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EXPANDING THE CAPABILITIES OF CO_2 LIDAR SYSTEM WITH NONLINEAR OPTICAL CHALCOGENIDE CRYSTALS

Wojciech Kuźnik, Iwan Kityk

WG Member - wojtek.kuznik@gmail.com

Czestochowa University of Technology, Poland

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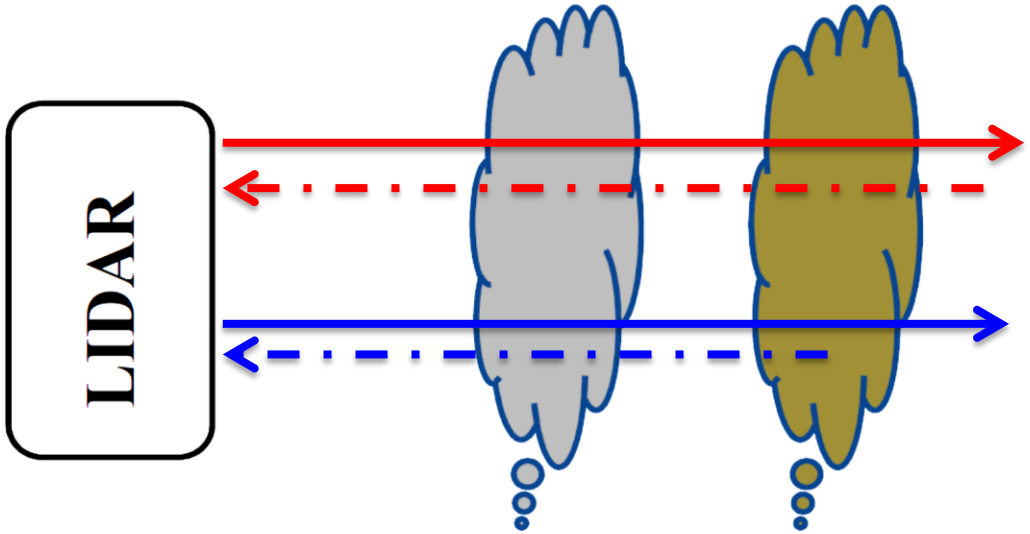
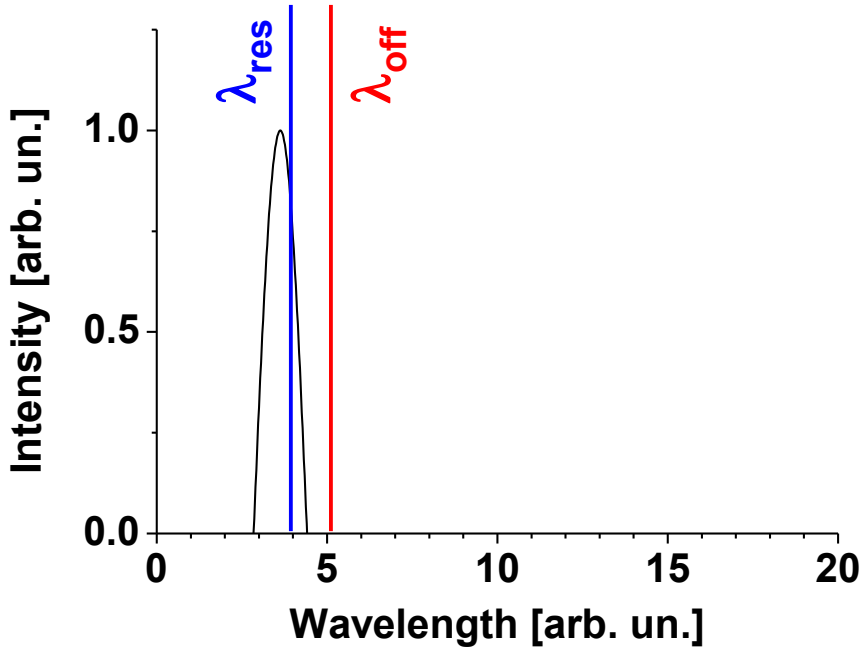
Presentations overview

- Short Description of Differential Absorption LIDAR (DIAL) system
- DIAL limitations
- Nonlinear Optics (NLO) overview
- Application of NLO crystals to overcome the DIAL limitations
- Why chalcogenide crystals?
- Results
- Challenges & Research Plans
- Conclusions

DIAL system

- Two wavelengths are used: one absorbed by the monitored gas λ_{res}, second off resonance λ_{off} used as reference
- Reflection of both wavelengths is measured



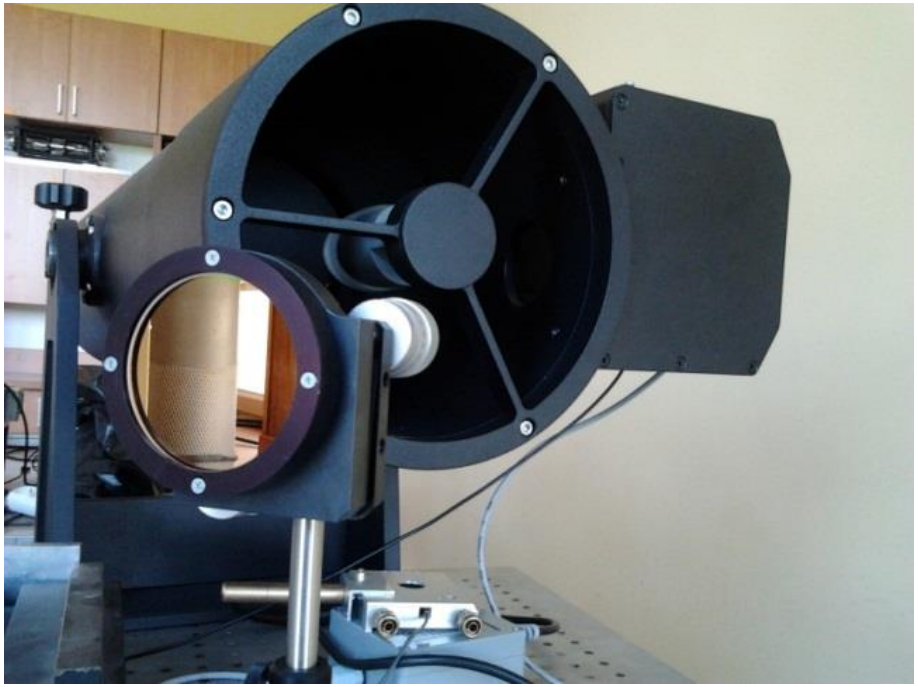


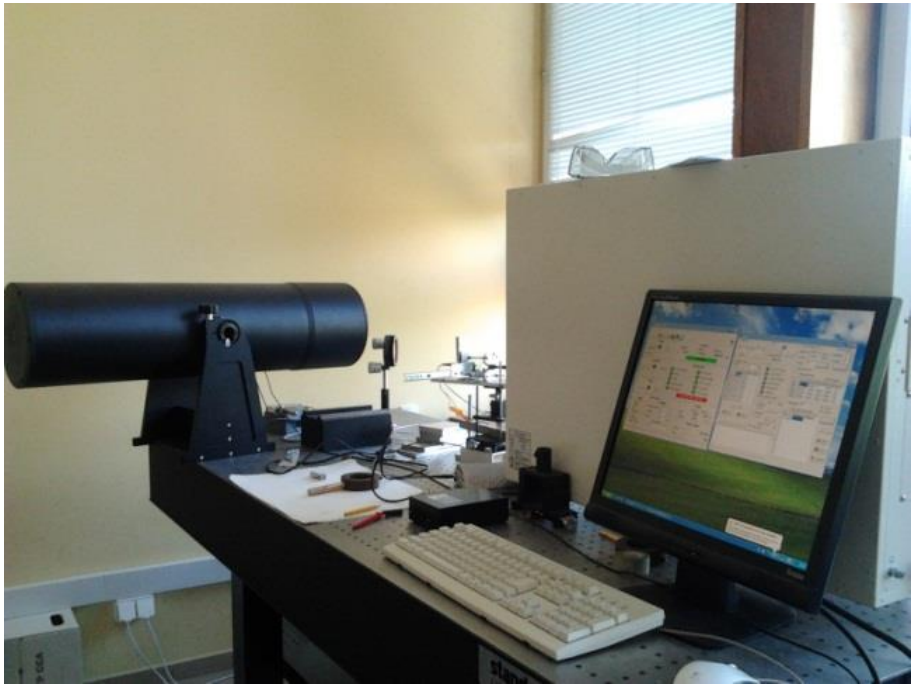
Range up to 15 km (our set-up 2 km)

May be mounted on vehicles

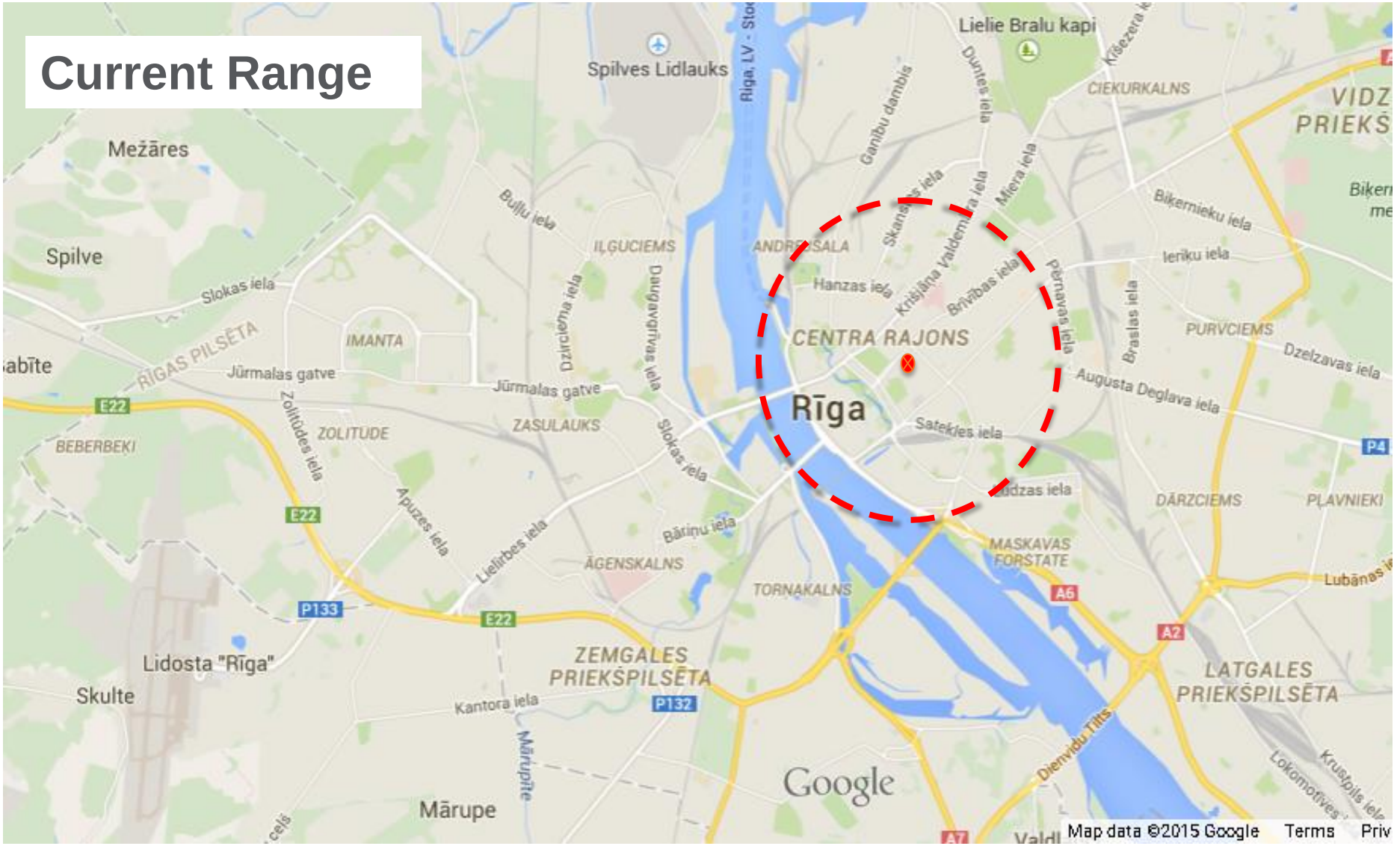
DIAL system - Czestochowa

- Laser wavelength 9 - 11.8 μm, pulse energy up to 90 mJ, pulse duration 1-4 μs,
- $TEM_{0.4}$, divergence less than 30', CdHgTe detector cooled with N_2, mirror diameter 20 cm (2 km range, may be upgraded)
- Currently: detection of CO_2 and O_3

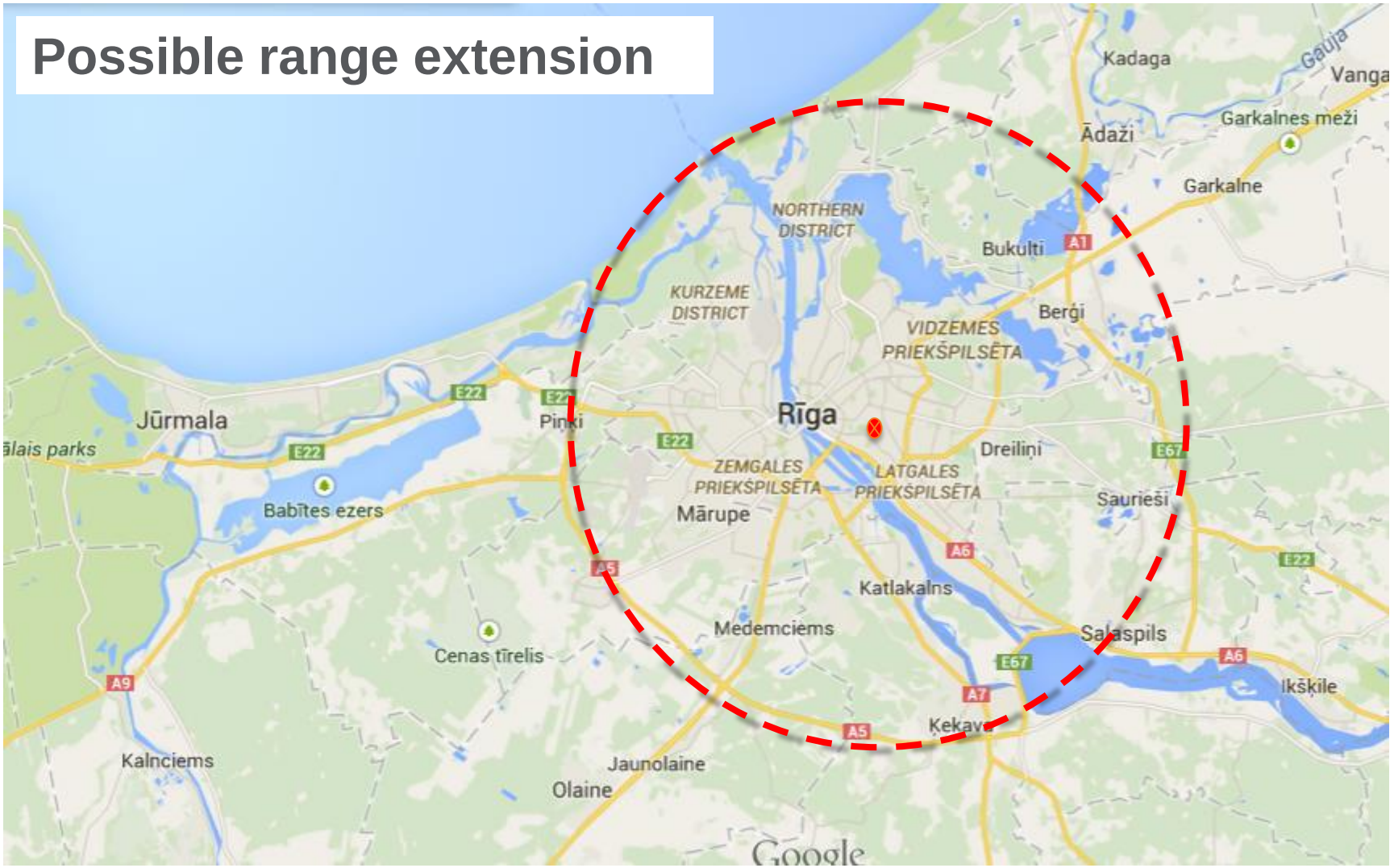




DIAL system - Czestochowa

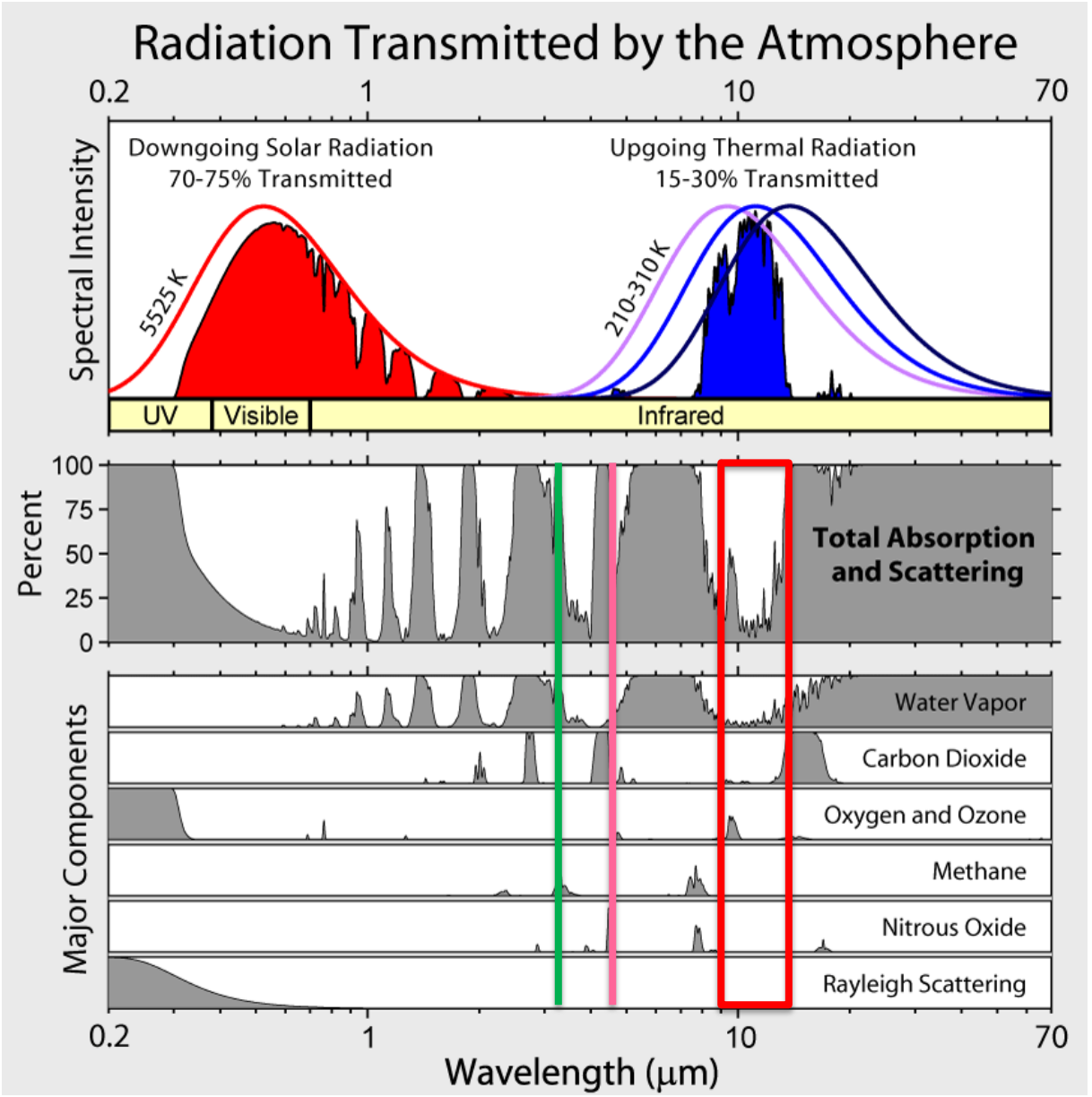


DIAL system - Czestochowa



Limitations of the DIAL CO_2

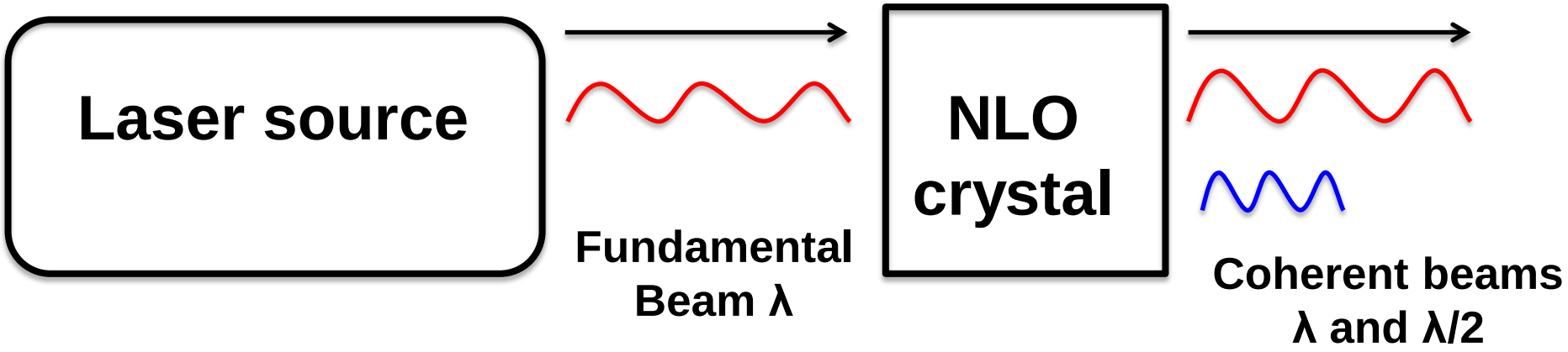
- 1 – Spectral range limited to 9-12 μm
- 2 – Absorption bands of different gases overlap



Nonlinear optics overview

- For high intensity of light (especially pulsed lasers) principle of superposition does not hold: 2 * input ≠ 2 * output -> Lambert-Beer Law does not hold
- The increase in recorded signal with light intensity may be substantially higher than in the case of linear optical phenomena
- Nonlinear Coherent Frequency Conversion – photons are combined, for example Second Harmonic Generation, Sum Frequency Generation – two photons combine to create one with higher energy
- The crystals allow for continuous wavelength tuning

Principle of the Nonlinear Frequency Mixing

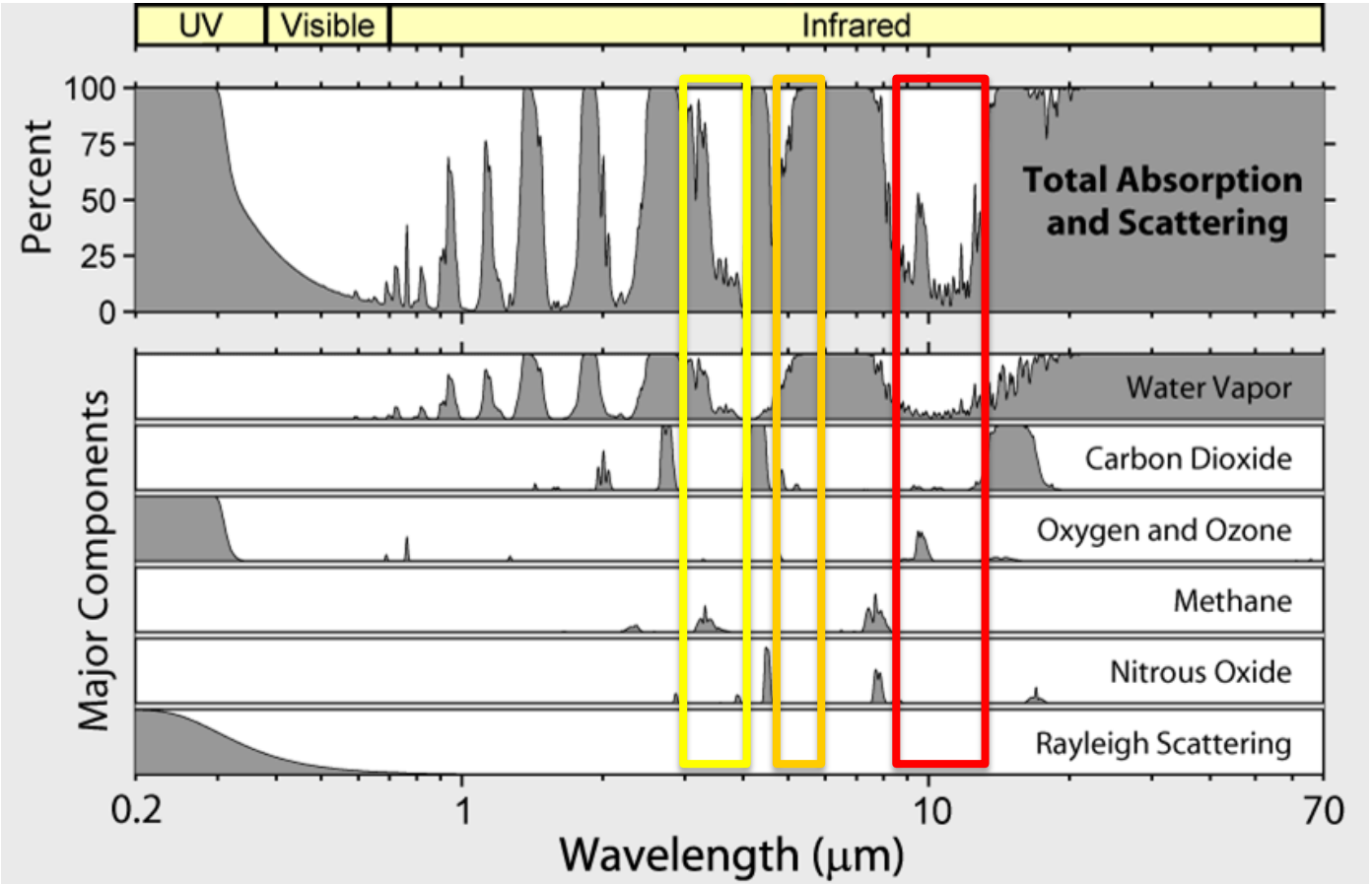


λ/2 - Second Harmonic Generation

$$\boldsymbol{P}_i(2\omega) \sim \chi_{ijk}^{2\omega} E_j(\omega)\, E_k(\omega)$$

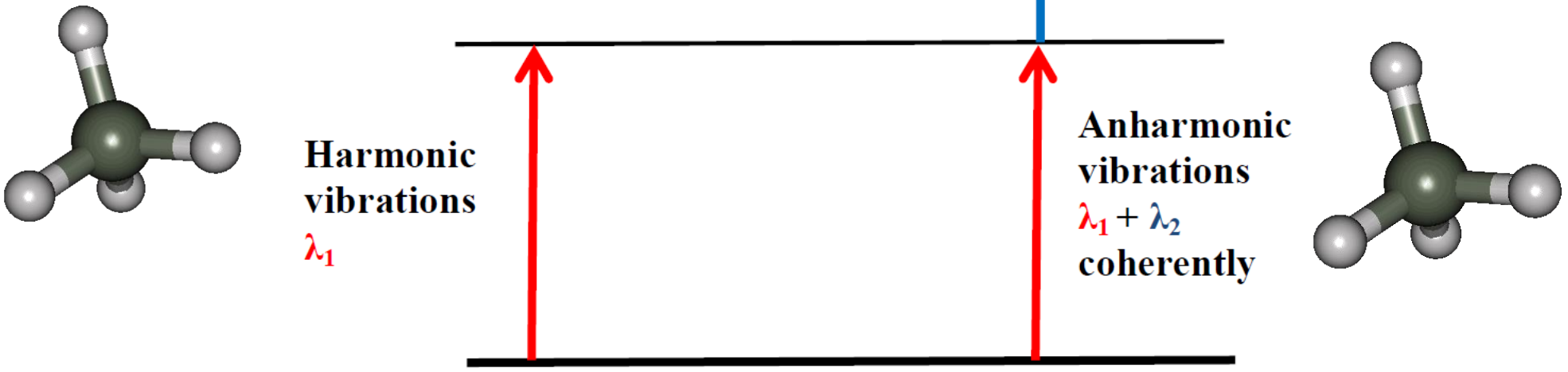
Solution to limited spectral range - SHG

- 1 – Spectral range limited to 9-12 µm
- SHG extends it to 4.5-6 µm
- THG extends it to 3-4 µm



Solution absorption overlap – coherent waves

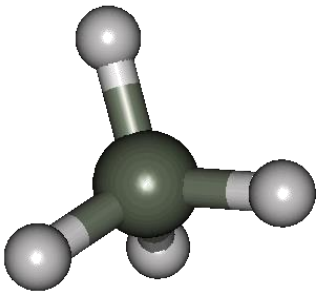
- For high intensity of light (especially pulsed lasers) principle of superposition does not hold.
- Anharmonic interaction 2 coherent waves with gases: hyper-Rayleigh scattering introduces additional information

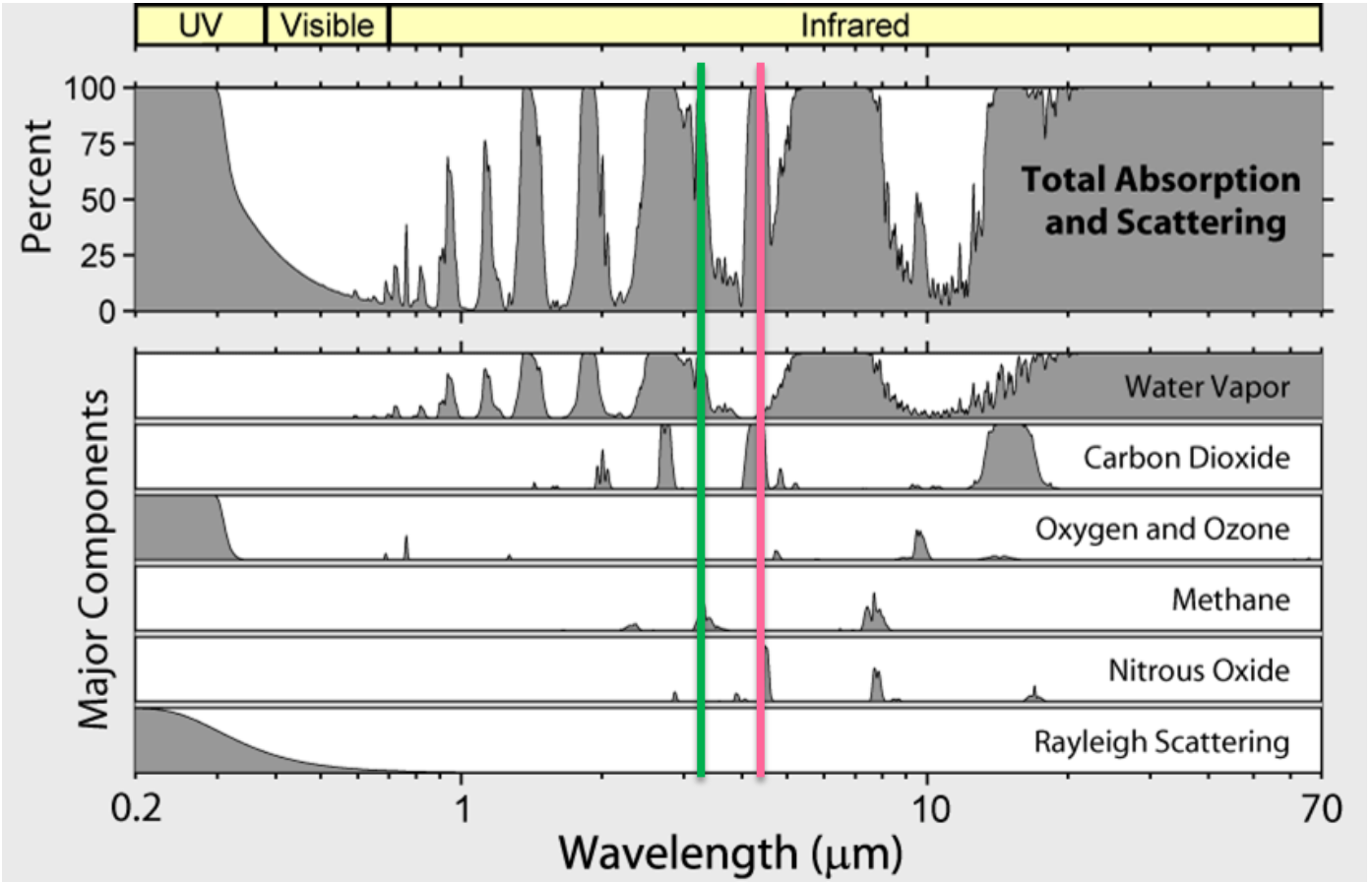


1% of increase in the nonlinear interaction with light results in 10% better selectivity

Solution absorption overlap – coherent waves

- 2 – Absorption bands of different gases overlap
- Use Two coherent waves – „see” anharmonic vibrations





Why chalcogenides?

- Transparent in the mid-IR region, sometimes as far as 2-20 μm
- Good signal to noise ratio
- Birefriengence may be large enough to meet phase matching conditions
- High energy of phonon subsystem assisting in NLO in the IR range – large NLO coefficients $\chi_{ijk}^{2\omega}$
- Relatively high photothermal stability up to 400-500 MW/cm^2 within spectral range 3-12 μm
- Not hygroscopic
- Good mechanical processability

Results obtained so far

- More than 50 crystals tested so far:

  Ag_3AsS_3, Ag_3SbS_3, $AgGaSe_2$, Tl_3AsSe_3, $CdGeAs_2$, $Ag_2In_2SiS_6$, $Ag_2In_2GeS_6$, $Ag_{0.98}Cu_{0.2}GaGe_3Se_8$, $Ag_xGa_xGe_{1-x}Se_2$(x=0.333; 0.25; 0.2; 0.167), $AgGaGe_3Se_8$:In(Si,Sn) …

- The most perspective groups are $AgGaGeS_4$ i $AgGaGeS_4$:Cu
- Maximum SHG efficiency obtained = 27%

Challenges

- Most chalcogenide crystals have <5% efficiency. Our goal is to increase it in the spectral range 3-9 μm using the solid state alloys of appropriate crystals
- Compromise between high efficiency and thermal stability has to be sought
- Decrease of the indesirable scattering caused by irreversible changes in crystals

Future research plans

- Laboratory for field testing on 70 m distance (under construction, almost finished) with measure chamber with controllable gases content
- Making the LIDAR mobile (a car)
- Testing more quaternary chalcogenide crystals from the $AgGaGe_3Se_8$ group and others
- Large NLO susceptibility coefficients – chances for high wave conversion yield
- Extension of signal to noise ratio in the wider spectral range
- Decrease of the minimum laser power density by the use of more efficient crystals

CONCLUSIONS

- It is possible to convert IR LIDAR wavelength to its half, one third or other wavelengths. LIDAR spectral range significantly extended!
- Coherent bicolor illumination increases measurement selectivity (anharmonic vibrations included), so far by 10%
- Conversion to SHG only 27% at best so far, research is ongoing
- The cost of the NLO upgrade would be 20 000 EUR – 10% of the equipment price