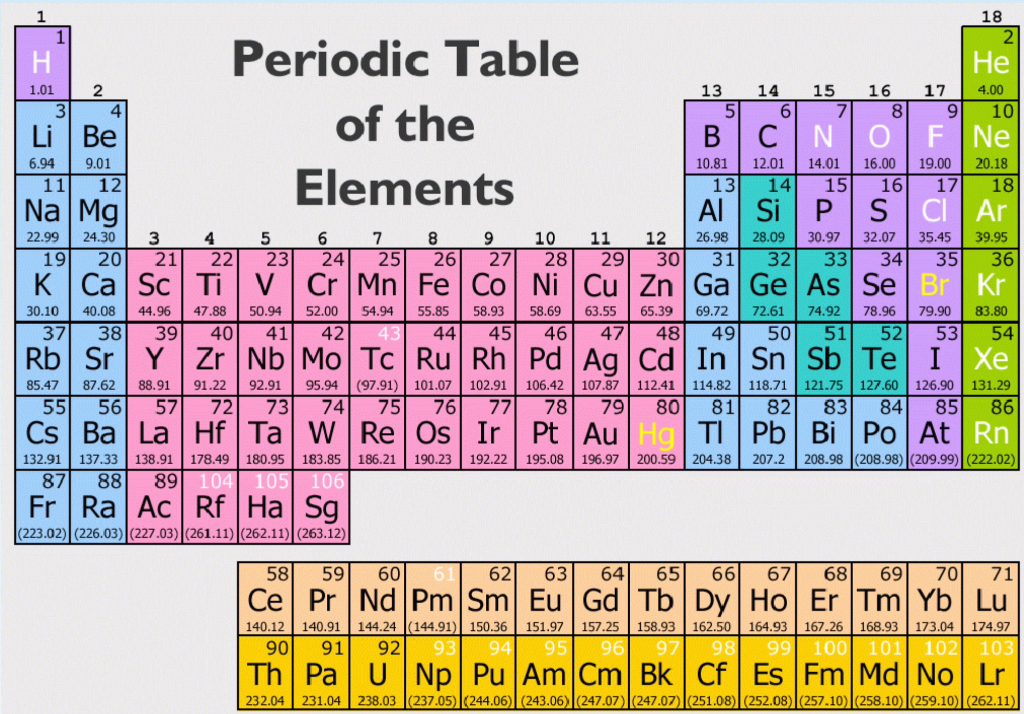
Time Resolved Optical Spectroscopy of Na_2IrO_3

Nicola Nembrini

What is Na_2IrO_3?



3d transition-metal oxides (Ti-Oxides, Mg-Oxides, Ni-Oxides, Cu-Oxides) are caracterized by 3d localized orbitals and small mass .

- Large on-site Coulomb repulsion,U (~5-6 eV)
- Small bandwith,W (~1 eV)
- Negligible spin-orbit coupling, SO

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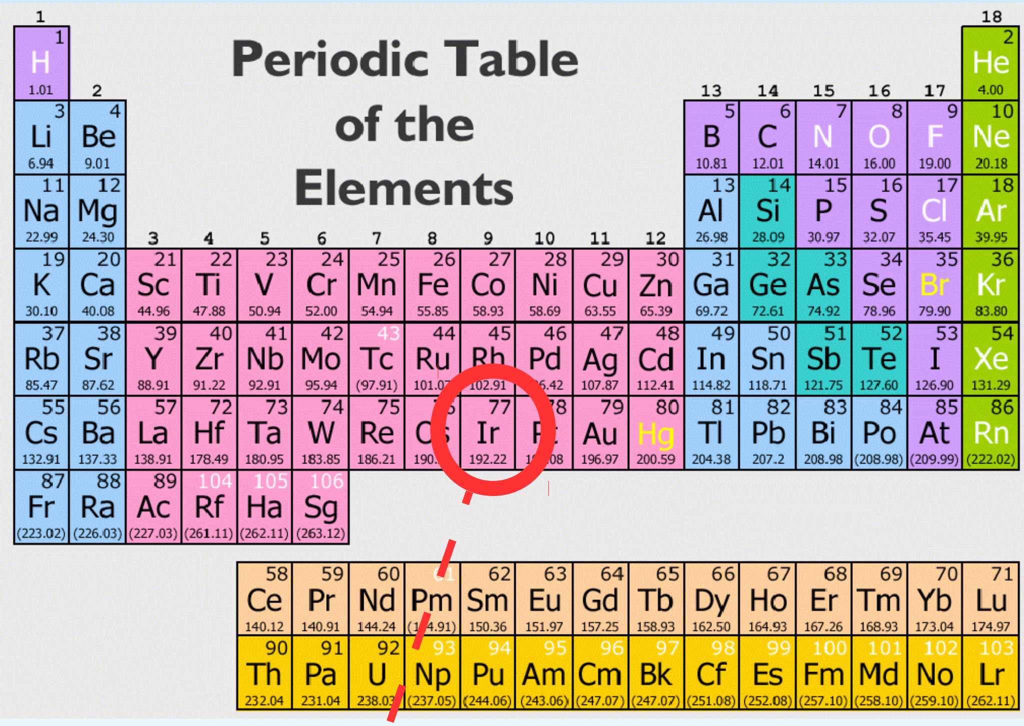

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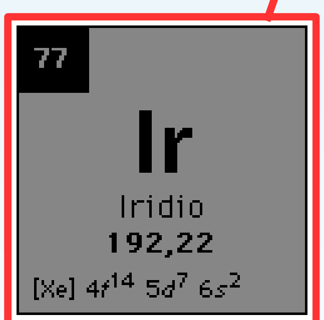
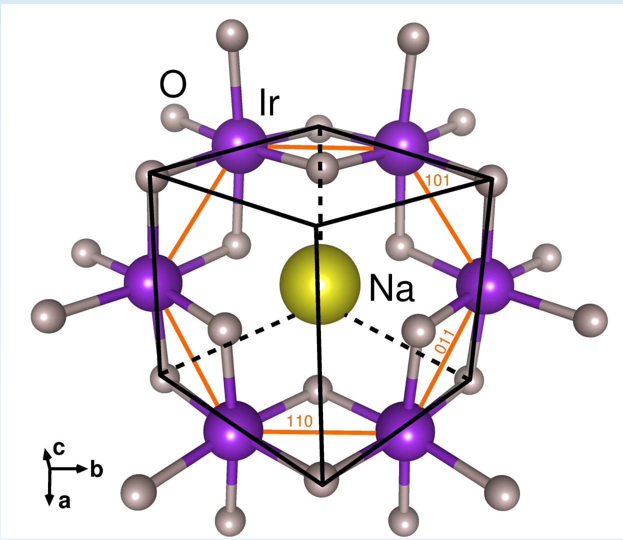


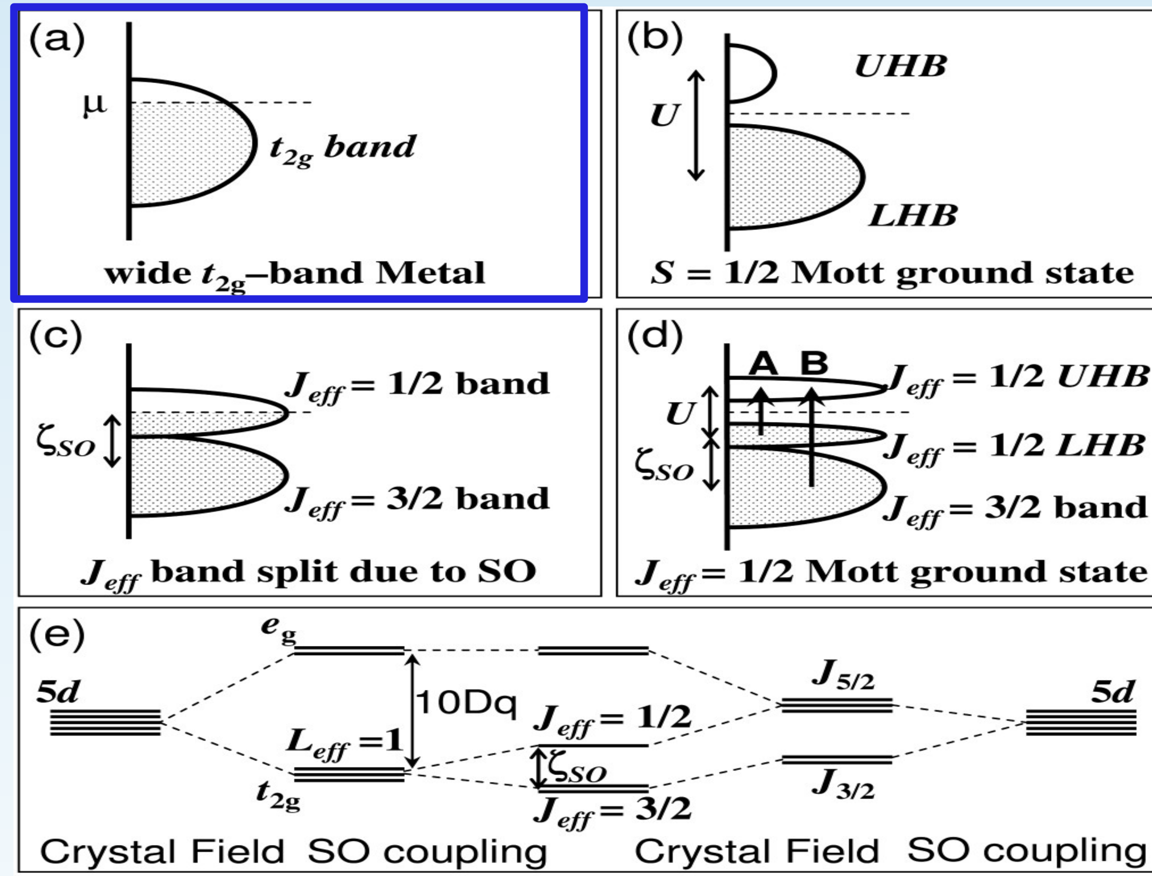
Na_2IrO_3 is 5d metal oxides:

- More delocalized orbitals (5d) → U ~ 1-2 eV
- large weight (SO ~ 1-2 eV) $H_{S.O.} = \frac{1}{2m^2c^2r}(\hat{S}\cdot\hat{L})\frac{dV}{dr}$ → Z

In this system U, W and SO are comparable (about 1-2 eV)

Properties of Na_2IrO_3 at equilibrium





- Crystal field
- Spin orbit coupling
- Coulomb repulsion

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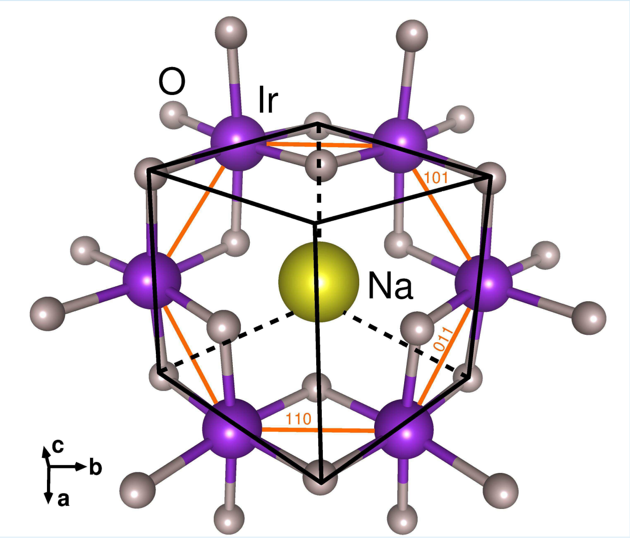

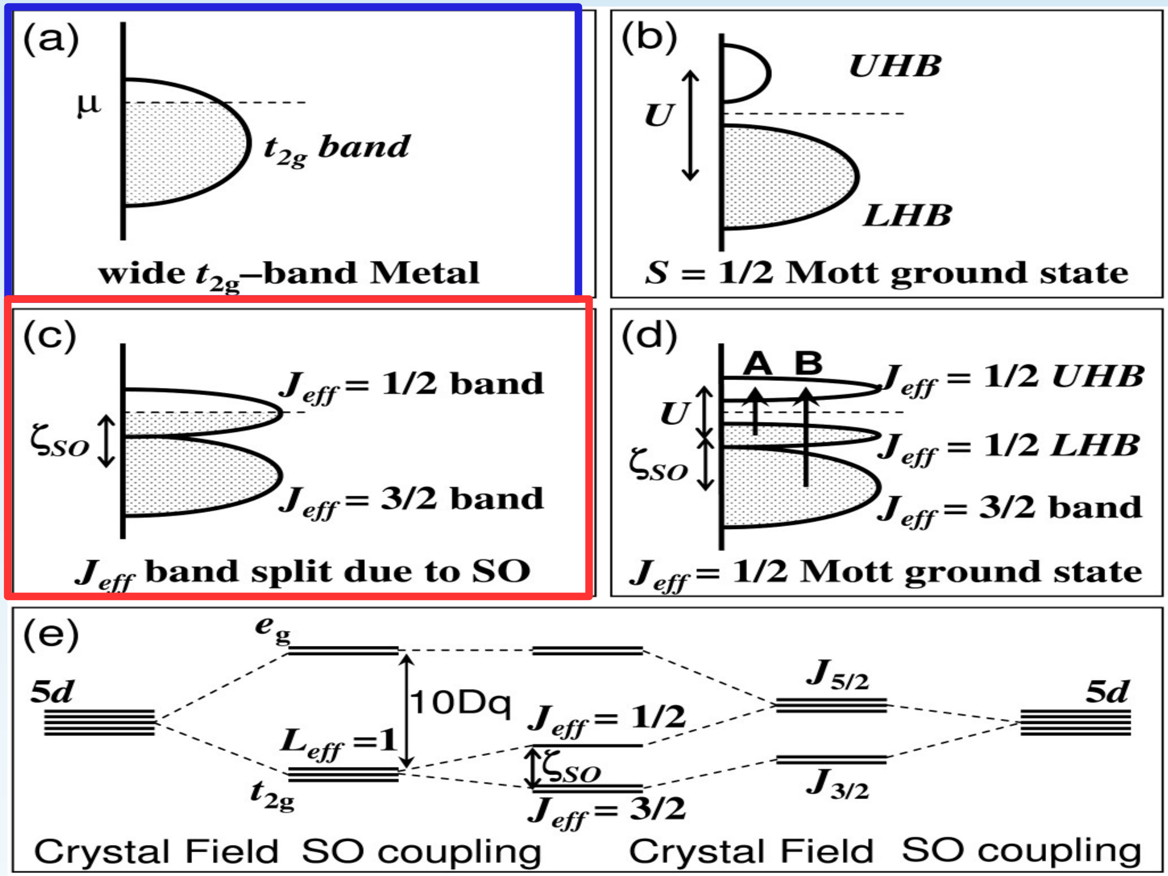

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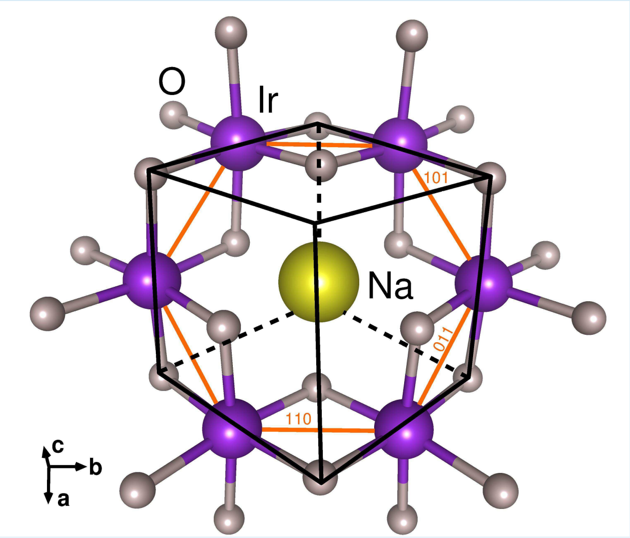
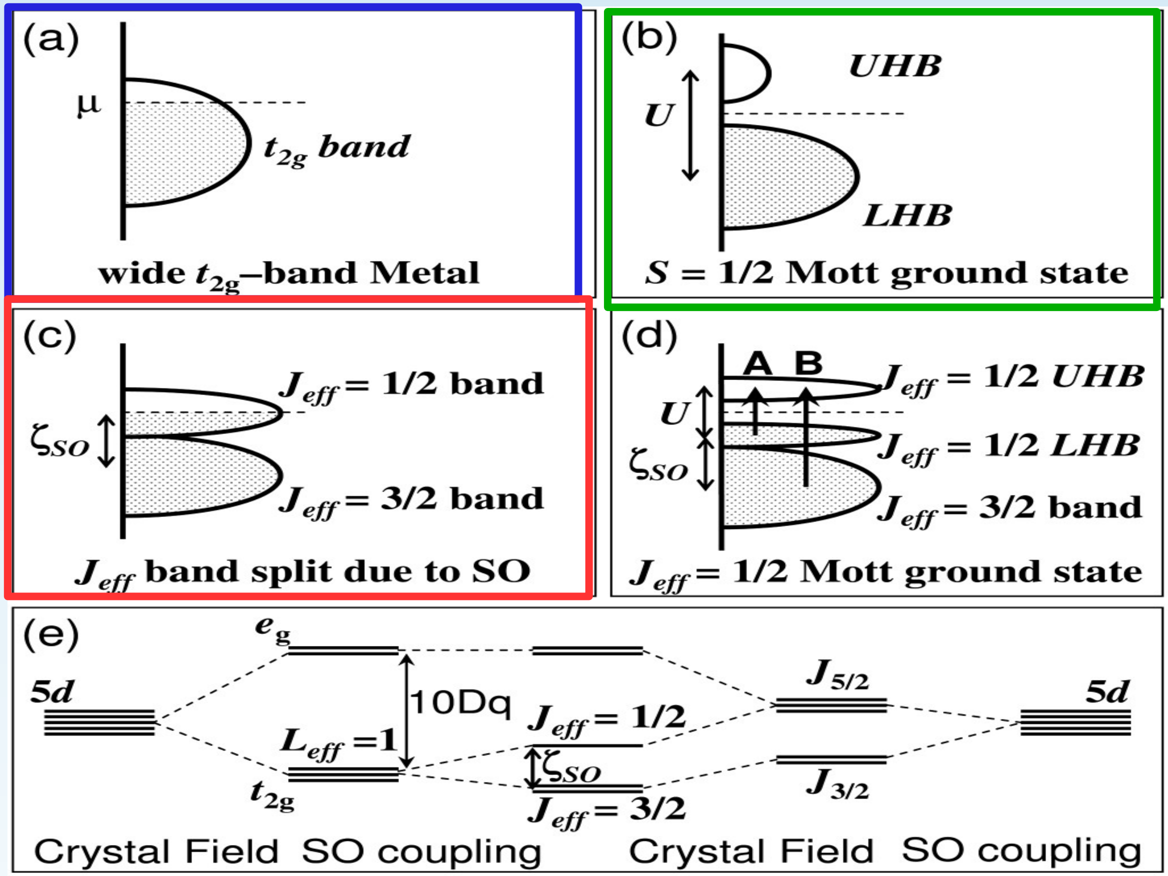




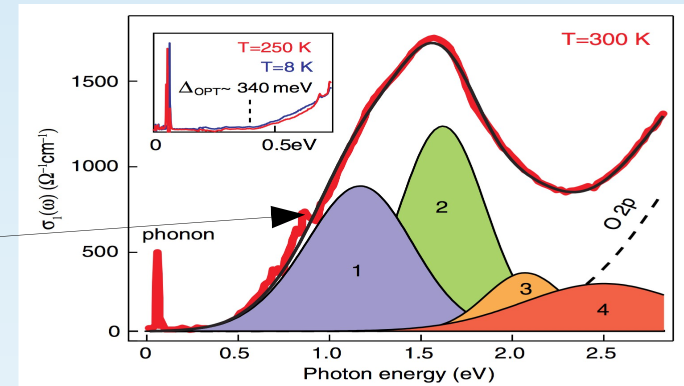
- Crystal field
- Spin orbit coupling
- Coulomb repulsion

Electronic band structure?

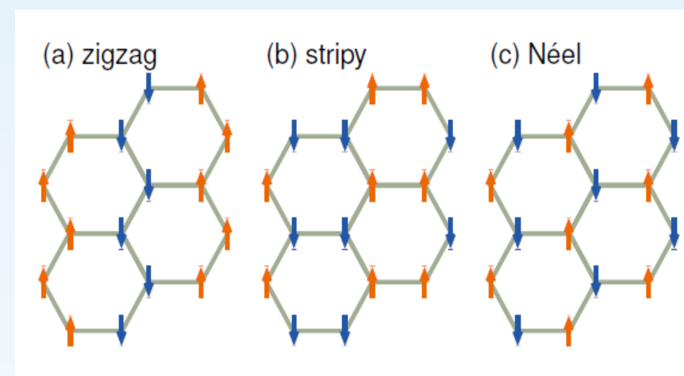
Physical properties of Na_2IrO_3 at equilibrium

- At T= 300K is a Mott Insulator (340meV)

Optical conductivity data (red line)
[R. Comin,A.Damascelli et al.]



- At T=15K present an antiferromagnetic order



Possible antiferromagnetic pattern of Na_2IrO_3

How W, SO and U determine these properties?

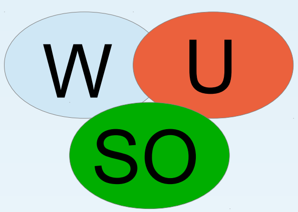
Time-resolved optical spectroscopy

Non equilibrium spectroscopy

At equilibrium state the energy scales are comparable.

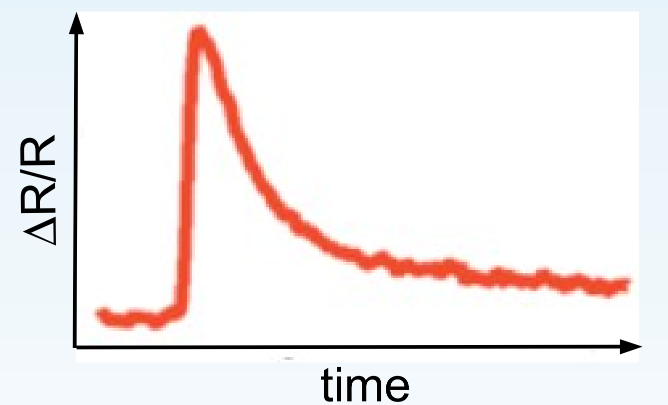
Bringing the system out of equilibrium is possible to decouple these contributions on time scale

At equilibrium comparable scale of energy

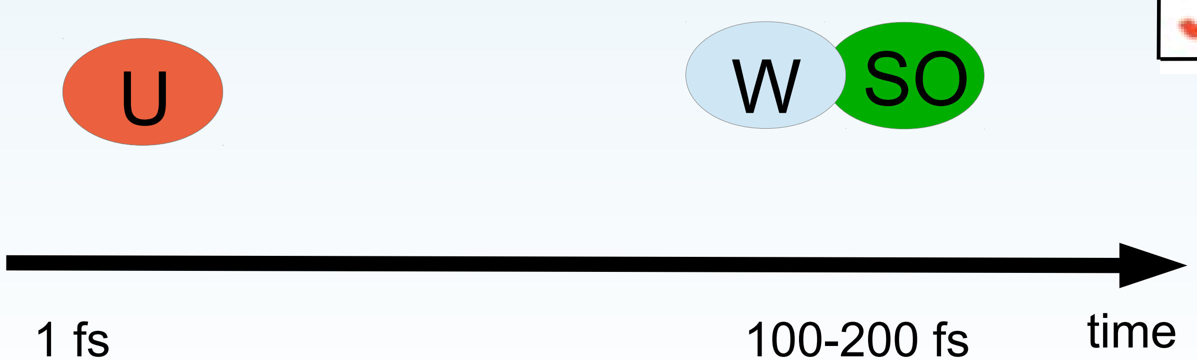


Tipically in time scale

U << SO and W



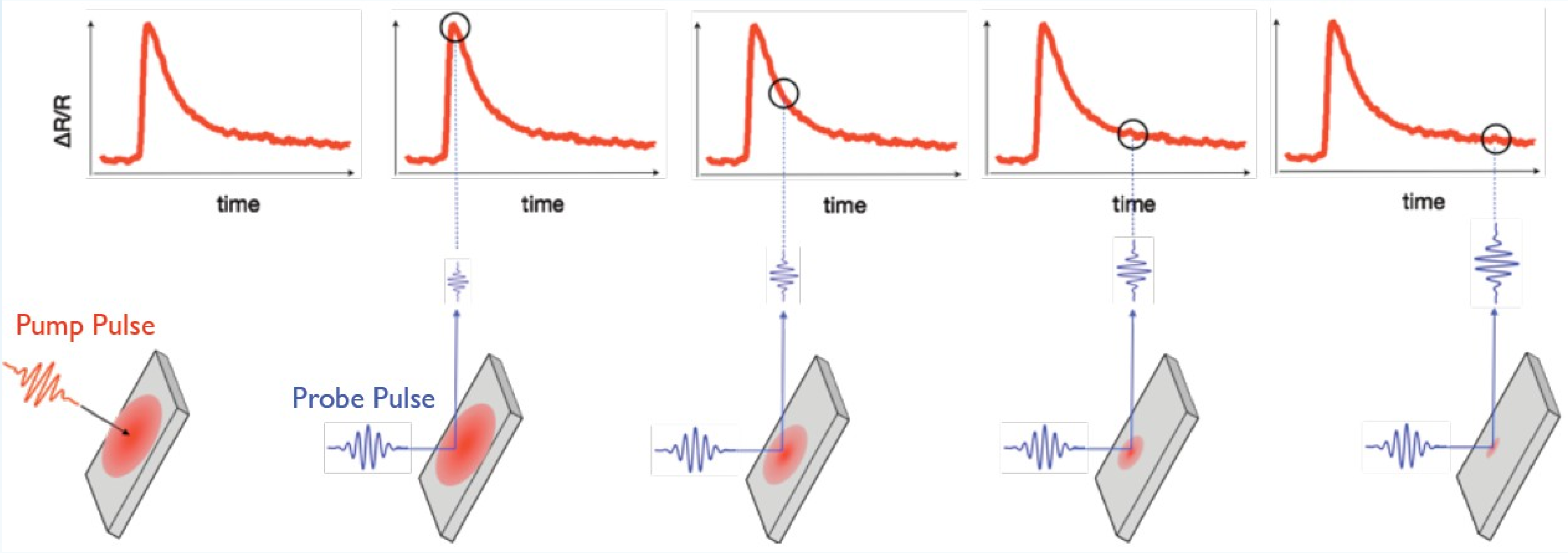
Out of equilibrium: different time scale



Time resolved spectroscopy: Pump & Probe

Two ultrashort pulses (120 fs) are used .

- Pump pulse (1.5 eV) to excite the system
- Probe pulse (singlecolor or supercontinuum) to take a snapshot of system's variation reflectivity for several delay times than pump pulse

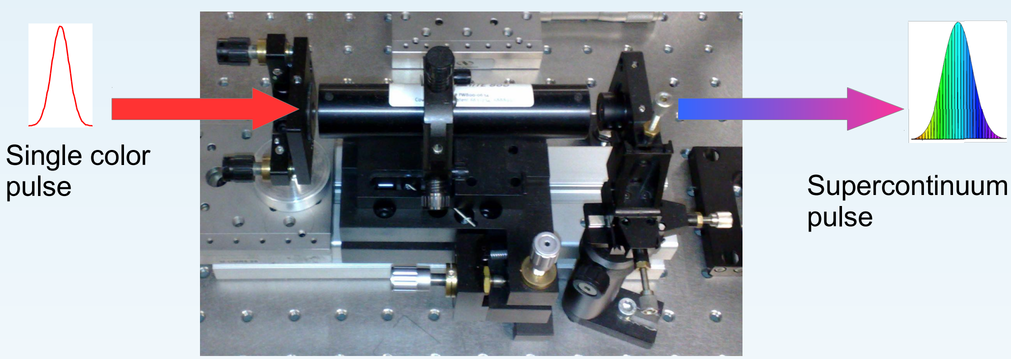


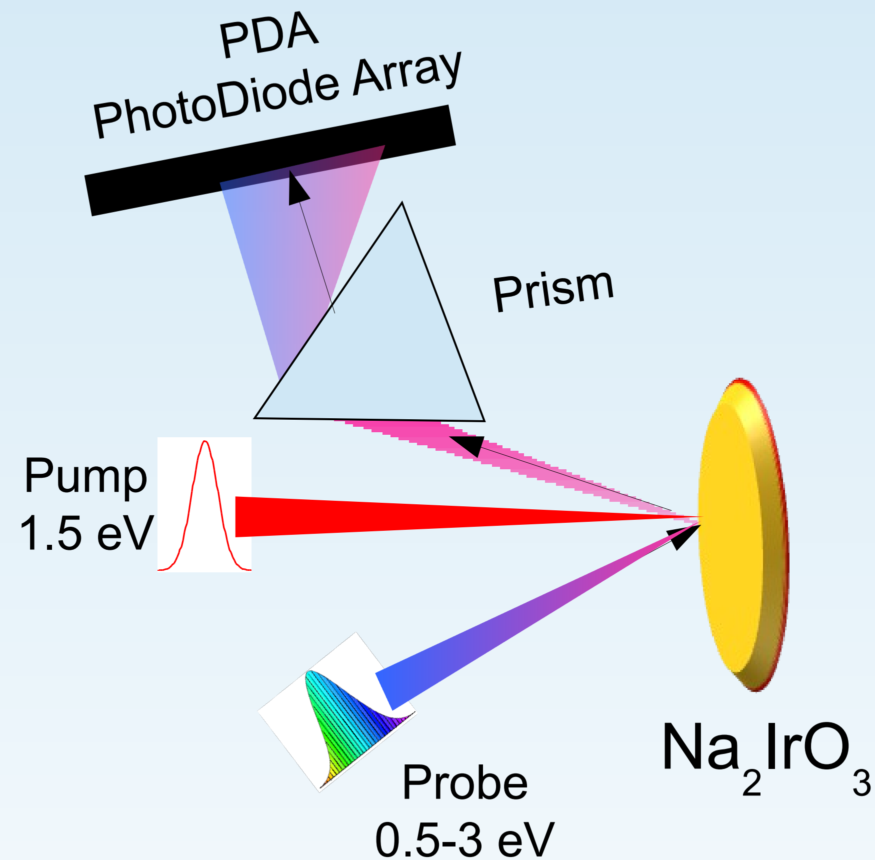
- Investigate dynamics of several ps (~10 ps)
- High time resolution of fs (~100 fs)

Time resolved spectroscopy Pump & Probe

Using a non-linear fiber is possible to generate ultrashort white-light pulses in order to have:

- Time-resolved measurements
- Frequency-resolved measurements

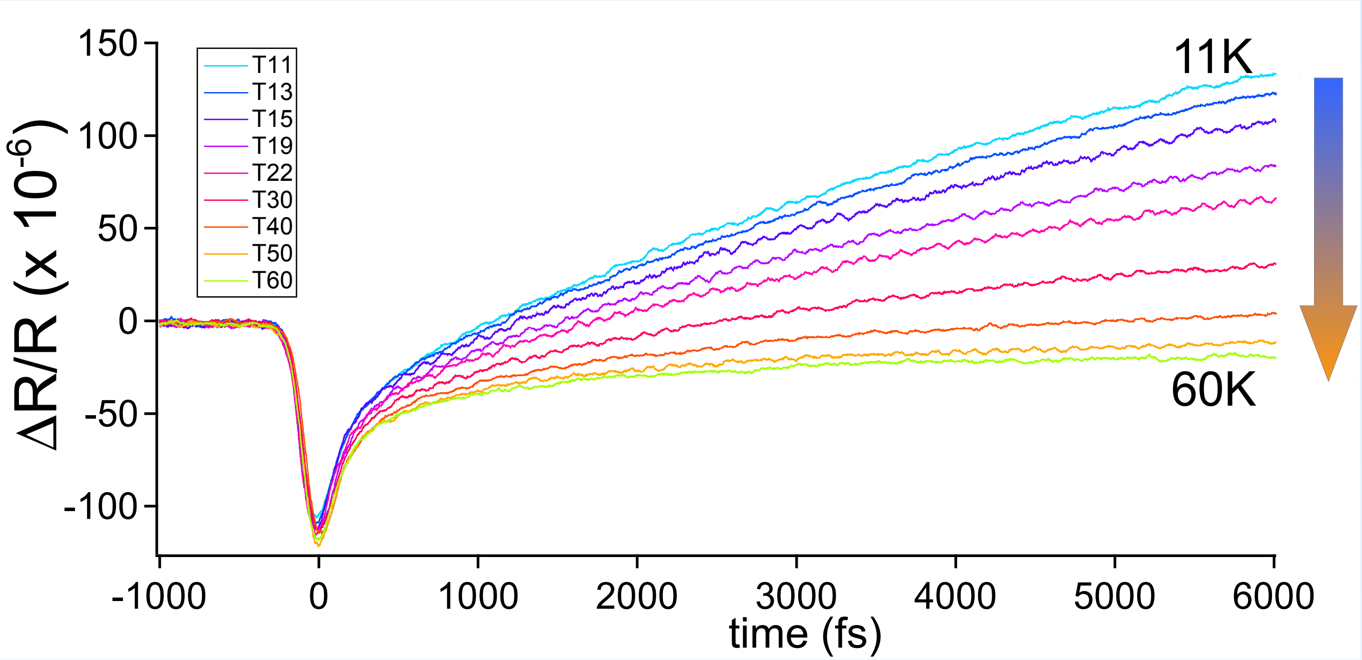




Experimental ΔR/R

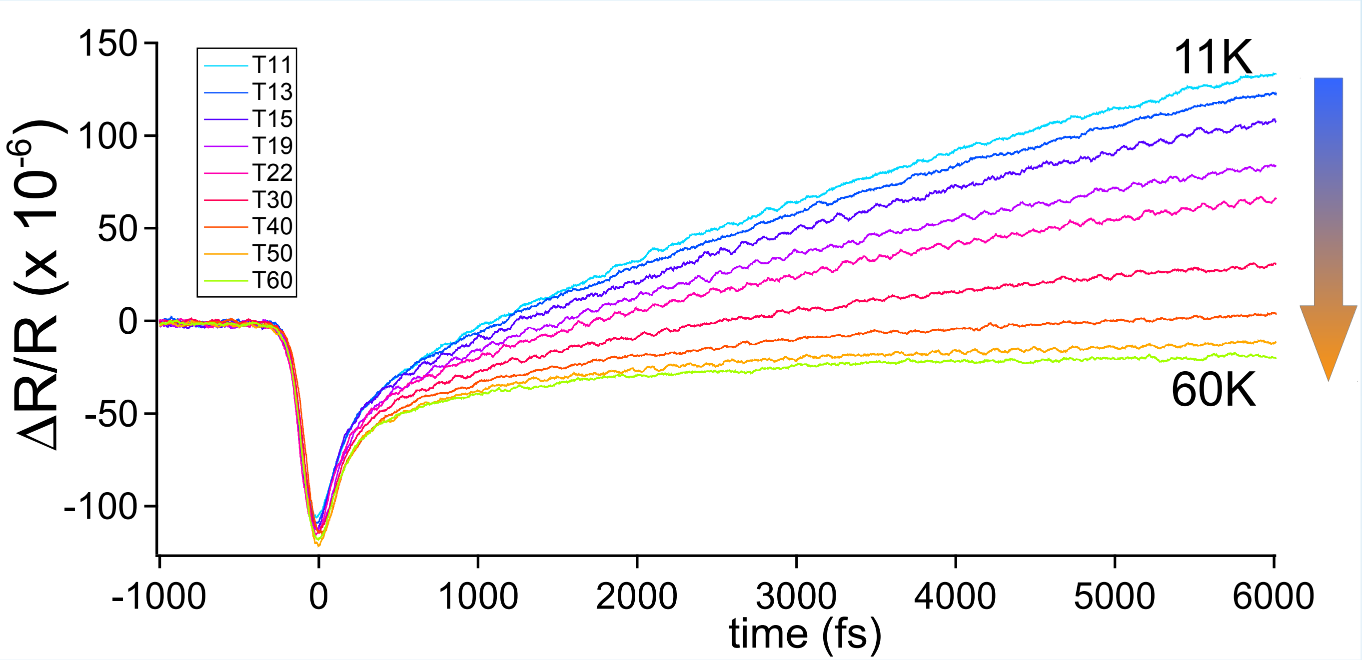
$$\frac{\Delta R}{R}(\omega,\tau) = \frac{R_{ex}(\omega,\tau) - R_{eq}(\omega)}{R_{eq}(\omega)}$$

Single color measurements as a function of T

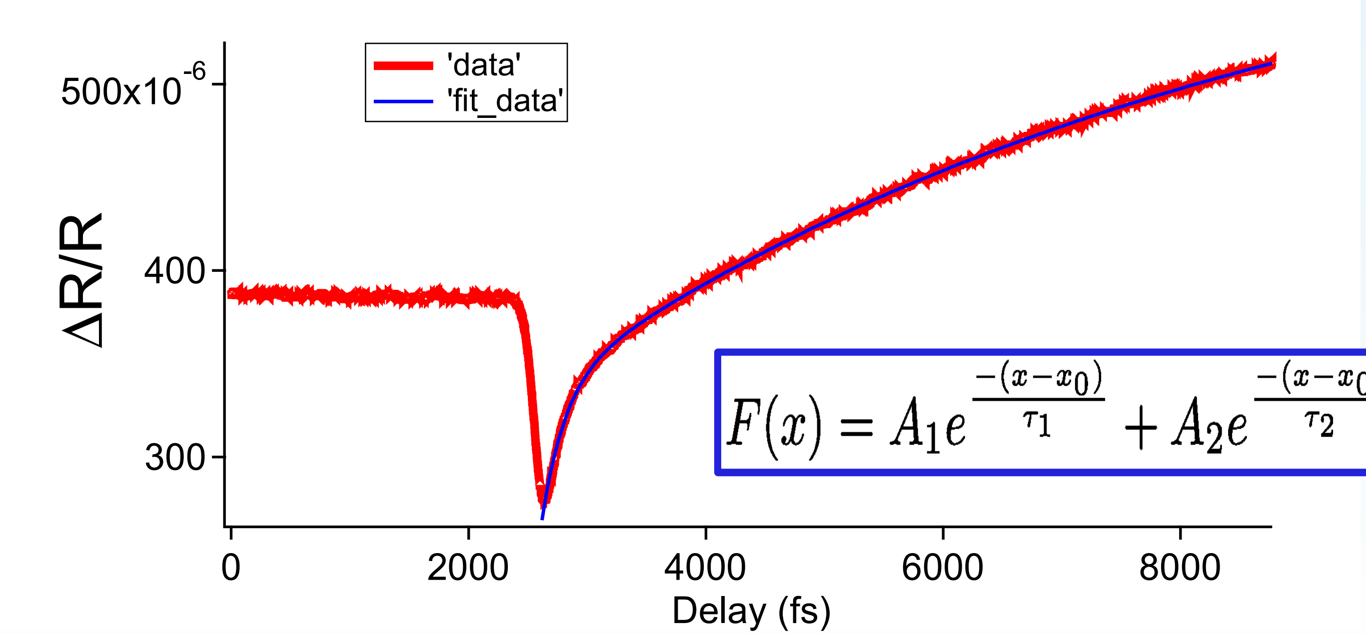


increasing temperature

Single color measurements as a function of T



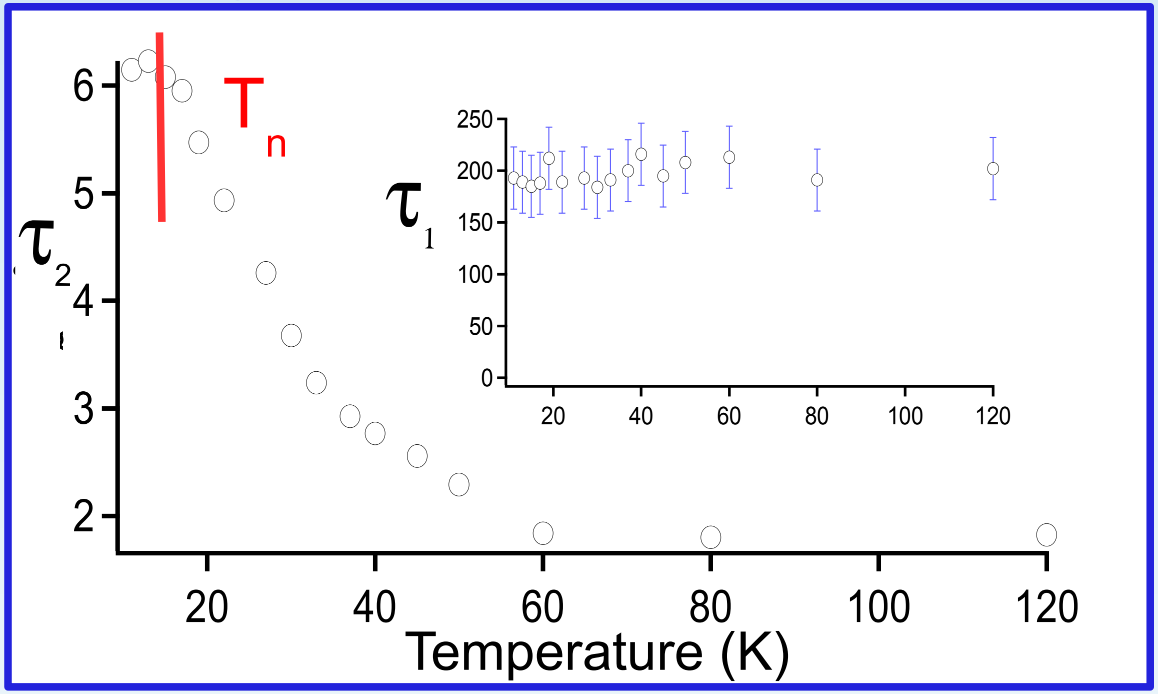
increasing temperature



$$F(x) = A_1 e^{\frac{-(x-x_0)}{\tau_1}} + A_2 e^{\frac{-(x-x_0)}{\tau_2}}$$

It is used a double exponential fit function

Single color measurements as a function of T



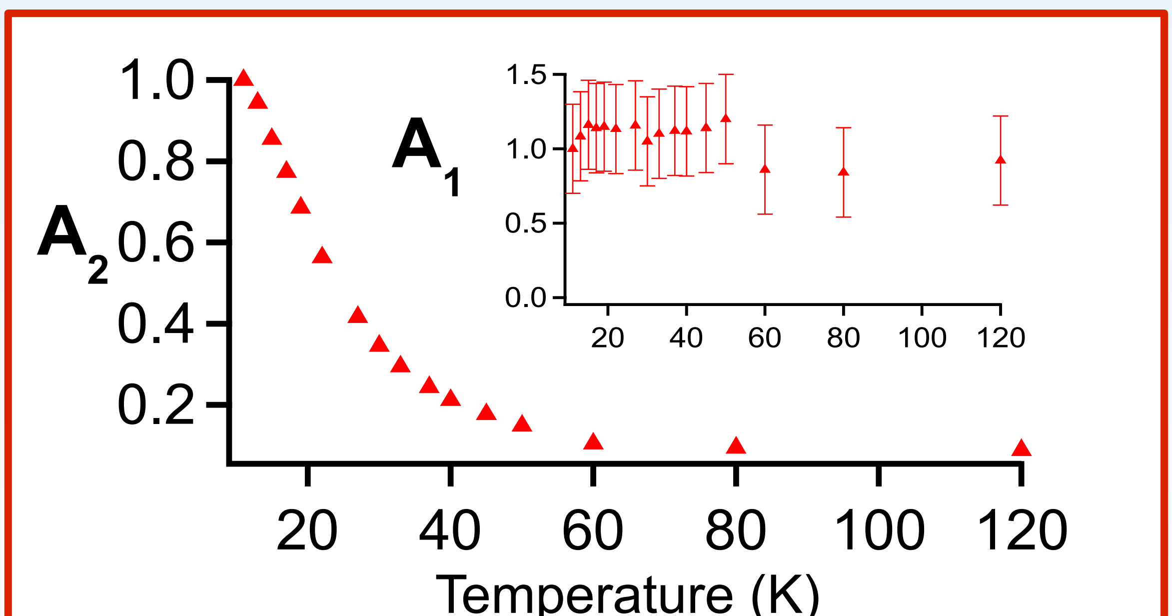
The fit's results evidences that on T onset:

τ_1 ~ constant at 200 fs

A_1 constant

τ_2 variation 6→2 ps

A_2 diverges at T_n



The experiment shows a temperature dipendence and a divergence at T_n

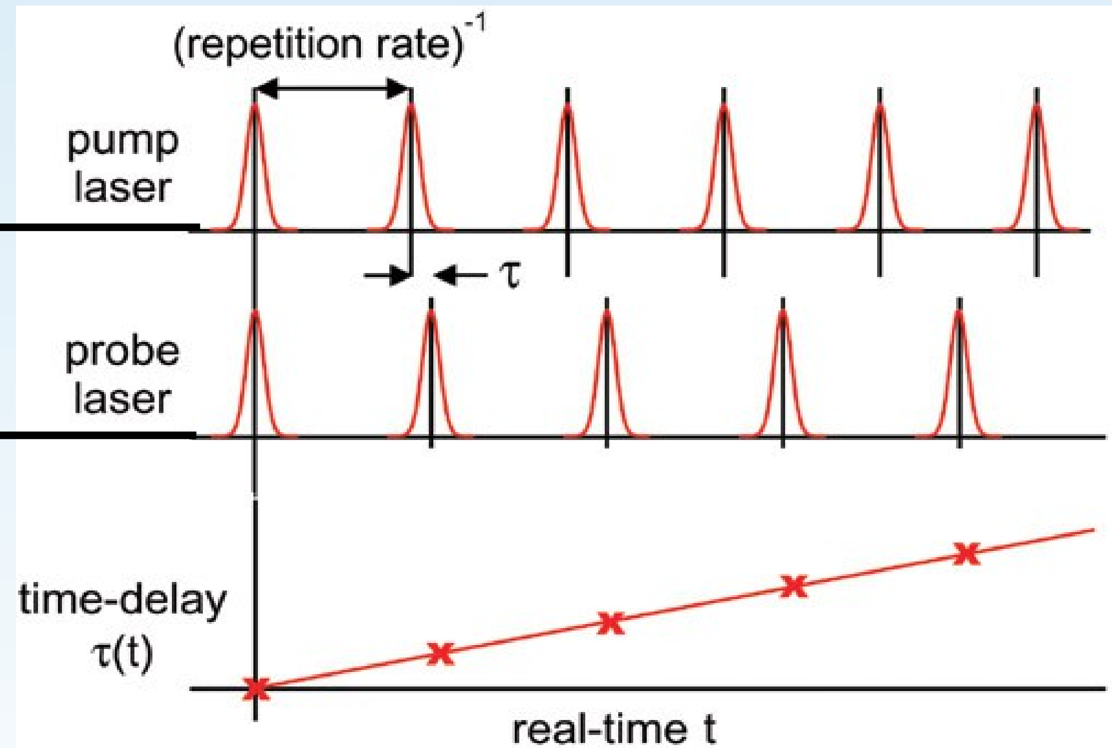
The dynamics on ns scale?

ASOPS tecnique

To achieve long-time windows (~10 *ns*) without losing the spatial coincidence is used ASOPS tecnique

f=100 MHz+ Δf

f=100 MHz



Properties:

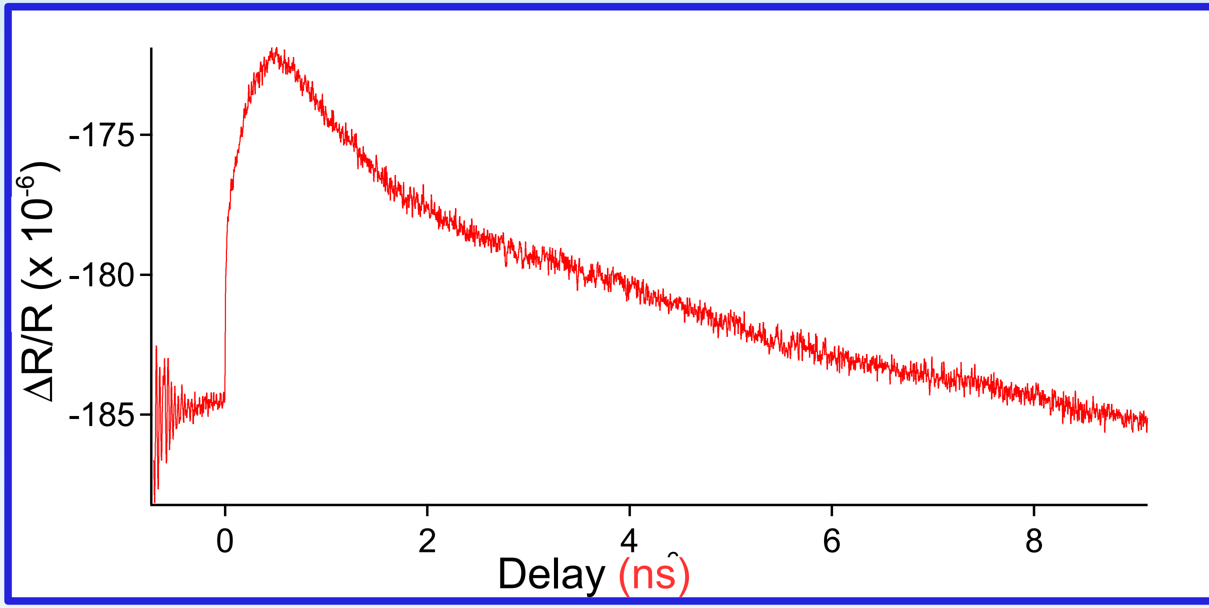
- without delay-line
- Dynamincs of several *ns*

ASynchronous OPtical Sampling tecnique

ASOPS measurements

Difference between single color mesurements:

- Pump pulse energy is 0.7 eV
- Time windows of ns
- Lower time resolution (100 fs)



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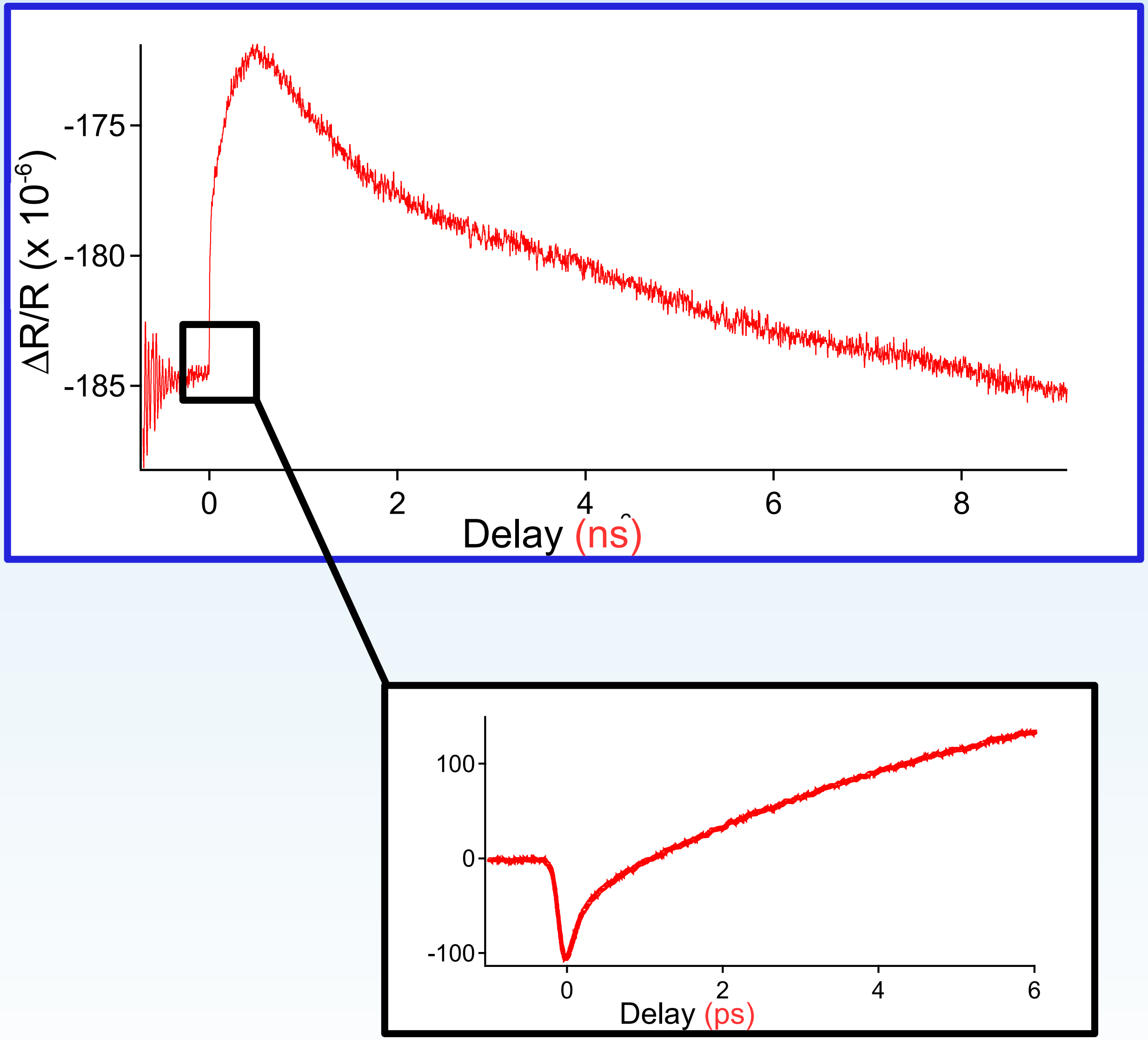

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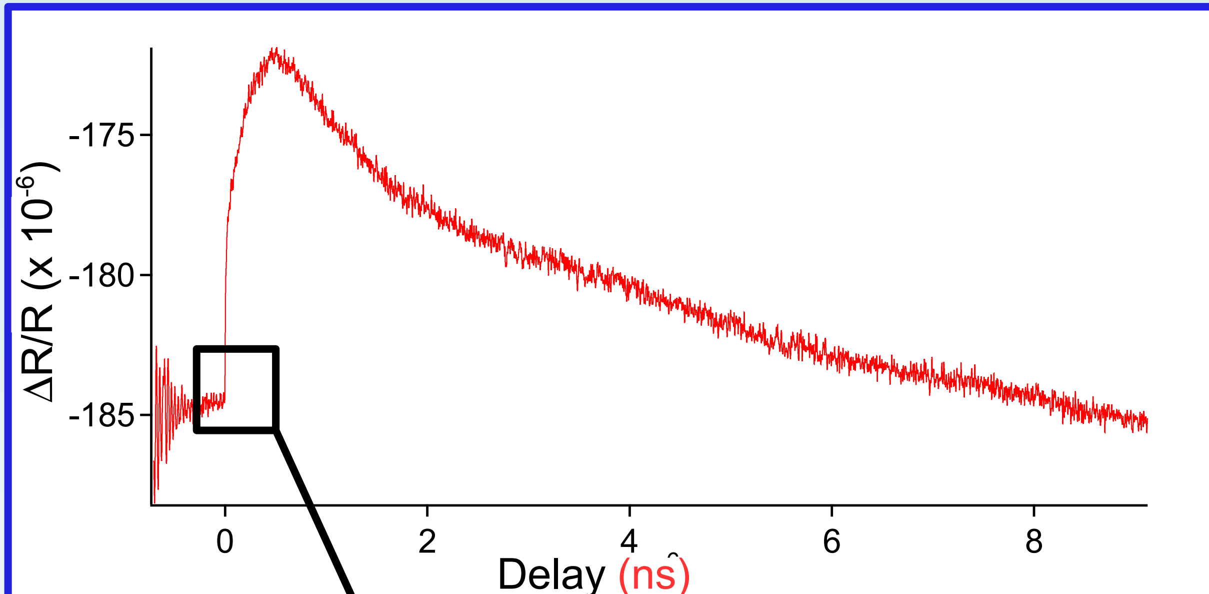
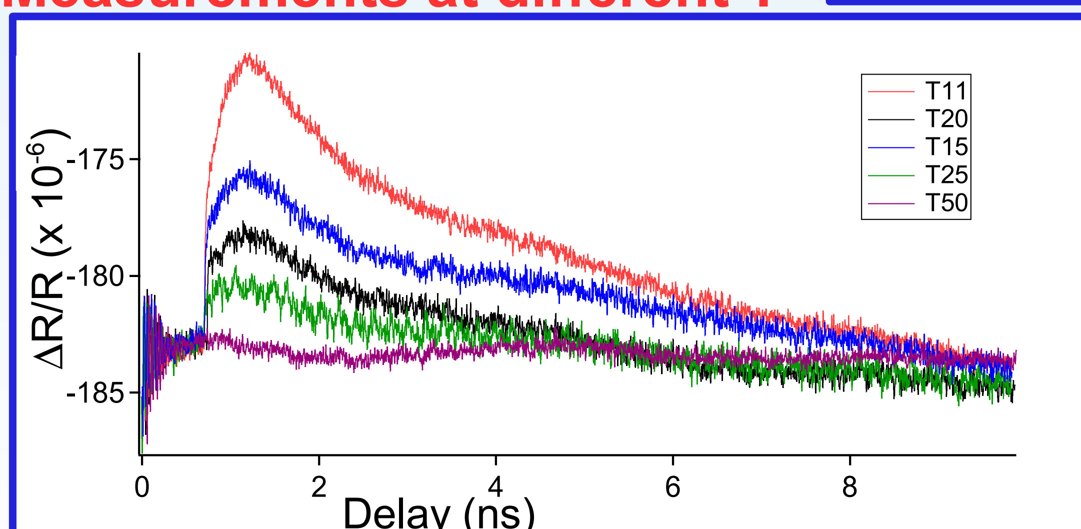
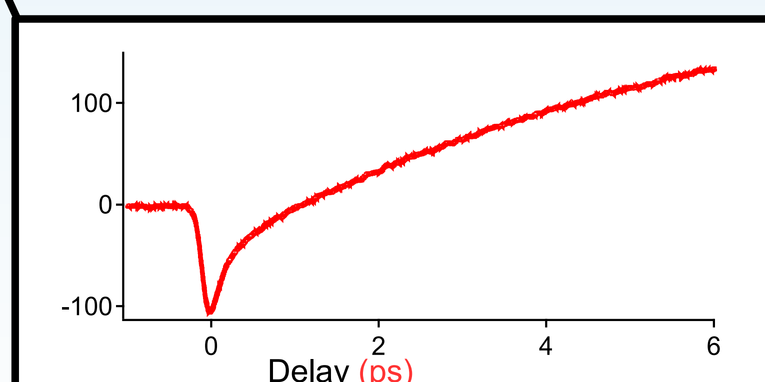
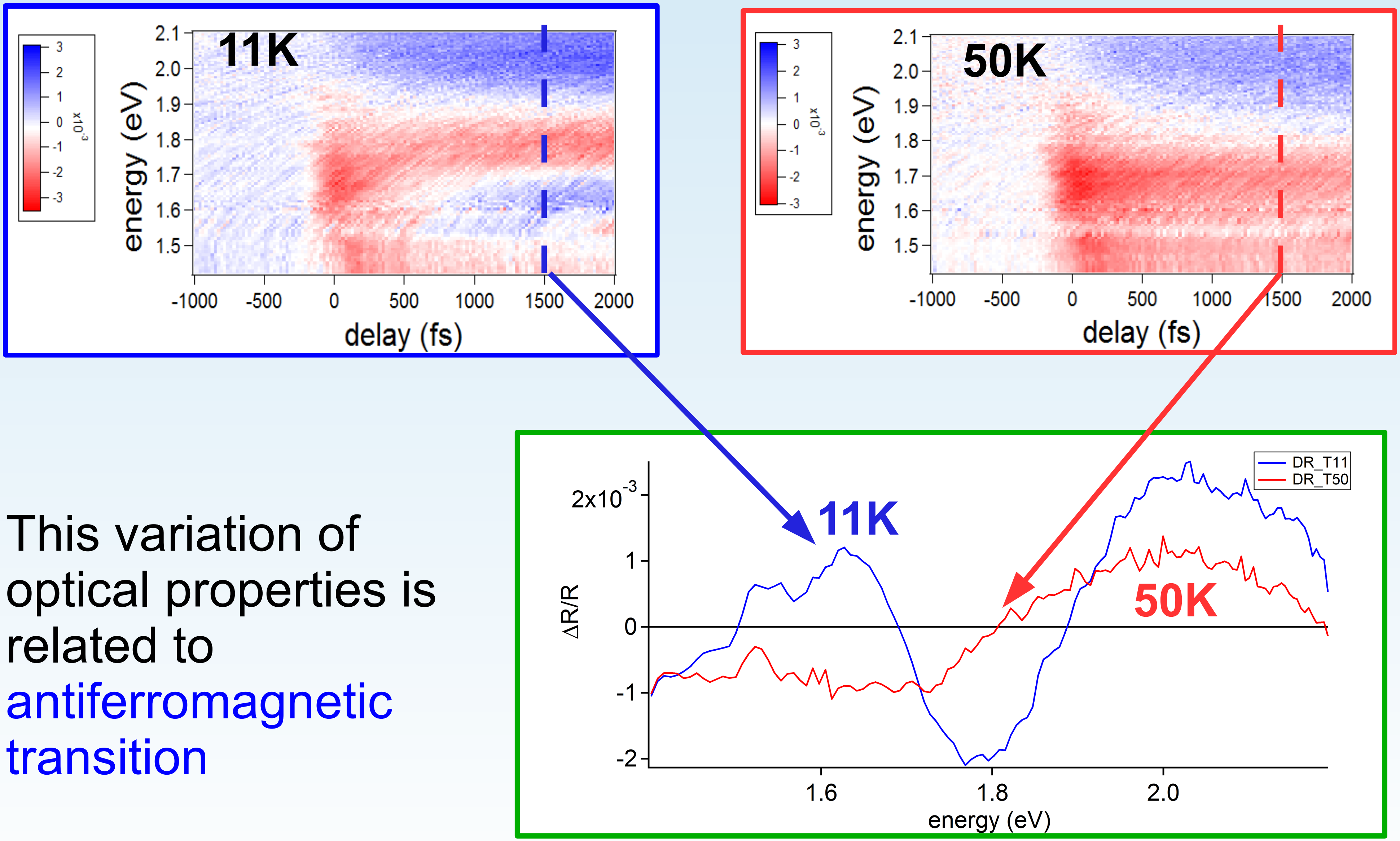


Measurements at different T





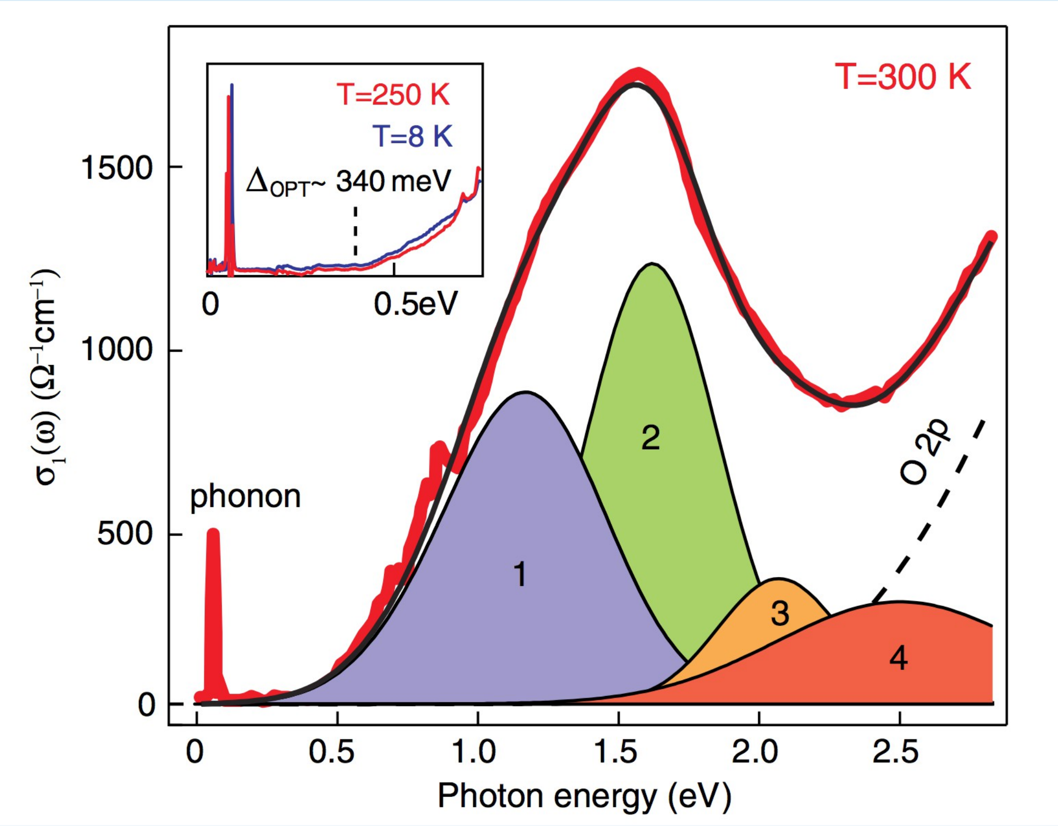
Supercontinuum measurements



This variation of optical properties is related to antiferromagnetic transition

Preliminary results and open problems

Differential fit to describe the variation of reflectivity



Optical properties at equilibrium (condictivity) with the Lorentz oscillators.

Lorentz oscillator

Oscillator weight

$$\epsilon(\omega) = \epsilon_\infty + \sum_j \frac{\omega_{pj}^2}{(\omega_{0j}^2 - \omega^2) - i\Gamma_j\omega}$$

Oscillator position

Oscillator width

Is possible to reproduce DR/R?

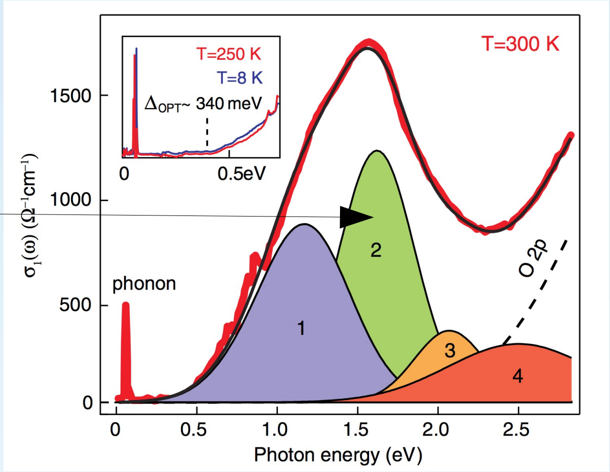
What are the parameter that changes?

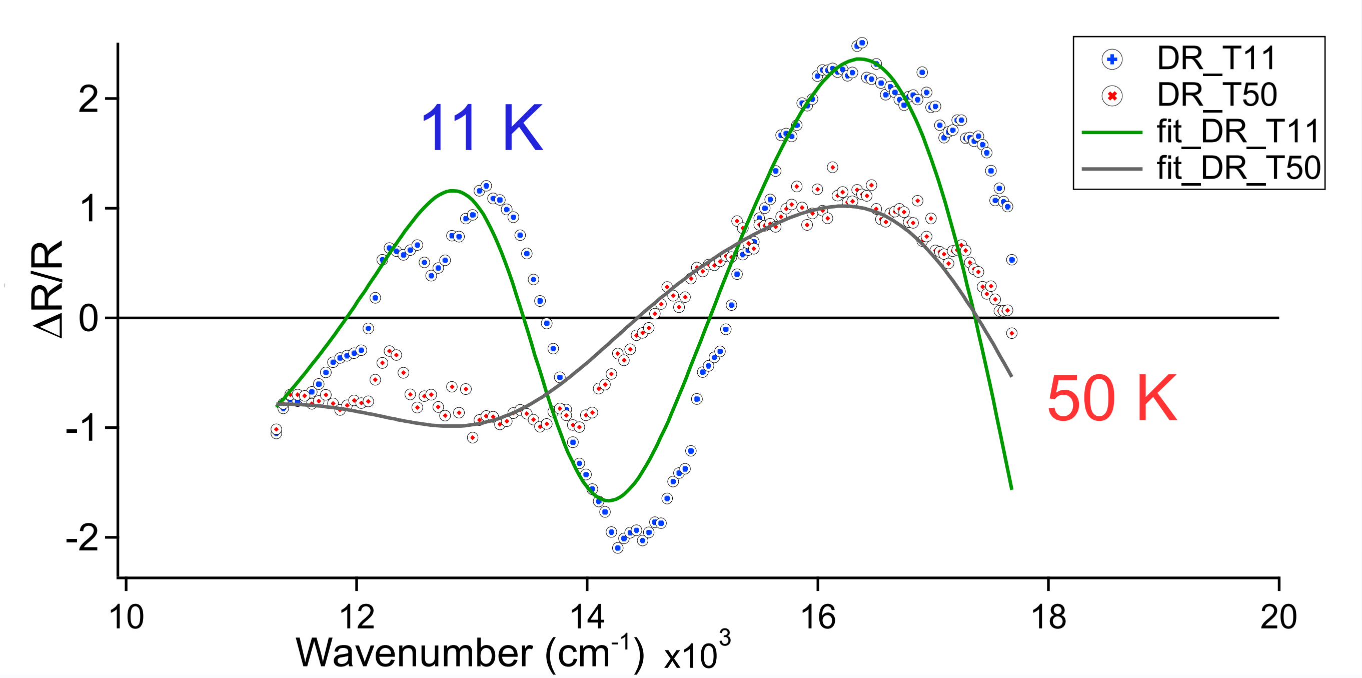
Preliminary results and open problems

Differential fit to describe the variation of riflectivity

Measure at 50 K → change ε_∞ and spectral weight of oscillator at 1.6 eV

Measure at 11 K → change ε_∞ spectral weight and position of oscillator at 1.6 eV





What does it mean?

Conclusions

- time-resolved optical spectroscopy is possible to decopule the different energy scales
- Variation of optical properties near Antiferromagnetic transition

In future:

Try to understand

What are the involved parameters?

Why these oscillators energy?

Thanks for yours attention!