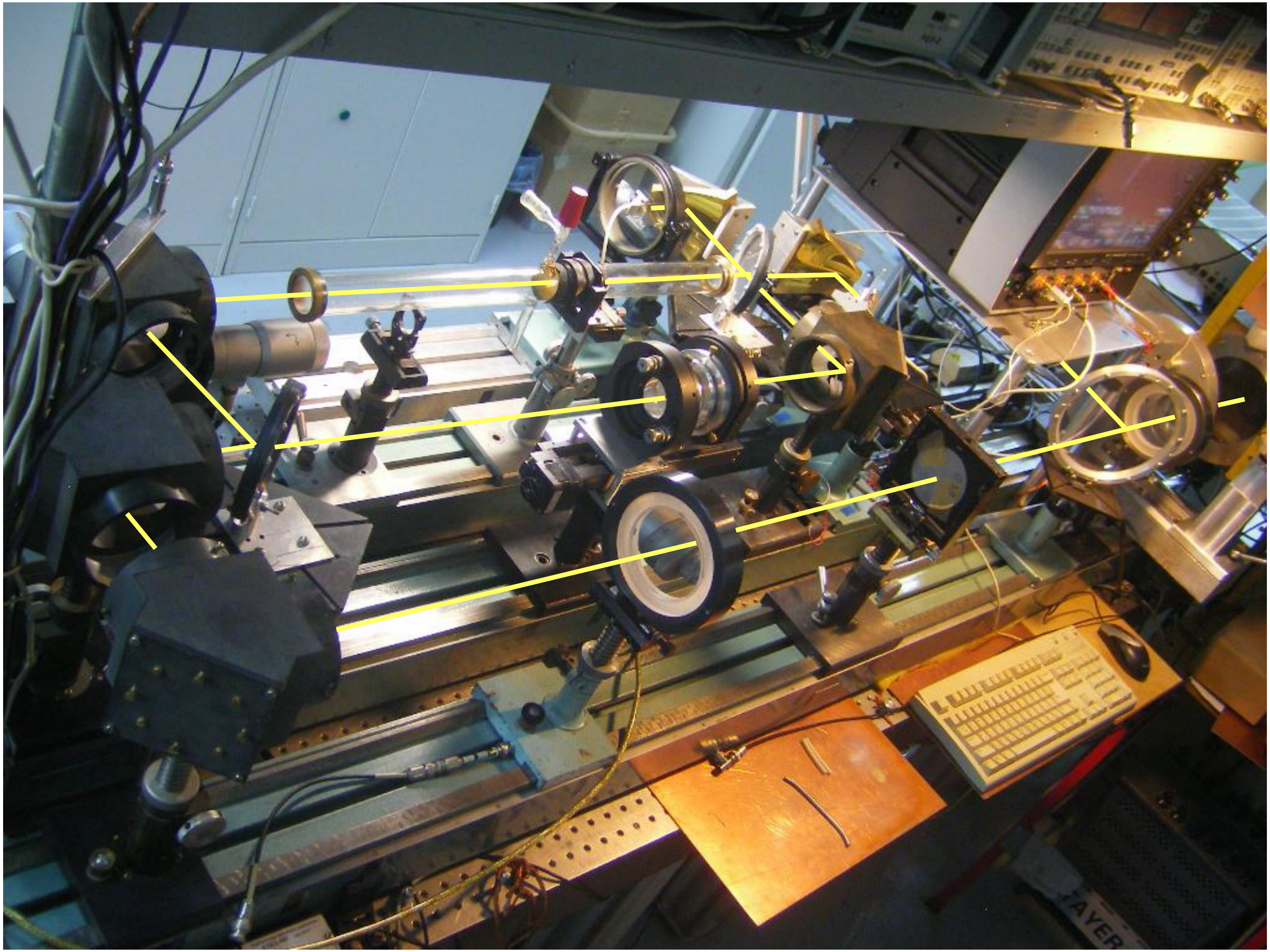
$$E_y(t) = E(t)\sin\varphi(t) \sim \frac{P_3^{(\pi/2)}(t) - P_4^{(3\pi/4)}(t)}{\sqrt{P_{ref}(t)}}.$$

Two-channel scheme:

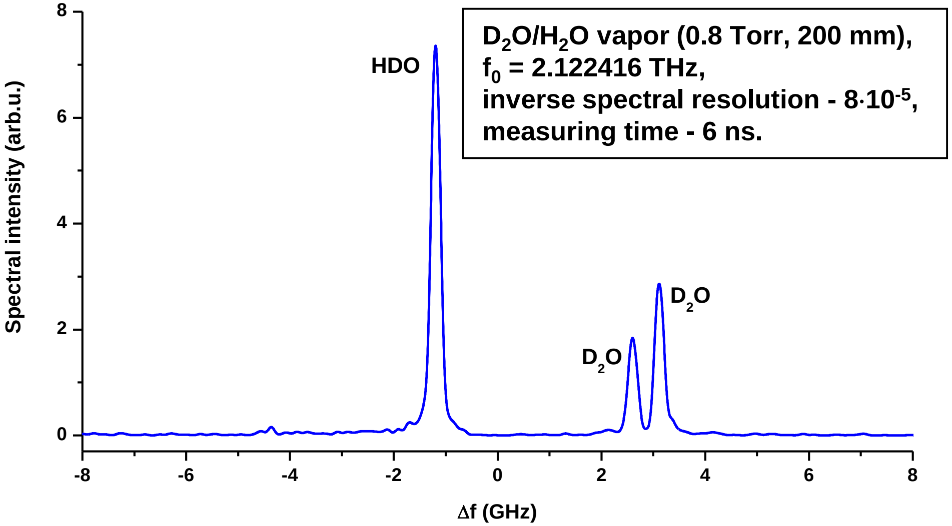
$$E_x(t) = E(t)\cos\varphi(t) \sim \sqrt{P_{1,2}(t)} - \sqrt{P_{ref}(t)};$$

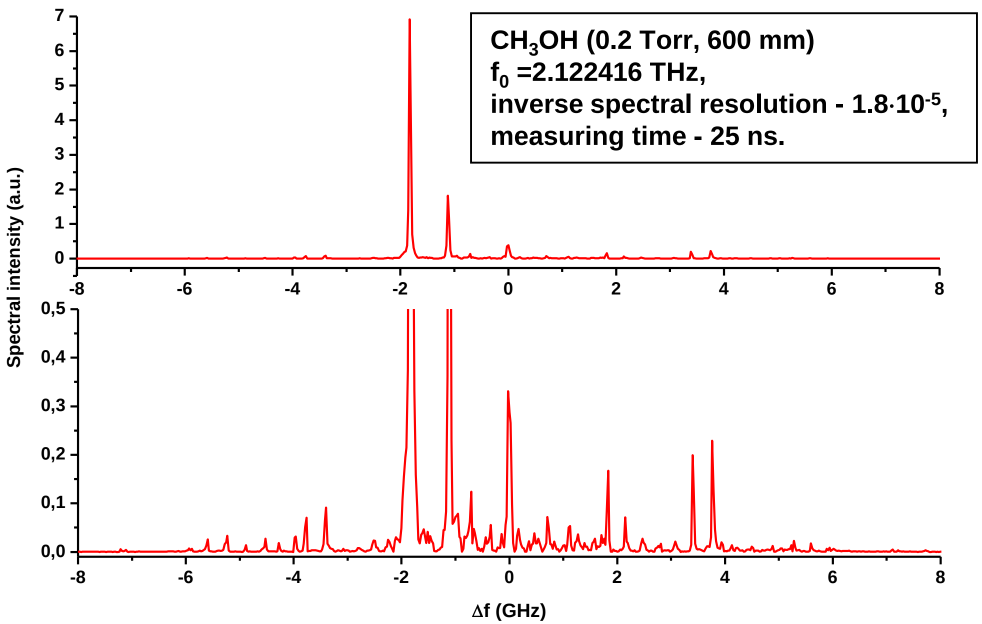
$$E_y(t) = E(t)\sin\varphi(t) \sim \sqrt{P_{3,4}(t)} - \sqrt{P_{ref}(t)}.$$

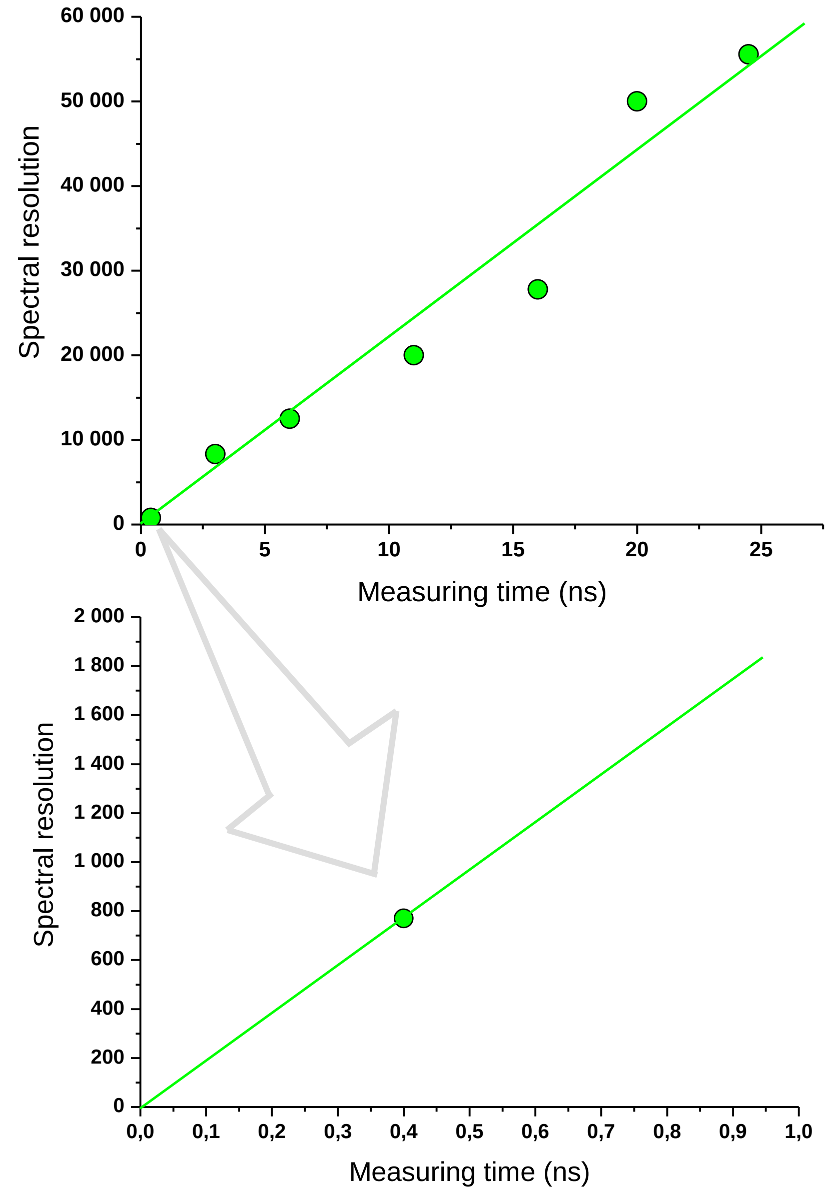
Ultrafast time-domain spectrometer (UTDS)



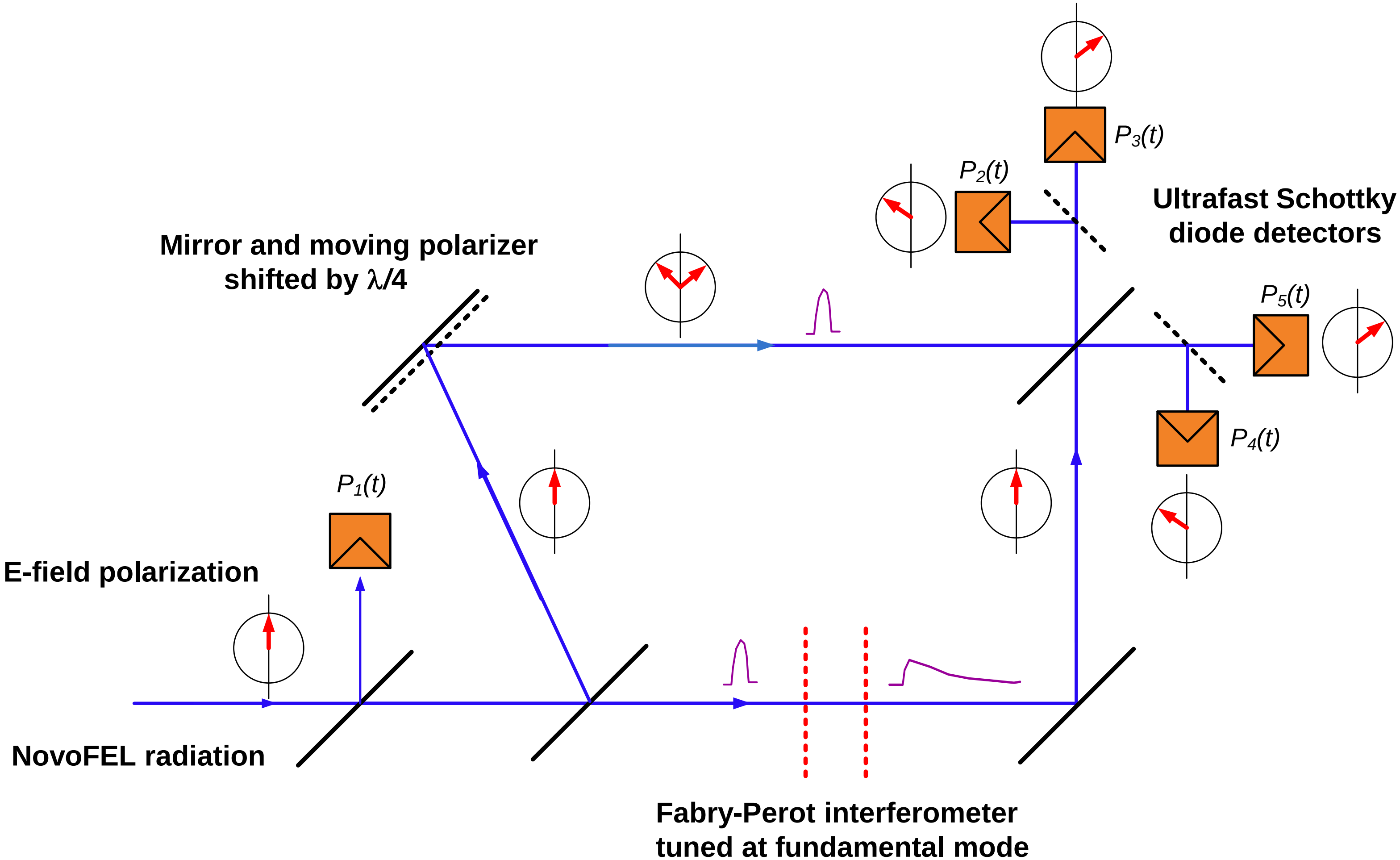
Experimental spectra and spectral resolution







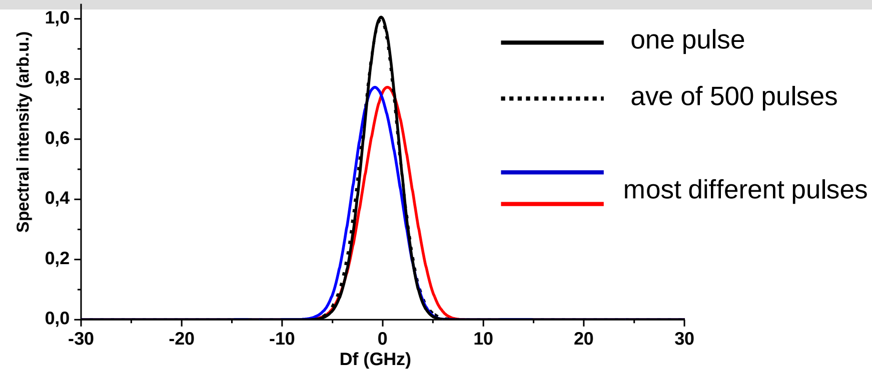
Modification of the ultrafast time-domain spectrometer for one-pulse diagnostics of NovoFEL radiation



Single-pulse spectroscopy of NovoFEL radiation

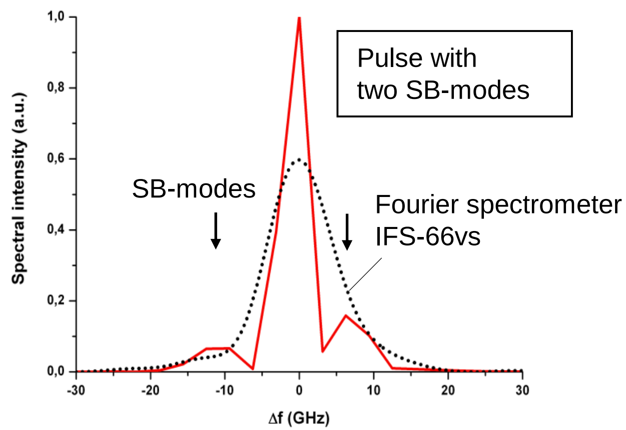
Stabilized regime

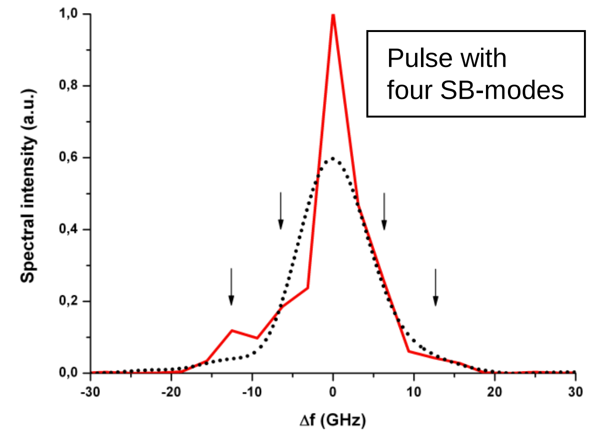
Spectral width (FWHM) – $2\cdot10^{-3}$ (4 GHz) – Fourier limit for 100 ps pulse
f_0 = 2.12 THz,
Inverse spectral resolution – $1.2\cdot10^{-3}$,
Measuring time – 0.4 ns.



Regime of moderate side-band (SB) instability

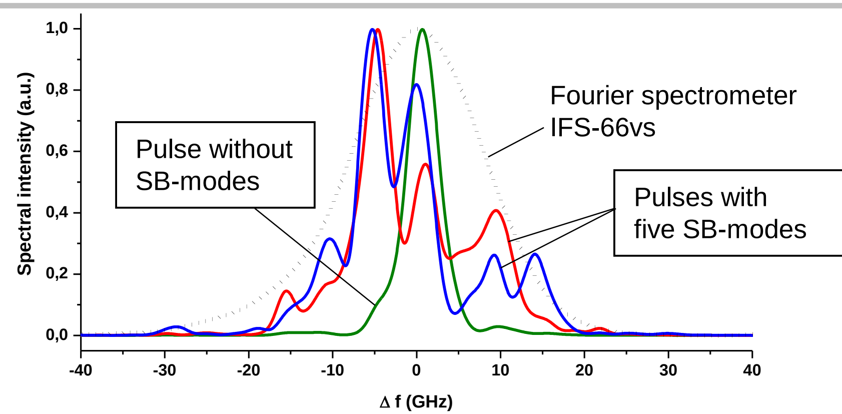
Integral spectral width – $5\cdot10^{-3}$ (10 GHz)
f_0 = 2.12 THz,
Inverse spectral resolution – $1.2\cdot10^{-3}$,
Measuring time – 0.4 ns.





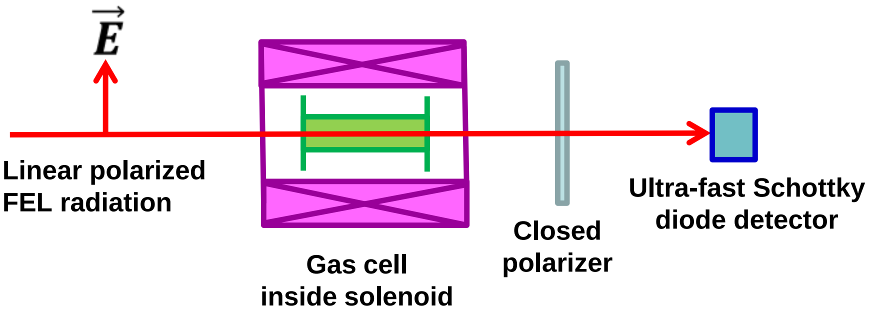
Regime of strong SB-instability

Integral spectral width – $9\cdot10^{-3}$ (18 GHz)
f_0 = 2.12 THz,
Inverse spectral resolution – $1.2\cdot10^{-3}$,
Measuring time – 0.4 ns.

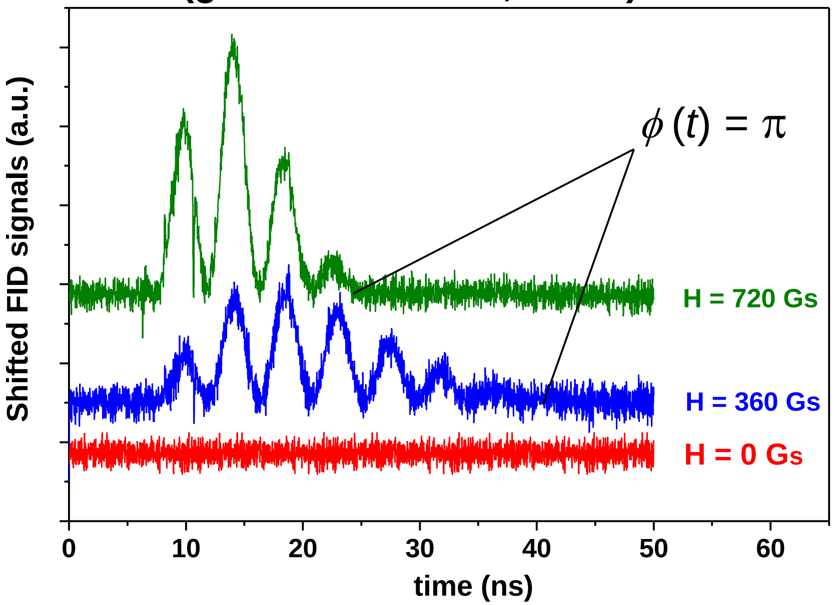


Non-Faraday rotation of polarization

Scheme of the experiment:

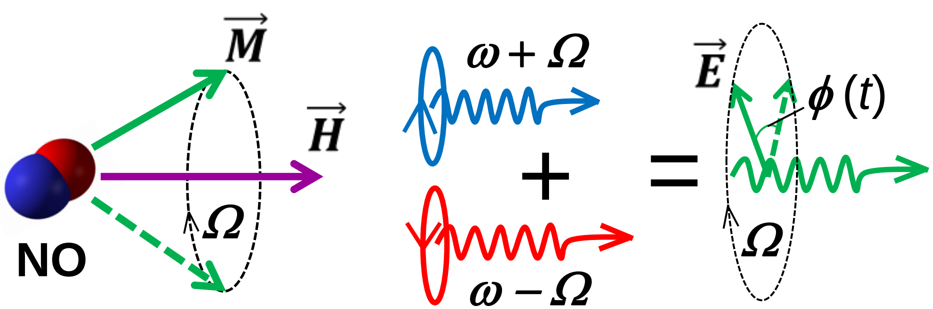


FID radiation of paramagnetic NO molecules (gas cell - 400 mm, 2 Torr) :



Features of the non-Faraday rotation of coherent FID radiation :

- large-scale effect
- rotation angle is time function

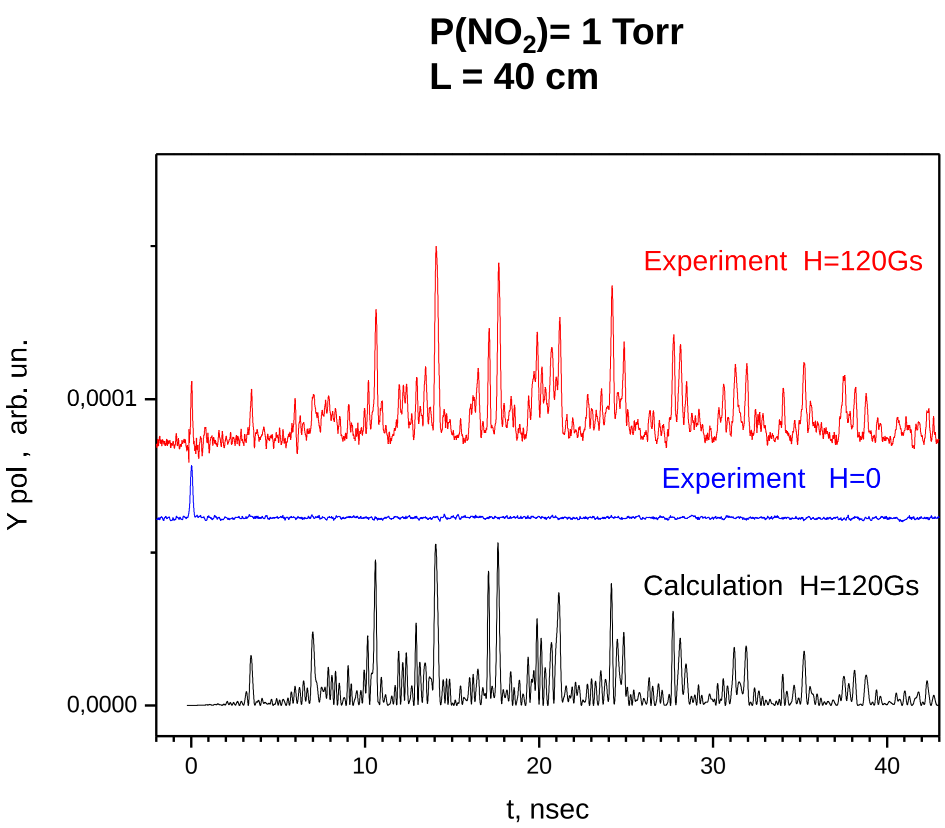


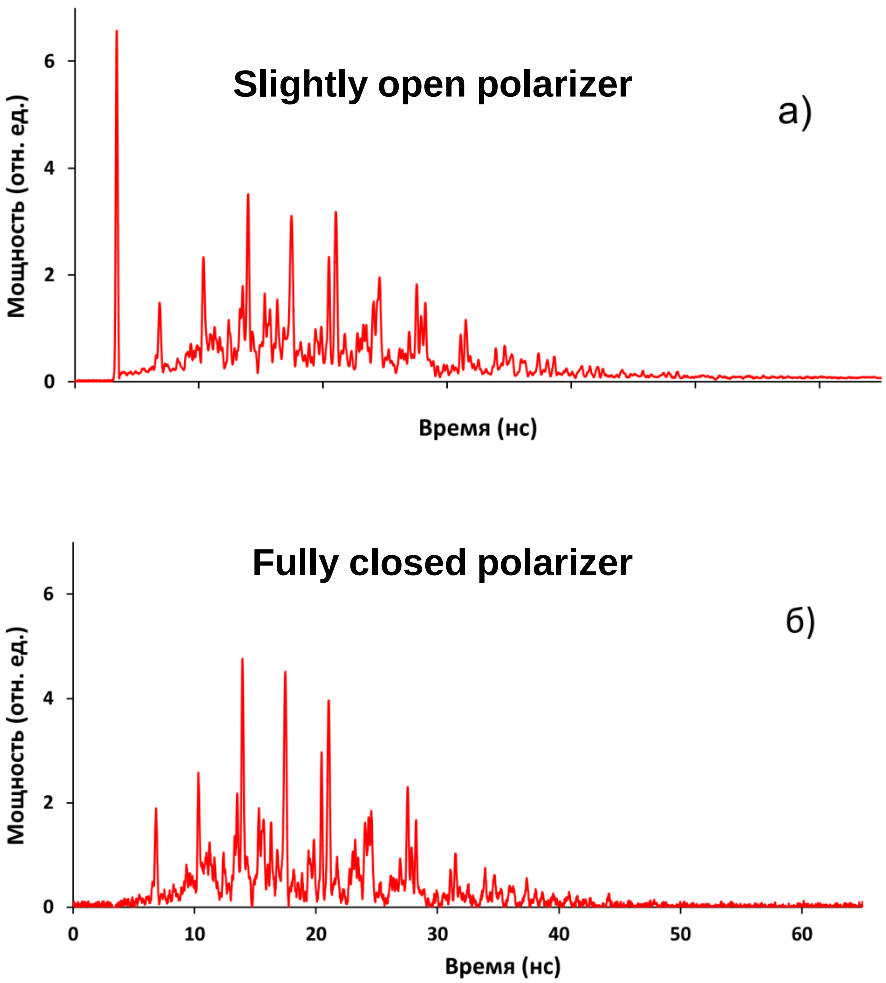
Applications:

- **high resolution molecular spectroscopy (H = 30-70 kGs, superconducting solenoid)**
- **ultrafast high-sensitive spectroscopy of short-lived chemical radicals**

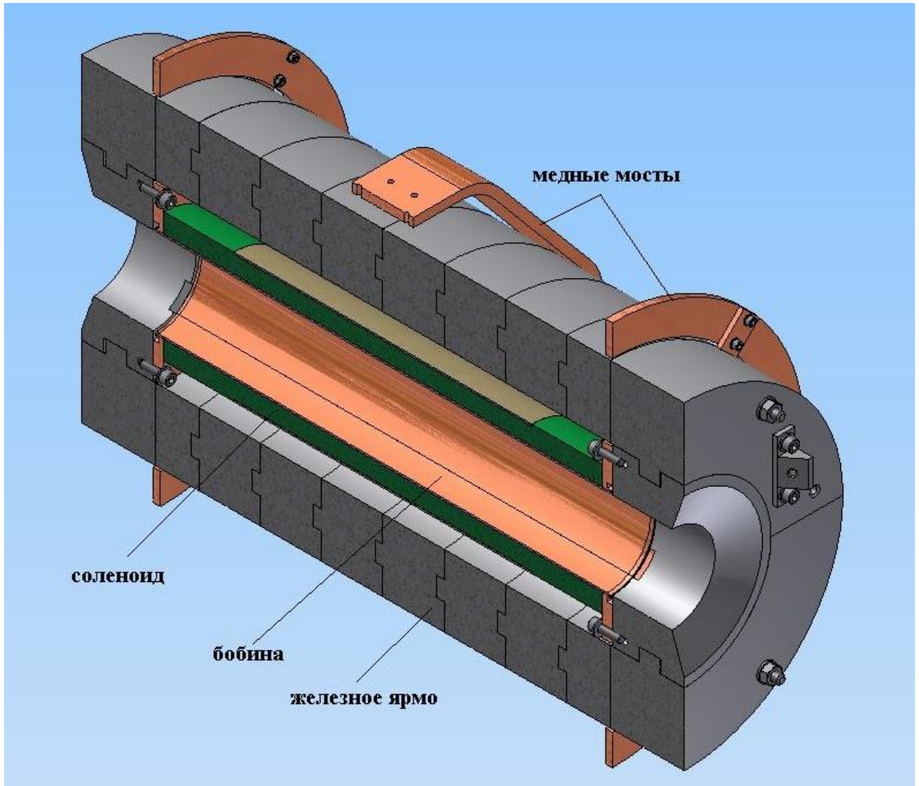
Chesnokov E.N., Kubarev V.V., Koshlyakov P.V., Getmanov Ya.V., Shevchenko O.A.
"*Non-Faraday rotation of the free induction decay in gaseous NO*", Chemical Physics Letters 636 (2015).

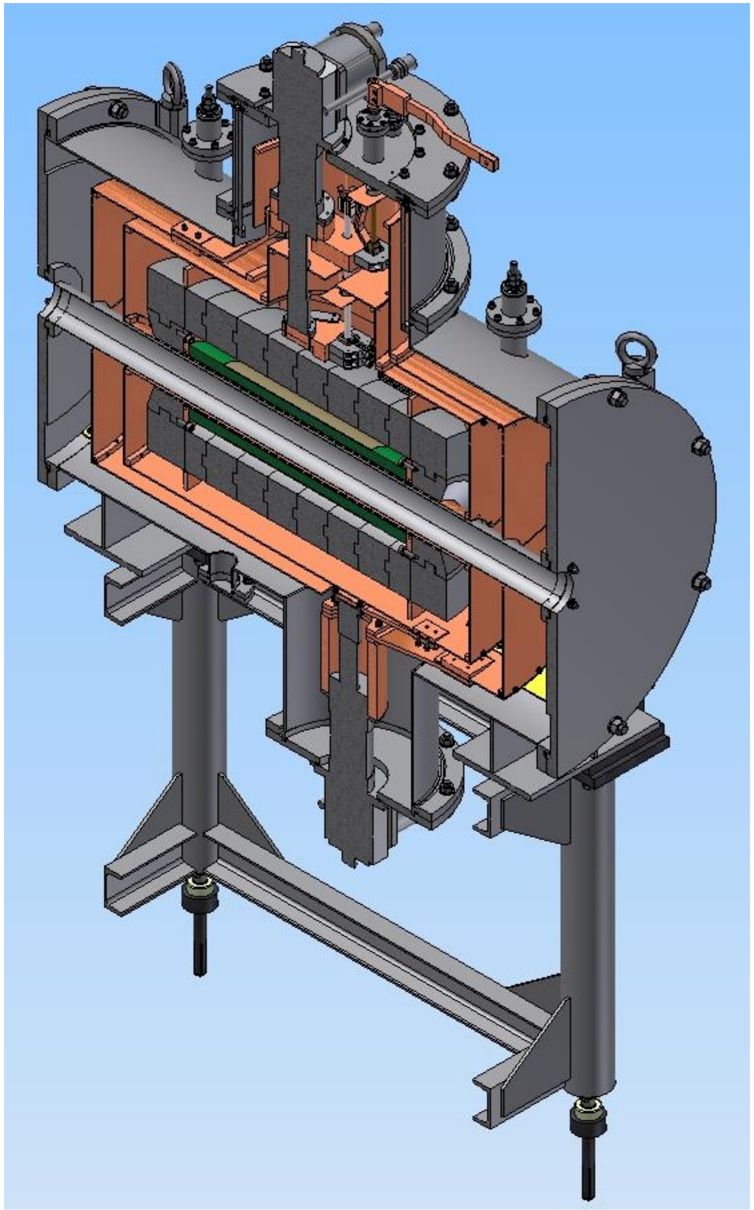
NO_2 FID in magnetic fields. Sensitive spectroscopy





Ultrafast spectroscopy in strong magnetic field





Liquid-free superconducting 6 T solenoid

Poster Session:
"Superconducting solenoid for superfast THz spectroscopy"
A. Bragin, S. Khruschev, V. Kubarev, N. Mezentsev, V. Shkaruba, G. Sozinov, V. Tsukanov

Thank you for attention !