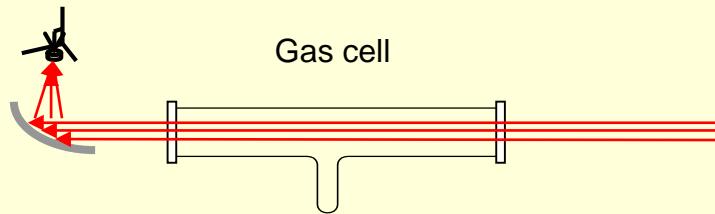


# Zero-area THz optical pulses in gases.

Evgeniy Chesnokov (ICKC SBRAS, Novosibirsk)  
Vitaly Kubarev (BINP, Novosibirsk)  
Pavel Koshlyakov (ICKC SBRAS, Novosibirsk)

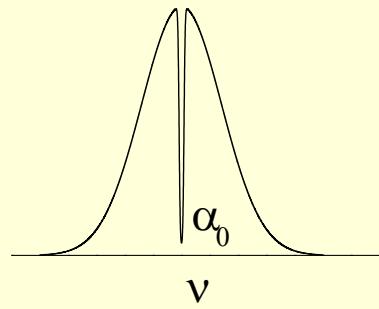
# Unusual transformation of the laser pulse.

Fast detector

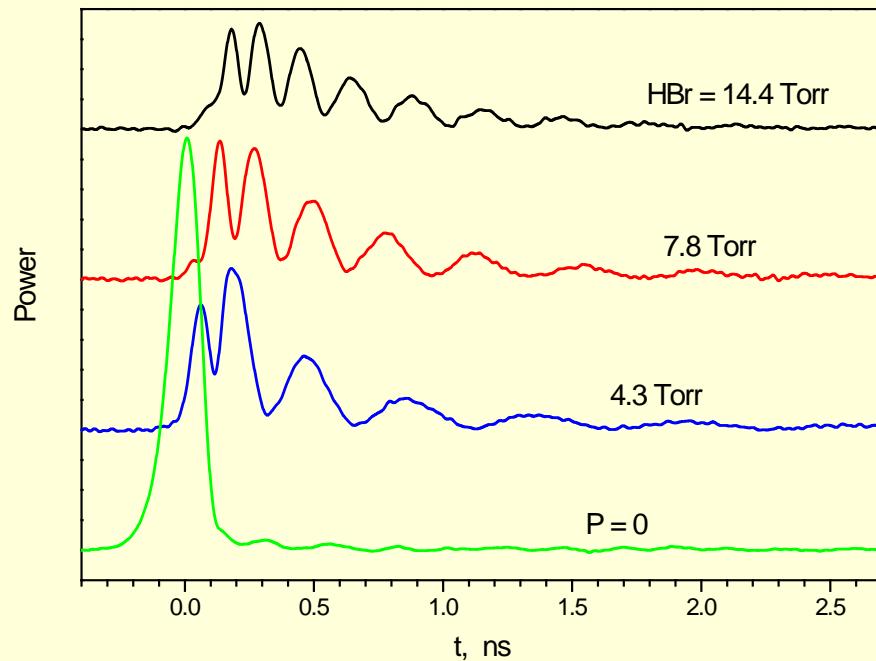


5.6 MHz  
5 – 20  $\mu$ J  
120 ps

Terahertz FEL pulses.



Narrow absorption line.  
High optical density.



# Area of the optical pulse.

$$E(z, t) = A(z, t) \cdot e^{-i(\omega_0 t - kz)}$$

Amplitude (slow)

Carrier frequency (fast)

Area

$$\theta(z) = \frac{d_{12}}{h} \int A(z, t) dt$$

In absorption media:

$$\theta \rightarrow 0;$$

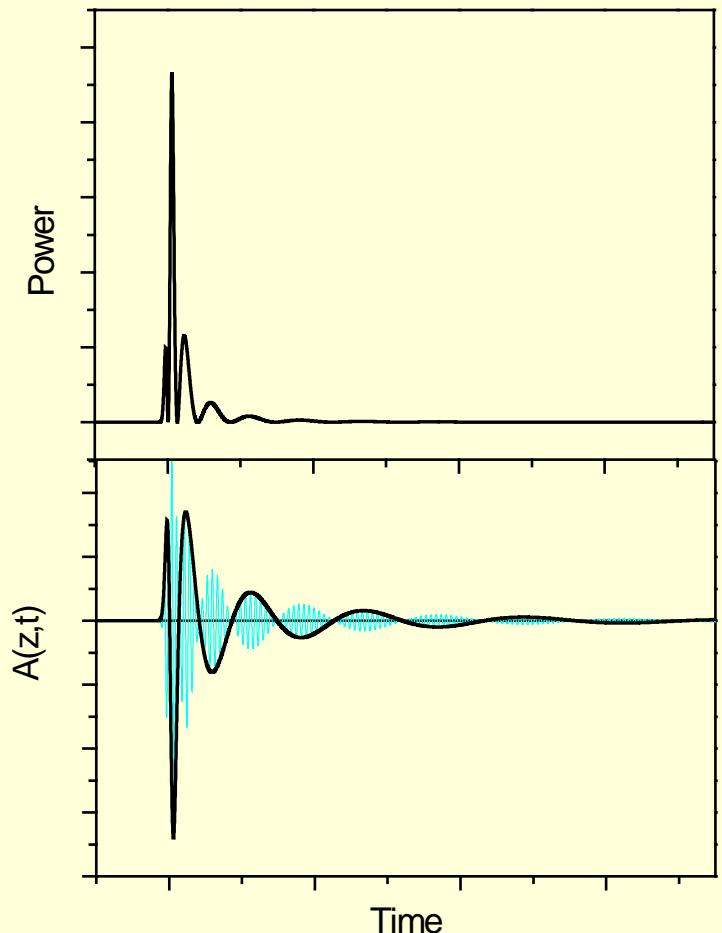
$$\theta \rightarrow 2\pi;$$

$$\theta \rightarrow 4\pi;$$

.....

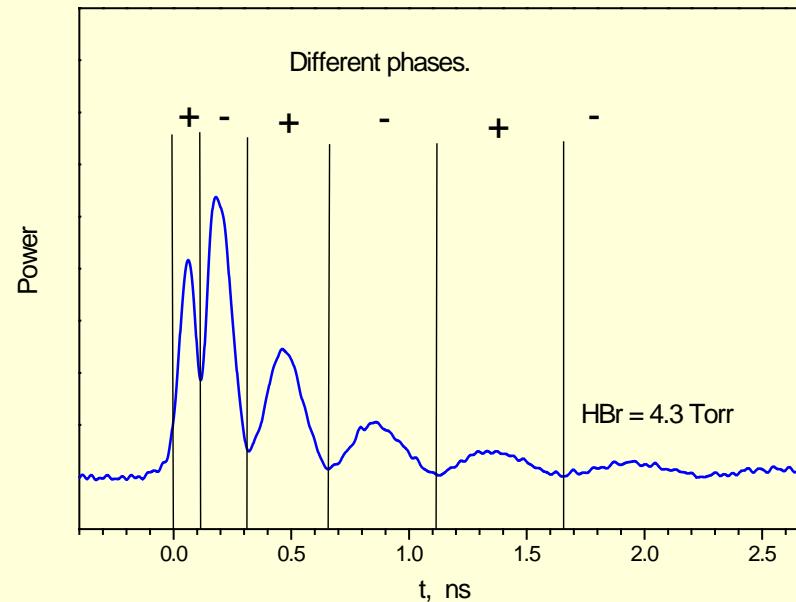
Beer -Lambert law.

$$\theta(z) = \theta_0 \exp(-\alpha_0 z)$$



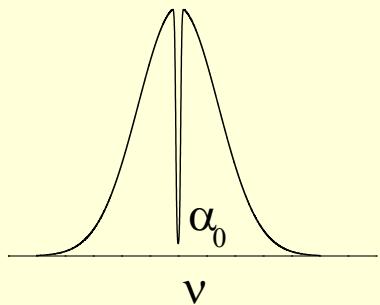
Our experimental signal.

$$\theta = 0.$$



1. How the zero-area optical pulse is forming?
2. How it transformed by passing through an absorbing medium?

# Exact analytical solution. Grisp 1972.



Assumptions.

1. Small area  $\theta \ll 1$
2. Single Lorentz absorption line.
3. Input: short optical pulse:  $E(z,t) = \delta(t-kz)$

$$n(\omega) = 1 + \frac{\alpha_0 c}{\pi \gamma \omega_0} \cdot \frac{i}{1 + 2i(\omega - \omega_0)/\gamma}$$

$$\frac{E(t)}{E_{empty}(t)} = -\sqrt{\frac{\chi}{t}} \cdot J_1(2\sqrt{\chi t}) \cdot \exp\left(-t \frac{\gamma}{2}\right)$$

$$\boxed{\frac{I(t)}{I_{empty}(t)} = \frac{\chi}{t} \cdot J_1^2(2\sqrt{\chi t}) \cdot \exp(-\gamma t)}$$

Tail of the pulse - Free Induction Decay. (FID)

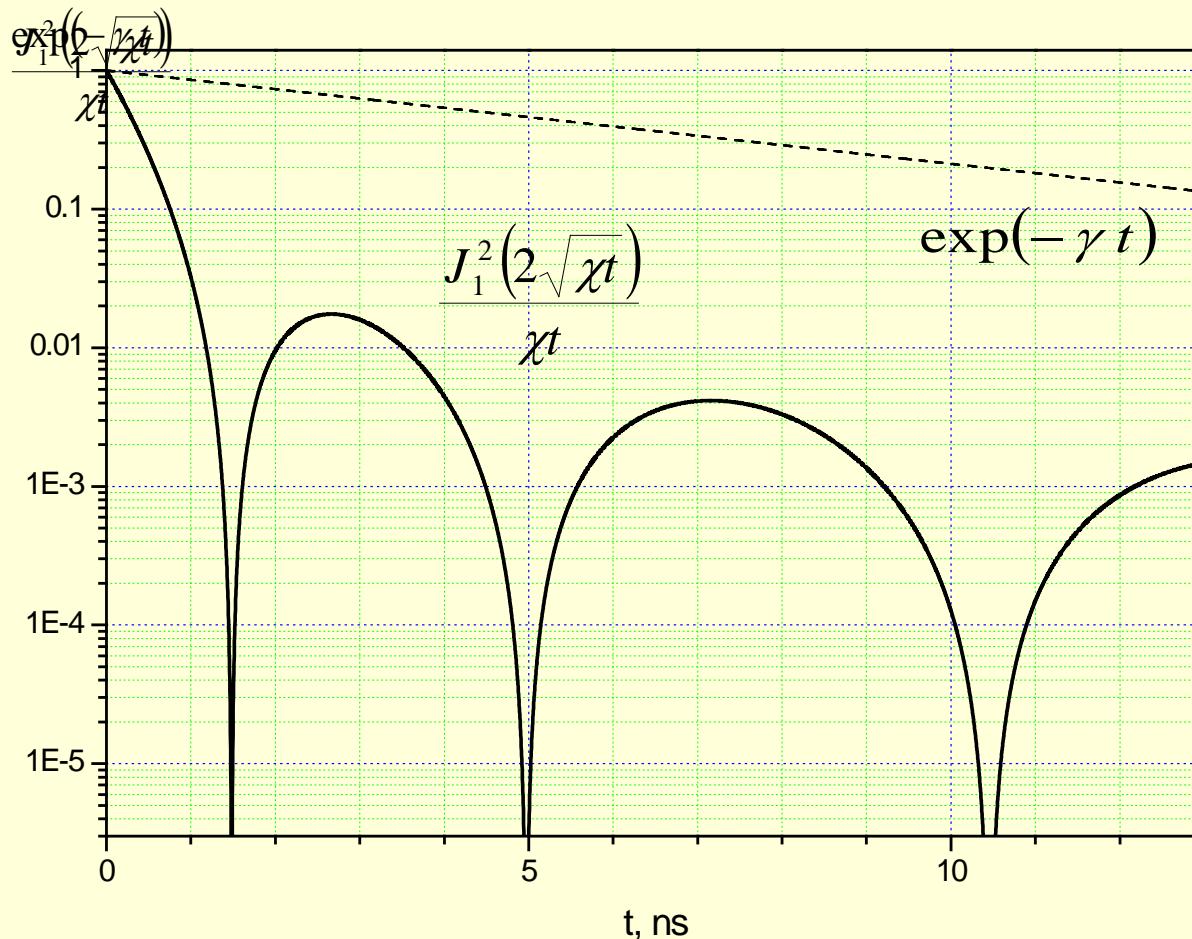
Two parameters:  $\gamma$  - linewidth,

$$\chi = \frac{L}{\pi} \int \alpha(\omega) d\omega \quad \text{-line integrated intensity}$$

# Formation of the zero-area pulses.

FID at high optical density. Two factors.

$$I(t) = \frac{\chi}{t} \cdot J_1^2(2\sqrt{\chi t}) \cdot \exp(-\gamma t) \quad \chi = l\alpha_0 / \pi$$

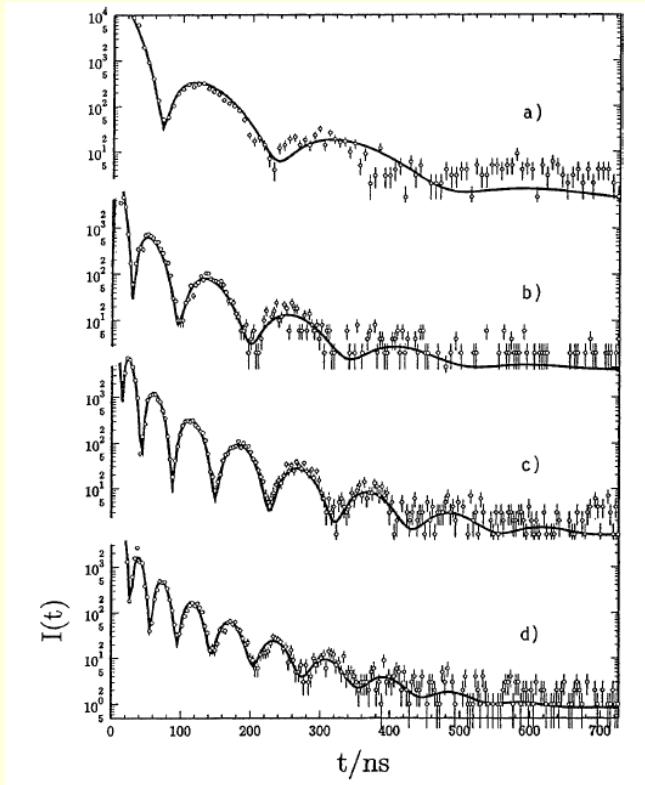


Values of the rates:  
 $\gamma = 0.155 \cdot 10^9 \text{ sec}^{-1}$   
 $\chi = 9.9 \cdot 10^9 \text{ sec}^{-1}$

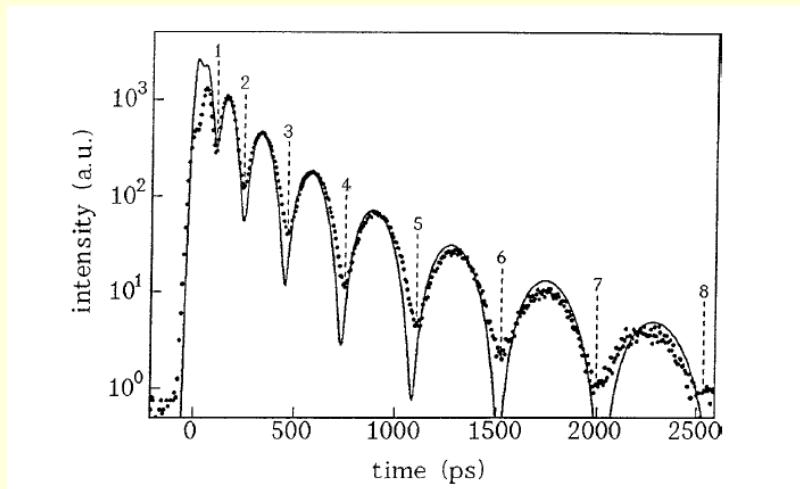
$$\begin{aligned} \chi/\gamma &= \\ &= \alpha(\omega_0)l/2 \end{aligned}$$

# The universality of the phenomenon.

Mossbauer spectroscopy,  $^{57}\text{Fe}$  the 122 keV



Exciton resonance in  $\text{Cu}_2\text{O}$  at 2eV



D. Fröhlich, A. Kulik, B. Uebbing, A. Mysyrowicz, V. Langer, H. Stolz and W. von der Osten, Phys. Rev. Lett. 67 (1991) 2343.

Y.V. Shvyd'ko, U. van Buerck, W. Potzel, P. Schindelmann, E. Gerdau, O. Leupold, J. Metge, H.D. Ruter and G.V. Smirnov, Phys. Rev. B 57 (1998) 3552.

How this formula works for molecular rotation spectra in terahertz?

# Experiments in terahertz region. Choice of the object.

Single absorption line within FEL spectra.

Rotation spectra.

HCl - - chlorine isotops, quadrupole quadrupole splitting.

HBr - - bromine isotops, quadrupole quadrupole splitting.

HF +++ ?? May be good.

CO - low dipole moment.

HCN +++ strong widely spaced lines.

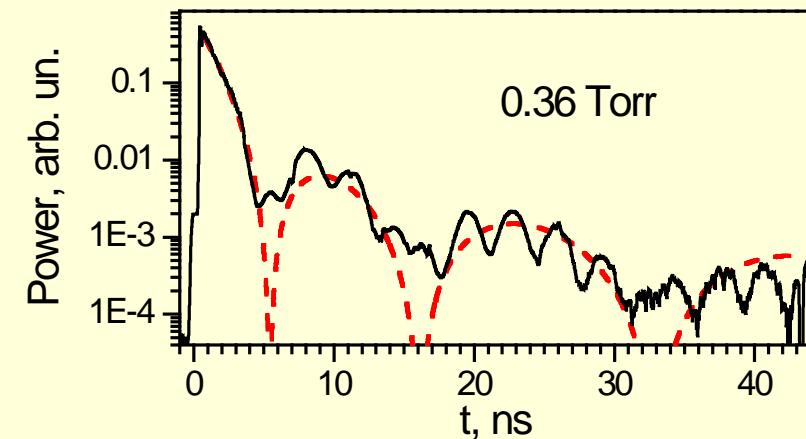
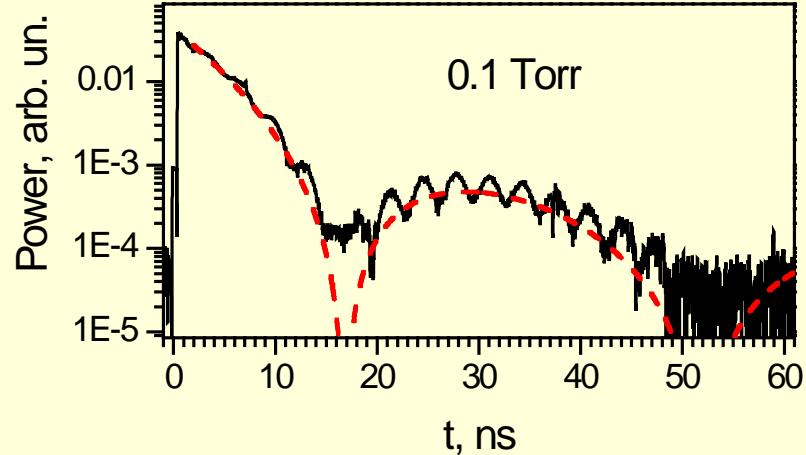
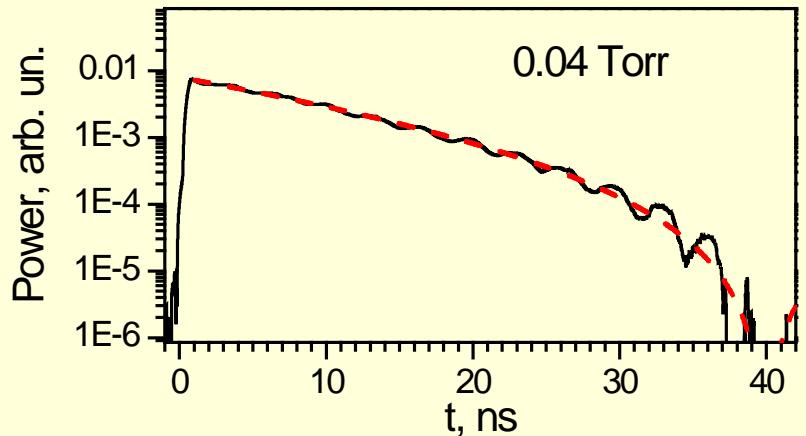
# Experiments with HCN.

Table 1. Calculated and experimental positions of the first minimum of FID signals at different pressures.

Pressure	0.04 Torr	0.1 Torr	0.36 Torr
t <sub>1</sub> experimental	39 ns	17 ns	5.5 ns
t <sub>1</sub> calculated	37 ns	15 ns	4.1 ns

$$I(t) = \frac{\chi}{t} \cdot J_1^2(2\sqrt{\chi t})$$

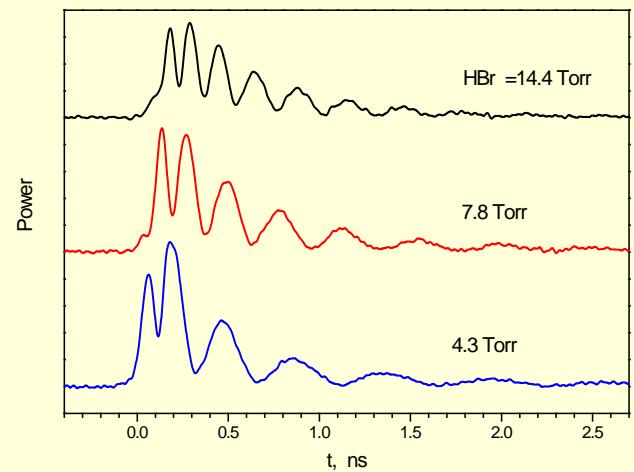
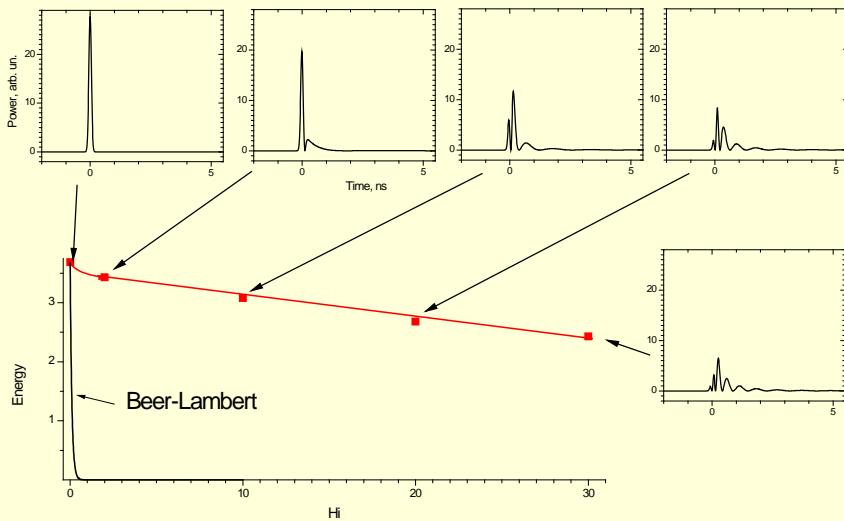
The formula is valid in Teraherts also.



# Energy dissipation

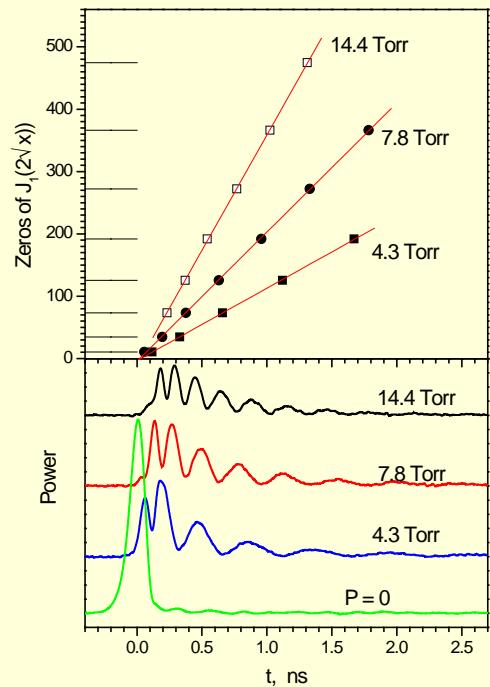
$$\left| \frac{d_{12}}{\hbar} \int A(z, t) dt \right|^2 = 0 \quad - \text{ transition probability}$$

According to perturbation theory - no energy absorption.

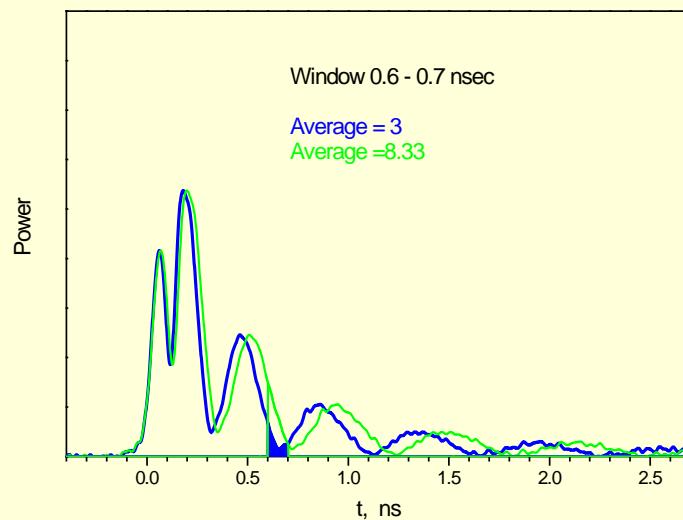


# Possible applications.

Precise measurement  
of the transition oscillator strength

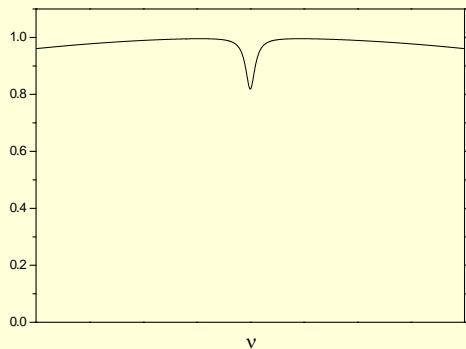


Detection of the small variation  
of concentration in time-resolved experiments.

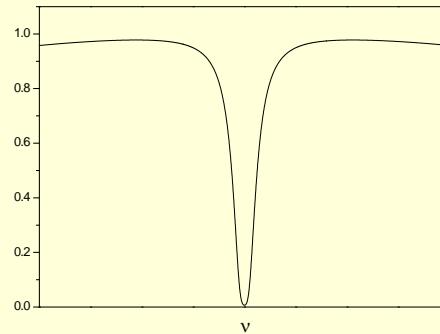


# Lifetime of the Free Induction Decay signals

Low optical density.



High optical density.



$$E_{FID}(t) \approx -\exp\left(-t \frac{\gamma}{2}\right)$$

$$E_{FID}(t) \approx -\frac{J_1(2\sqrt{\chi t})}{\sqrt{\chi t}}$$

Lifetime might be shorter.

The narrow absorption lines and a large optical density are typical for pulse propagation in the atmosphere.

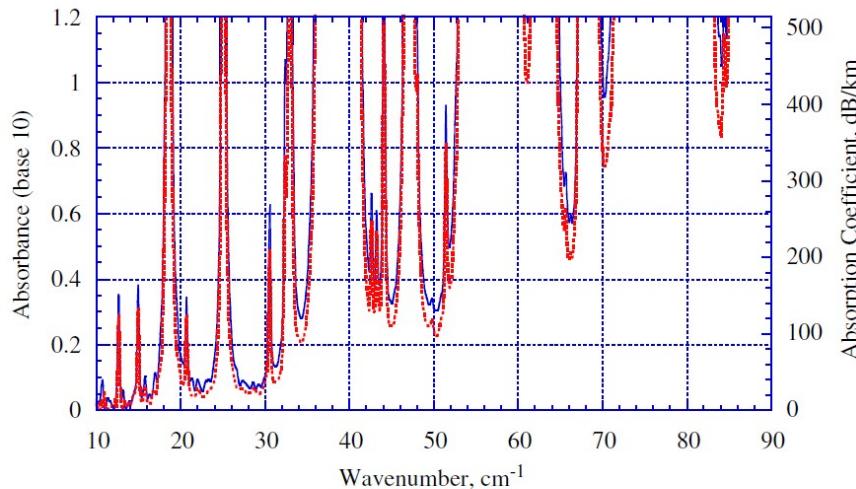


Fig. 3. Absorption spectra of a mixture composed of water vapor at 0.67 kPa and nitrogen at 70 kPa. Top and bottom spectra were obtained at 293 and 333 K, respectively.

Path length = 23 m

Thank you for attention.