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Ultrafast photo-induced dynamics of molecular materials under high pressure

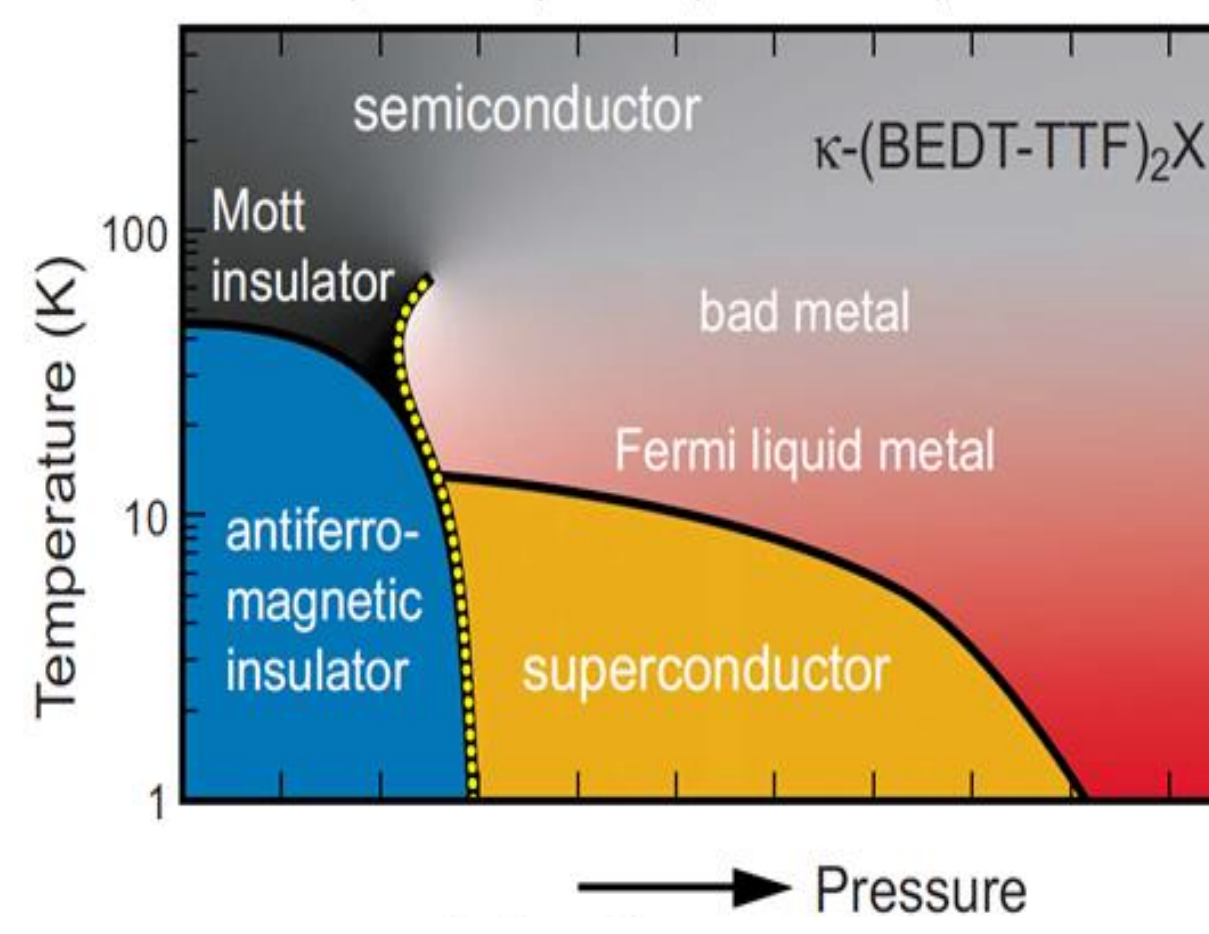
T. Gauthier, G. Privault, J-C. Ameline, E. Trzop, G. Huitric, M. Hervé, G. Azzolina, N. Godin, M. Maesato, G. Saito and R. Bertoni

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1| State of the art [1]

For a long time, the study of photo-induced phase transition was restricted to the sole use of temperature as external control parameter. Recent studies have demonstrated the possibility to add the use of **hydrostatic pressure to explore the phase diagram** of material. The idea is to look for parts of the phase diagram near critical points where gigantic photo-response may occur.



Example of complex phase diagram (P,T) of molecular material

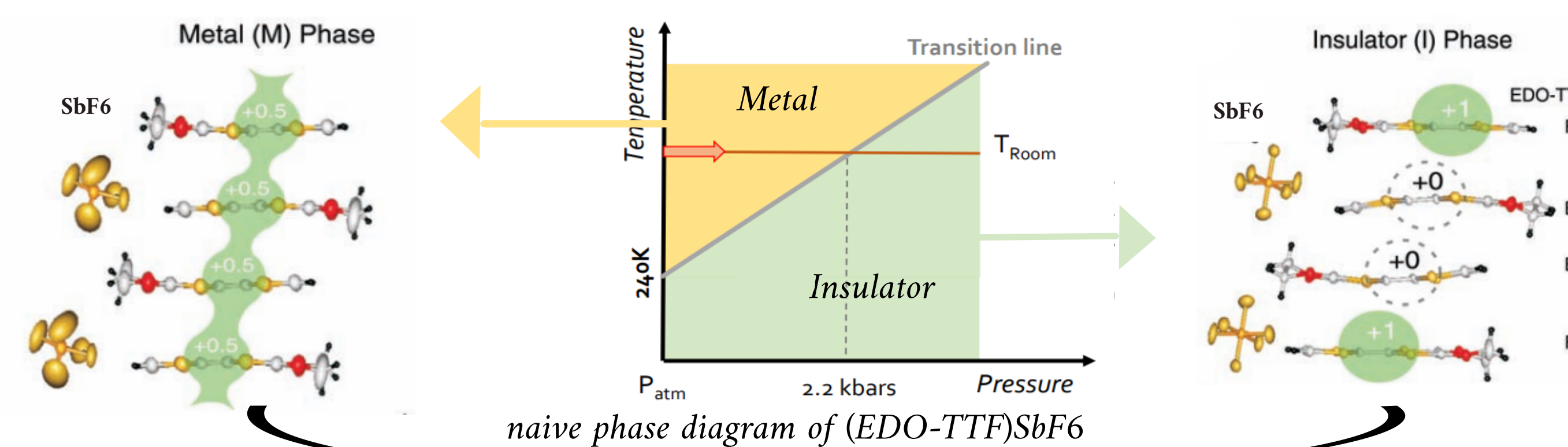
Goals

- Exploring the Pressure Temperature Phase Diagram of Molecular Material
- Seek for peculiar the Photo-Induced Dynamics in well defined parts of the Phase Diagram

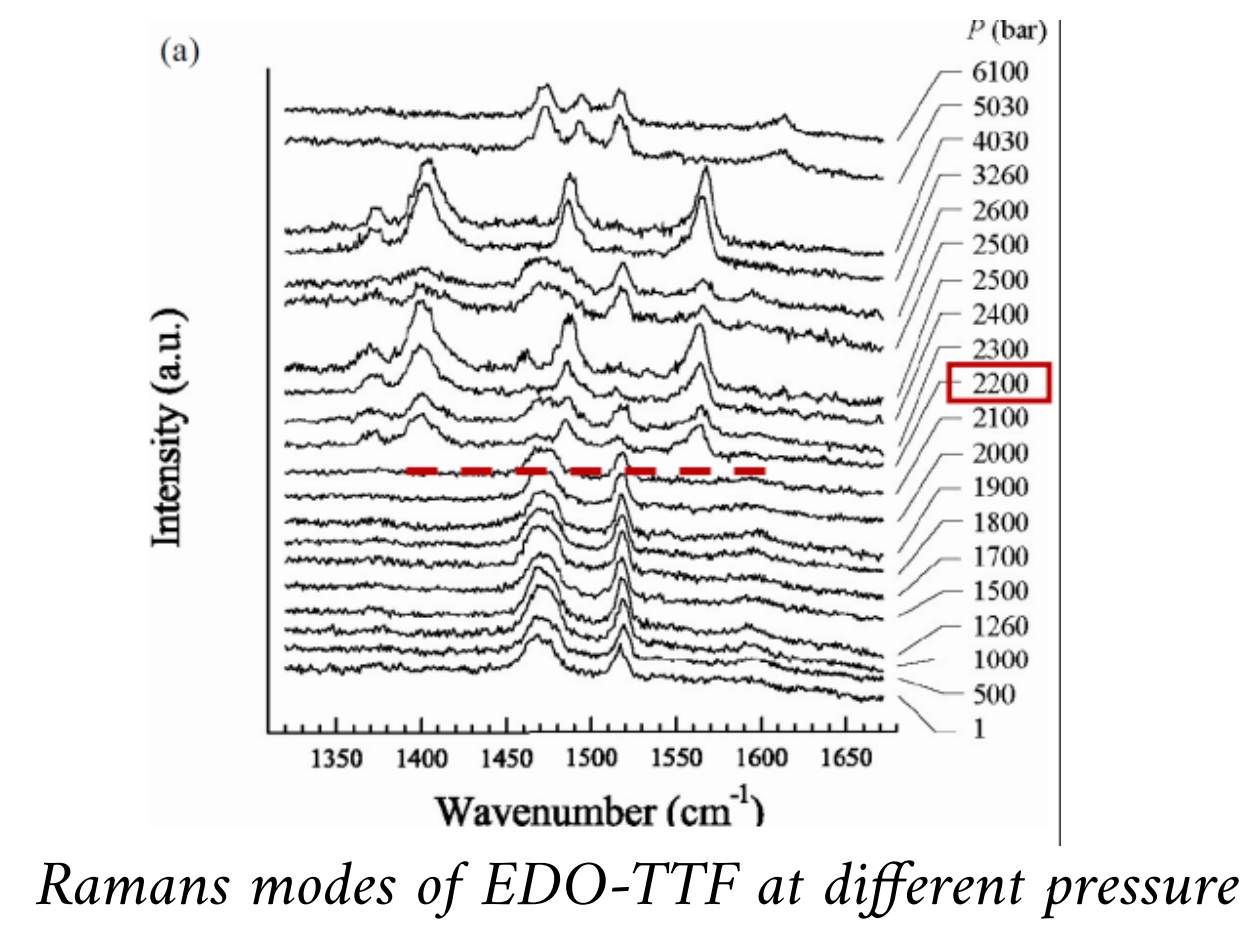
Using Pump-Probe experiment to study optical response of different phase, the width of pulses permit us to follow electronics and structural dynamics.

• Seek for peculiar the Photo-Induced Dynamics in well defined parts of the Phase Diagram

2| (EDO-TTF)SbF6 phase transition [2][3]



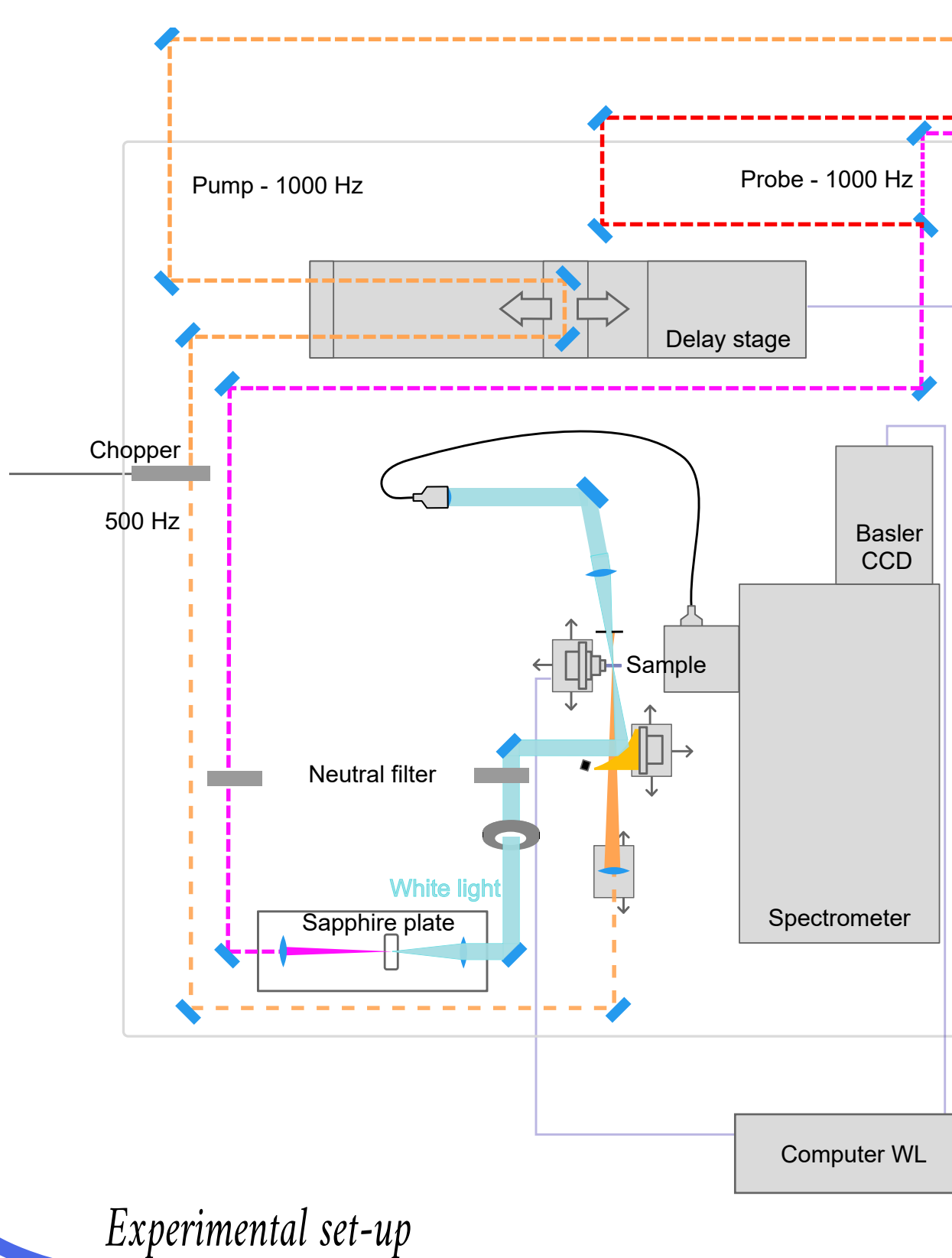
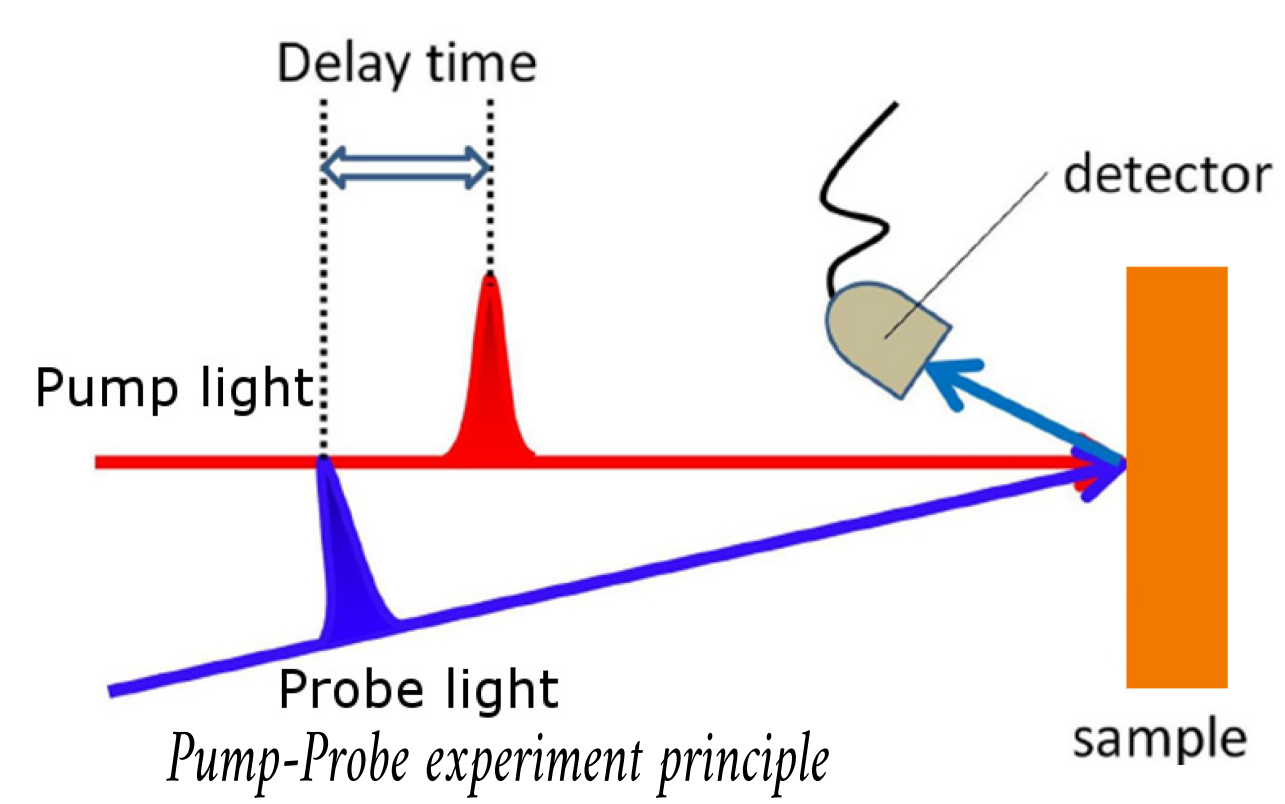
First order phase transition with **symmetry breaking**, the unit cell is doubling between Metal to insulator phase.



Ramans modes of EDO-TTF at different pressure

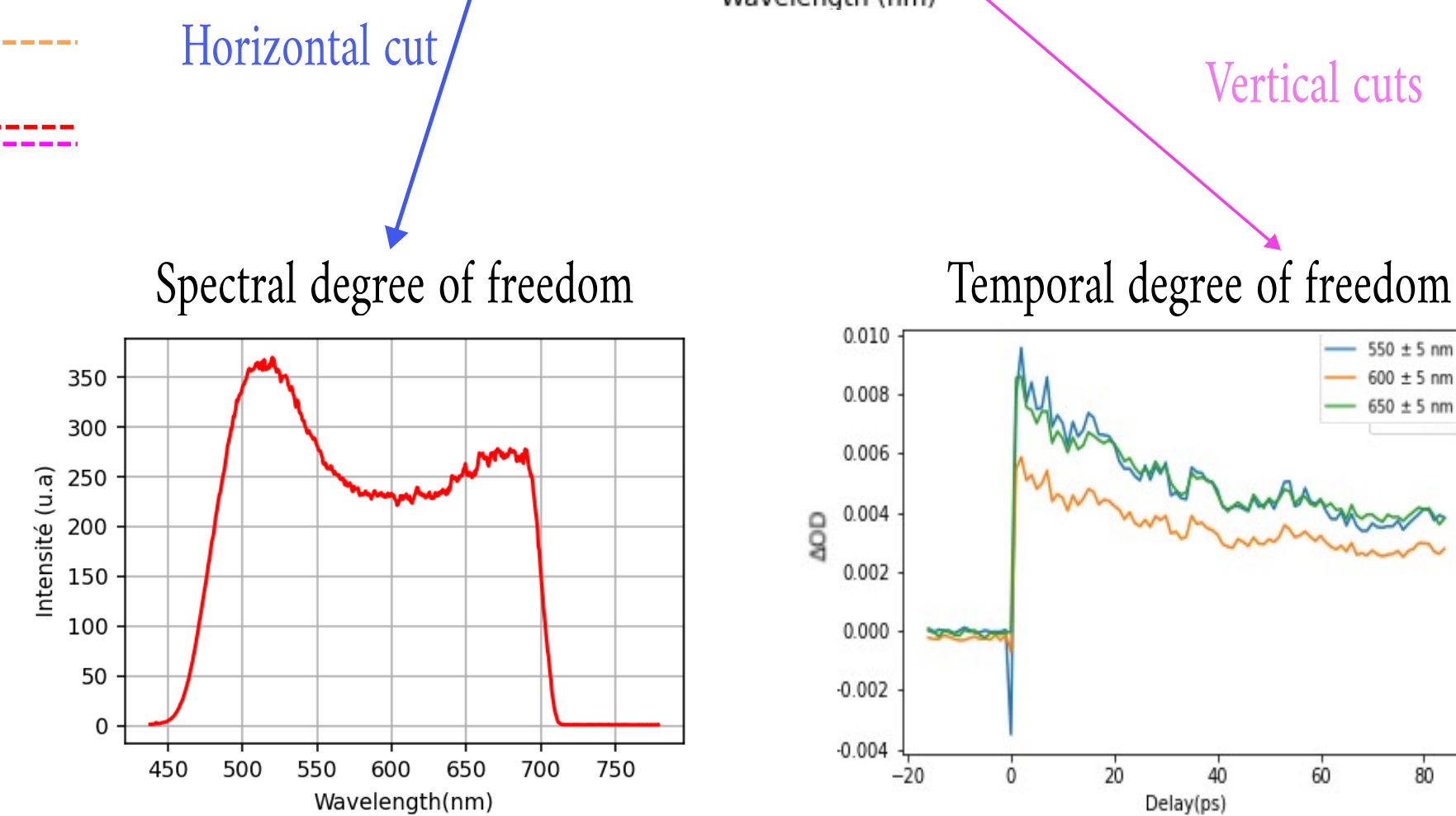
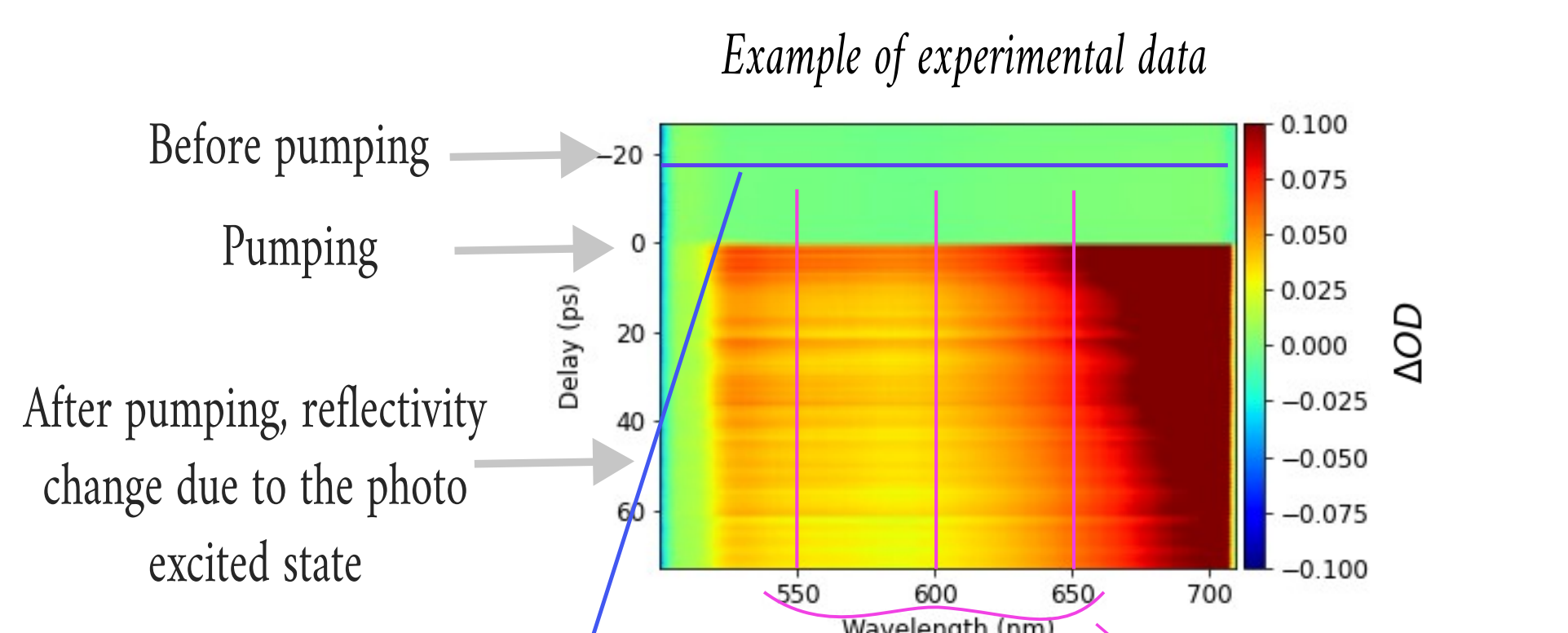
Reflectivity of (EDO-TTF)2SbF6 salt measured at 635 nm in cooling (▼) and warming (▲) modes. The first-order phase transition is characterized by a **thermal hysteresis** spanning from 227K to 249 K.

Optical set-up



Experimental set-up

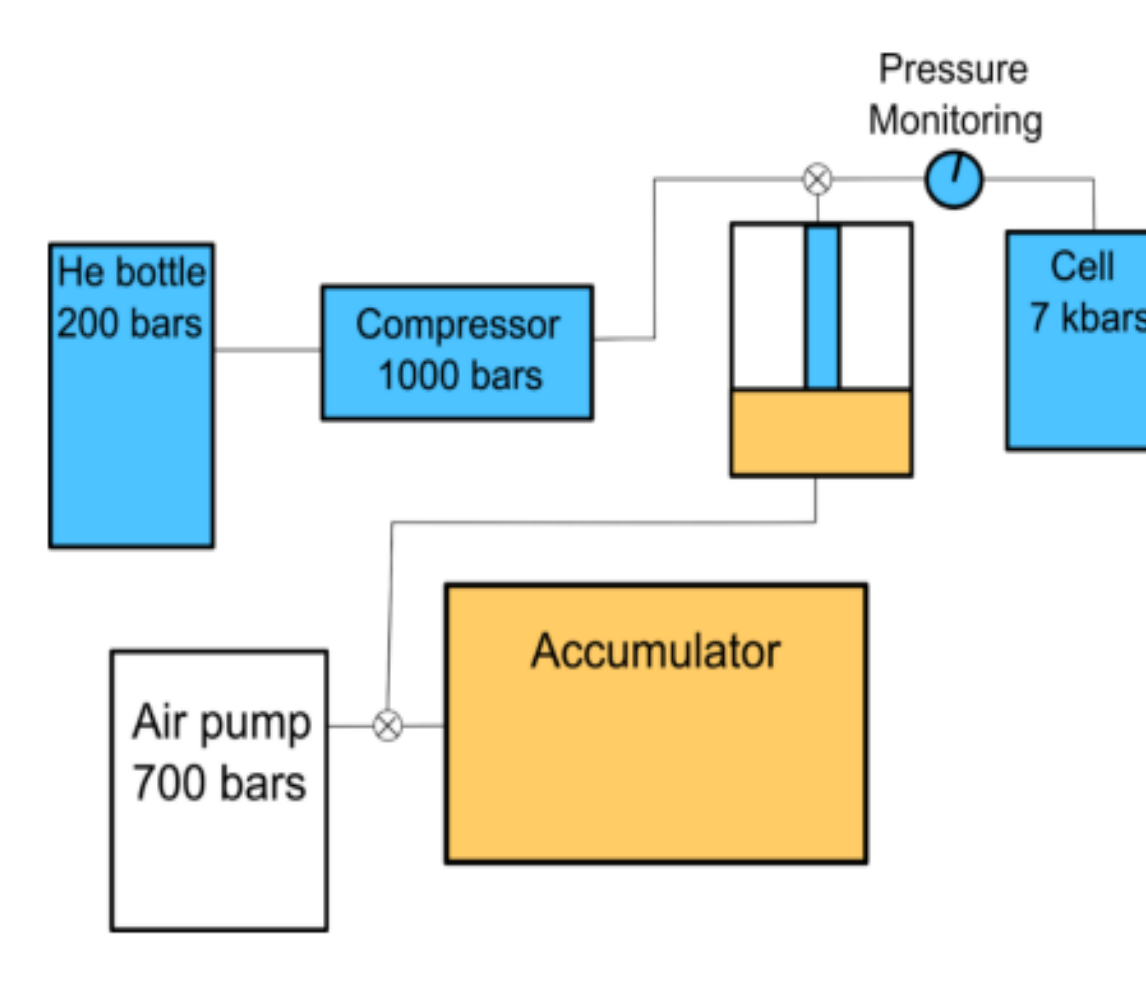
3| Experimental techniques



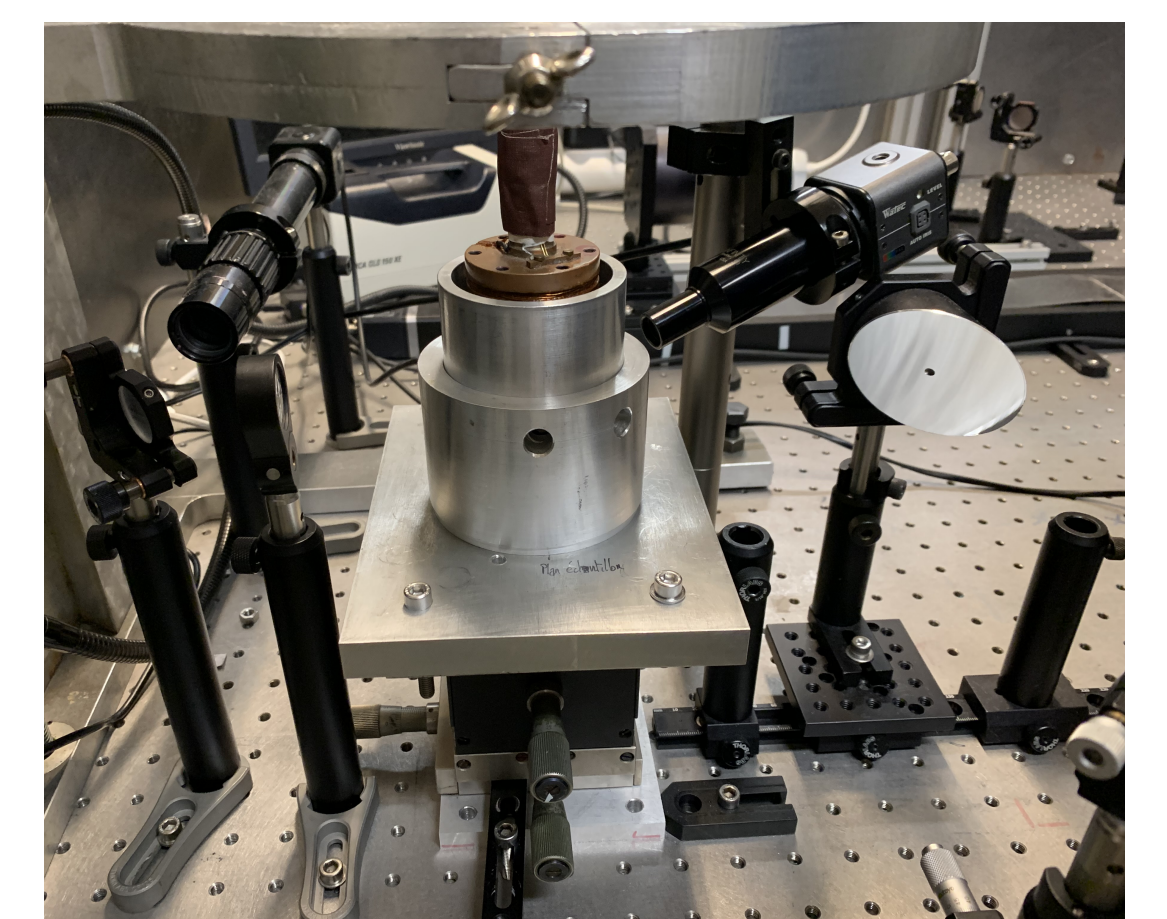
Supercontinuum/ White light Spectroscopy:

- (470-750 nm)--> Electronic transitions
- Time resolution ~ 100 fs (electronic and structural dynamic)
- Pump (450-2200 nm)

Pressure Set-up



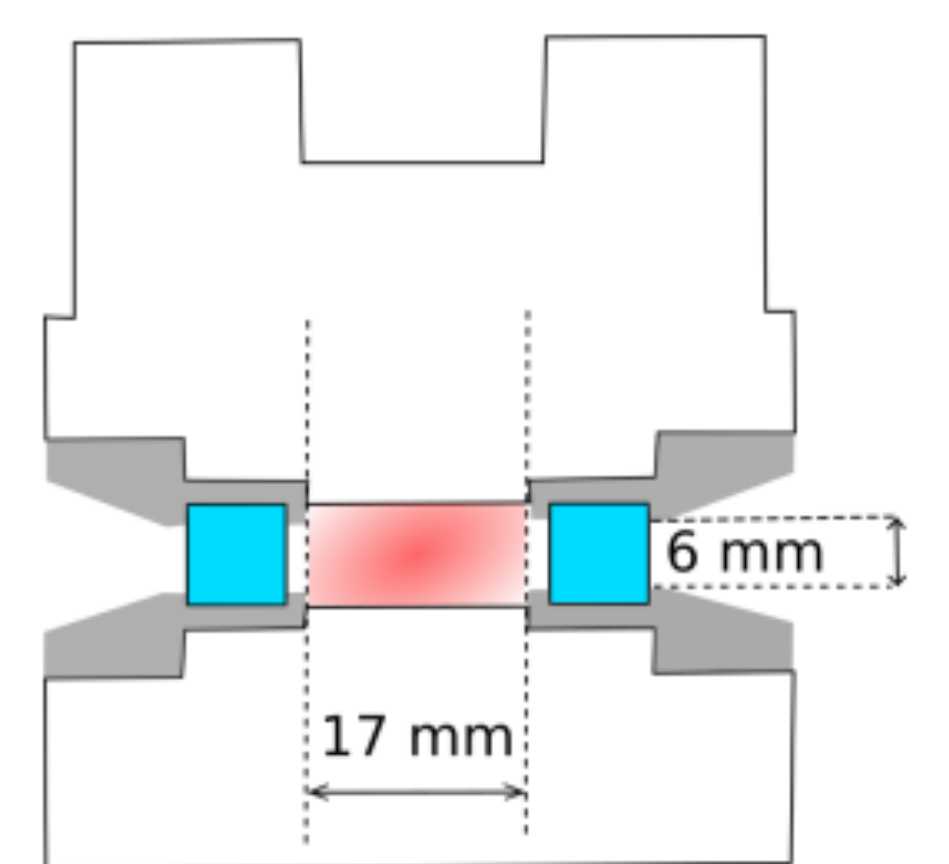
Multi-stages He gas based cell



Picture of the high pressure cell in the optical setup

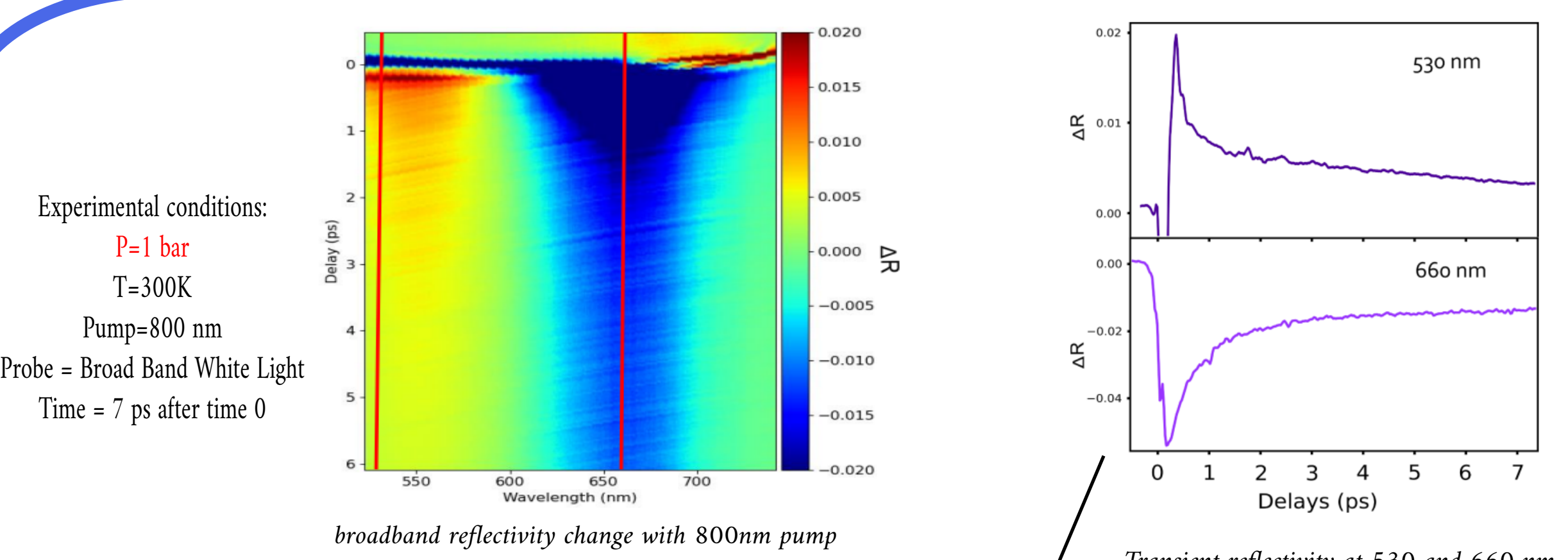
- Specifications:
- Precision of few bars (measure in operando)
 - Ramping up/down of few bars per minutes
 - Sapphire windows allowing optical Transmission/Reflection

Apply **hydrostatic pressure with Helium gas**. Material in the cell can make several square millimeters of surface as opposed to a cell with diamond anvil where the samples are of the order of 10 μm^2



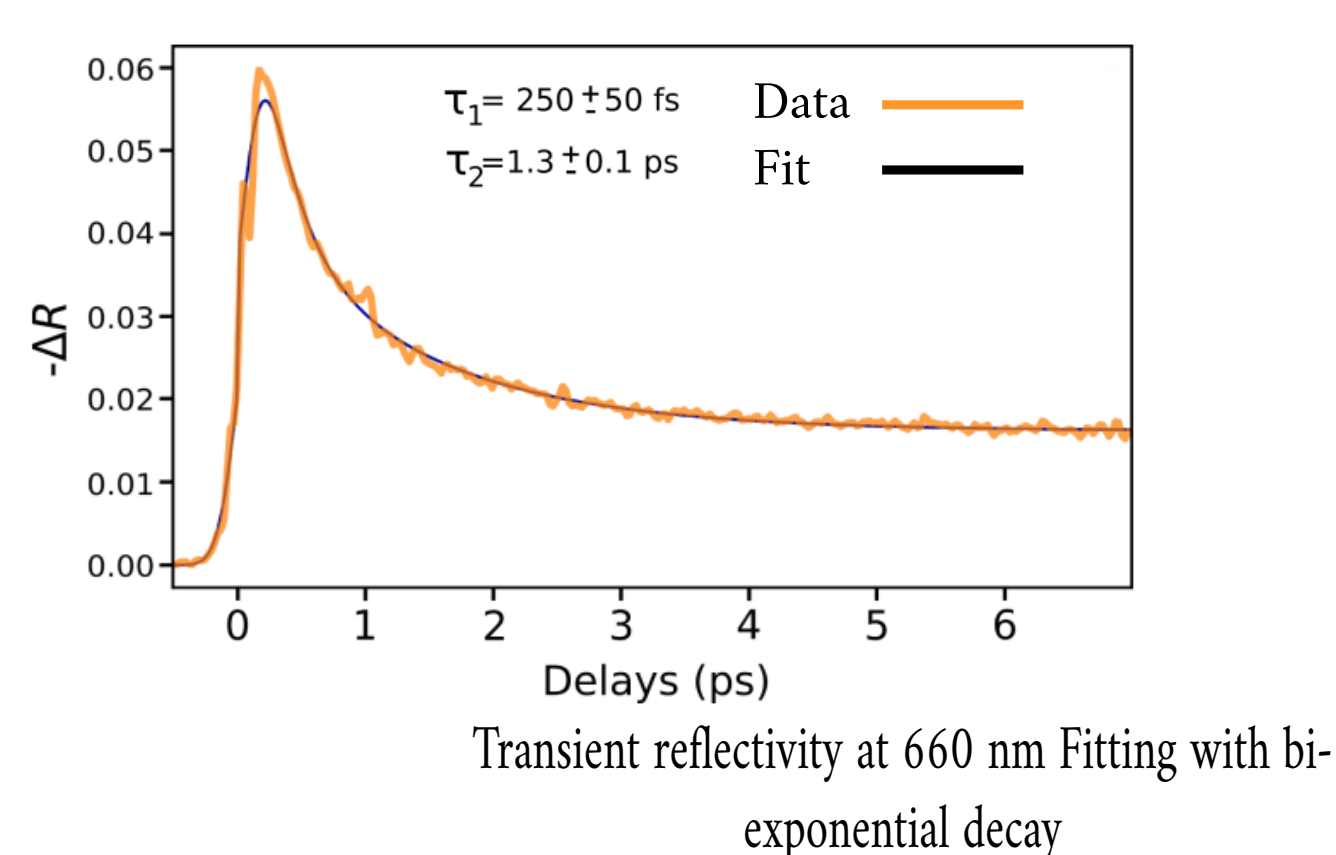
Schematic cut of high pressure cell

4| Results



Model with bi-exponential decay:

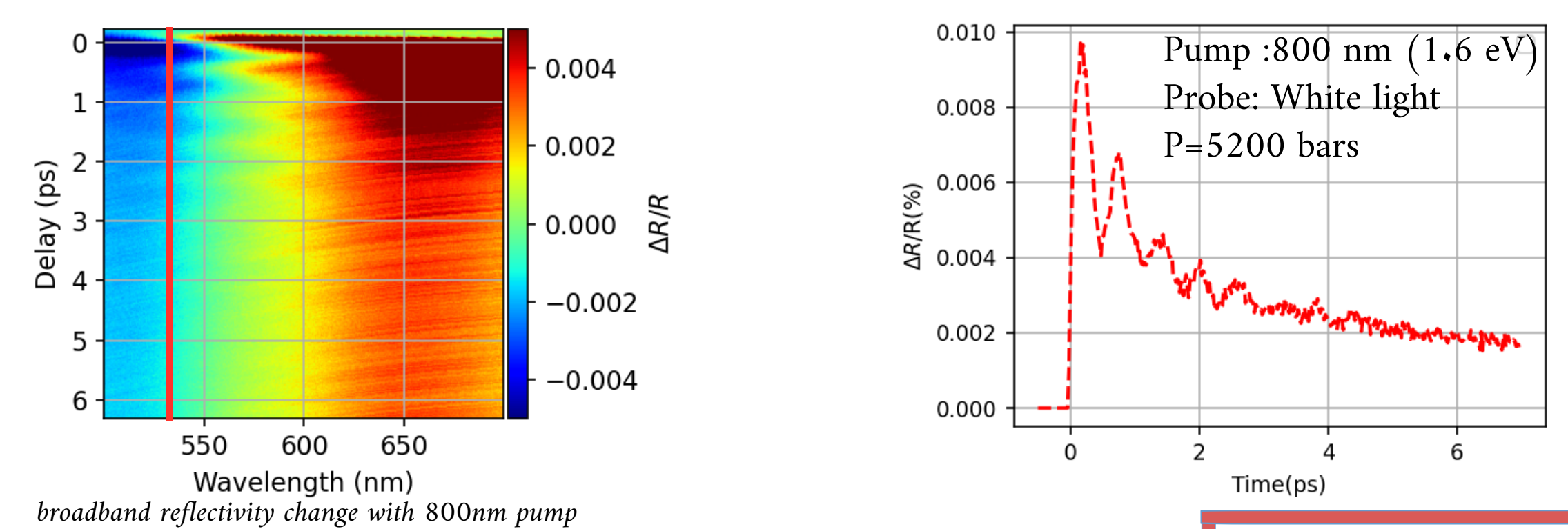
- 250 fs electron-electron interaction?
- 1.3 ps electron-phonon interaction?



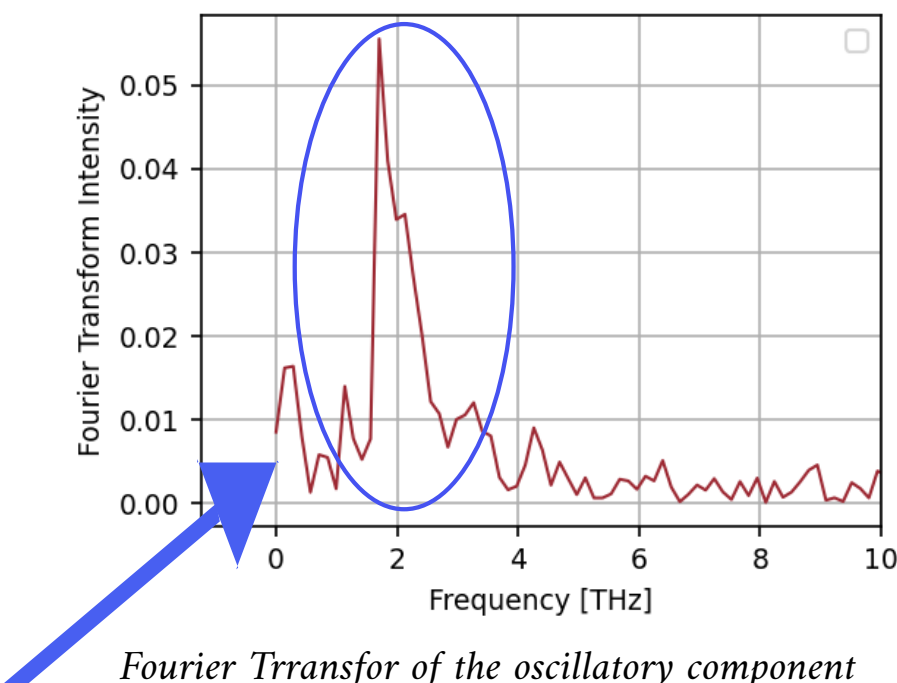
At 1 bar and 300K, the electronic excitation with the 800 nm pump do not couple with any structural movement because at this thermodynamic condition we are in the metallic phase. We observe the relaxation of excited carriers with exponential law.

Experimental conditions:

- P=5200 bars
- T=300K
- Pump=800 nm
- Probe=White Light
- Time = 7 ps after time 0



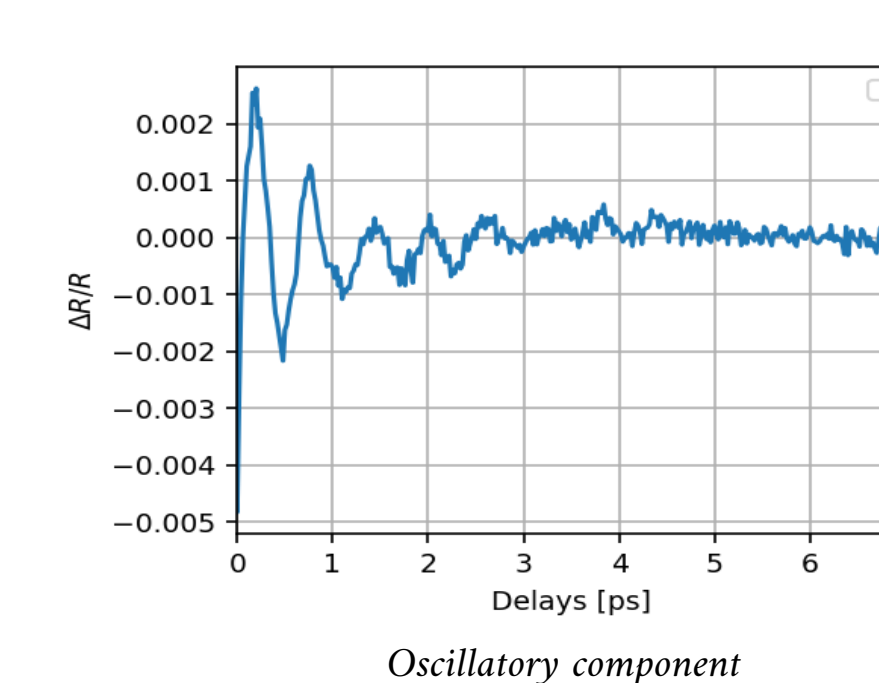
broadband reflectivity change with 800nm pump



1.67 THz optical mode

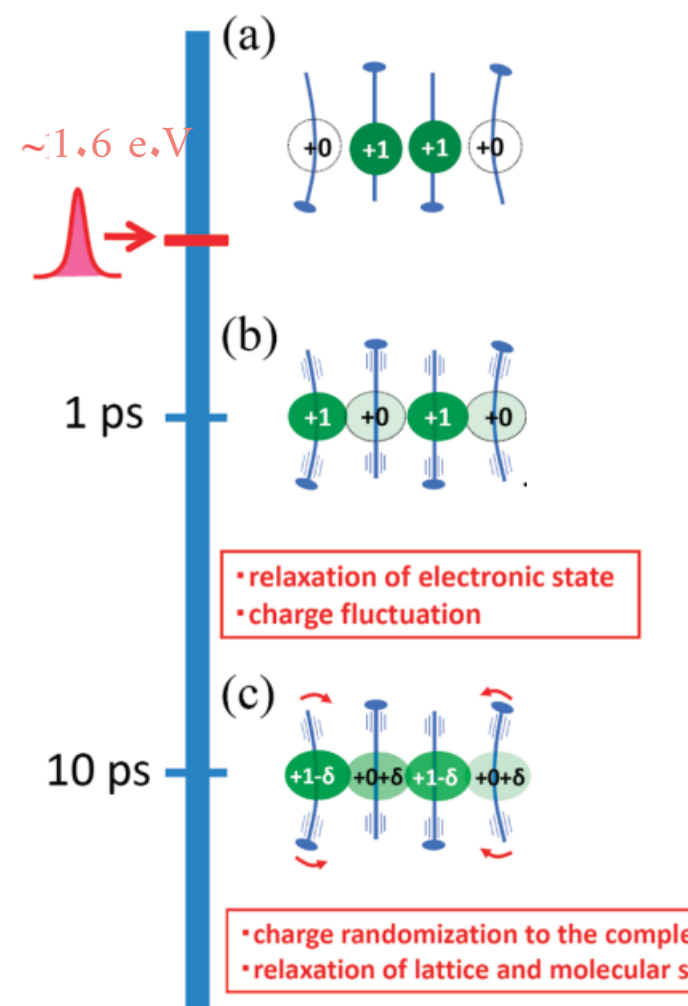
Optical mode at 1.67 THz coming from the electronic excitation of the pump and which couples to structural movements

Using the FT result to fit the data.



At 5200 bars and 300K the system is in the Insulating phase (a), electronic transfer create a new charge ordering (b) which couples with structural movement, this movement induce change in dielectric constant which is directly related to optical measurement.

Schematic of proposed time evolution of the charge pattern and molecular structure after photoexcitation of the CT transition in the insulating phase of (EDO-TTF)2SbF6 [4]



5| Conclusion

- Broadband ultrafast optical spectroscopie under high pressure (0-6kbars)
- Time resolution as good as 100 fs
- Observation of coherent optical phonons
- Possibility to distinguish the metallic and the insulator phase of (EDO-TTF)SbF6

[1] D. Faltermeier et al, Phys Rev. B 76 (16), 165113 (2007)

[2] M. Chollet et al, Sciences, 307 (5706) 86, 2005

[3] M. Maesato et al, J. of Phys.: Conference Series 148 (1), 012004, 2009

[4] Naoto Fukazawa, Minoru Shimizu, Tadahiko Ishikawa, et al, The Journal of Physical Chemistry C, American Chemical Society, Mar 1, 2012

