

Time Resolved Optical Spectroscopy of Na_2IrO_3

Nicola Nembrini

What is Na_2IrO_3 ?

**Periodic Table
of the
Elements**

1 H 1.01																	18 He 4.00														
3 Li 6.94	4 Be 9.01											13 B 10.81	14 C 12.01	15 N 14.01	16 O 16.00	17 F 19.00	10 Ne 20.18														
11 Na 22.99	12 Mg 24.30											13 Al 26.98	14 Si 28.09	15 P 30.97	16 S 32.07	17 Cl 35.45	18 Ar 39.95														
19 K 39.10	20 Ca 40.08	21 Sc 44.96	22 Ti 47.88	23 V 50.94	24 Cr 52.00	25 Mn 54.94	26 Fe 55.85	27 Co 58.93	28 Ni 58.69	29 Cu 63.55	30 Zn 65.39	31 Ga 69.72	32 Ge 72.61	33 As 74.92	34 Se 78.96	35 Br 79.90	36 Kr 83.80														
37 Rb 85.47	38 Sr 87.62	39 Y 88.91	40 Zr 91.22	41 Nb 92.91	42 Mo 95.94	43 Tc (97.91)	44 Ru 101.07	45 Rh 102.91	46 Pd 106.42	47 Ag 107.87	48 Cd 112.41	49 In 114.82	50 Sn 118.71	51 Sb 121.75	52 Te 127.60	53 I 126.90	54 Xe 131.29														
55 Cs 132.91	56 Ba 137.33	57 La 138.91	72 Hf 178.49	73 Ta 180.95	74 W 183.85	75 Re 186.21	76 Os 190.23	77 Ir 192.22	78 Pt 195.08	79 Au 196.97	80 Hg 200.59	81 Tl 204.38	82 Pb 207.2	83 Bi 208.98	84 Po (208.98)	85 At (209.99)	86 Rn (222.02)														
87 Fr (223.02)	88 Ra (226.03)	89 Ac (227.03)	104 Rf (261.11)	105 Ha (262.11)	106 Sg (263.12)																										
																		58 Ce 140.12	59 Pr 140.91	60 Nd 144.24	61 Pm (144.91)	62 Sm 150.36	63 Eu 151.97	64 Gd 157.25	65 Tb 158.93	66 Dy 162.50	67 Ho 164.93	68 Er 167.26	69 Tm 168.93	70 Yb 173.04	71 Lu 174.97
																		90 Th 232.04	91 Pa 231.04	92 U 238.03	93 Np (237.05)	94 Pu (244.06)	95 Am (243.06)	96 Cm (247.07)	97 Bk (247.07)	98 Cf (251.08)	99 Es (252.08)	100 Fm (257.10)	101 Md (258.10)	102 No (259.10)	103 Lr (262.11)

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

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

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77
Ir
Iridio
192,22
[Xe] 4f ¹⁴ 5d ⁷ 6s ²

Na_2IrO_3 is **5d metal oxides**:

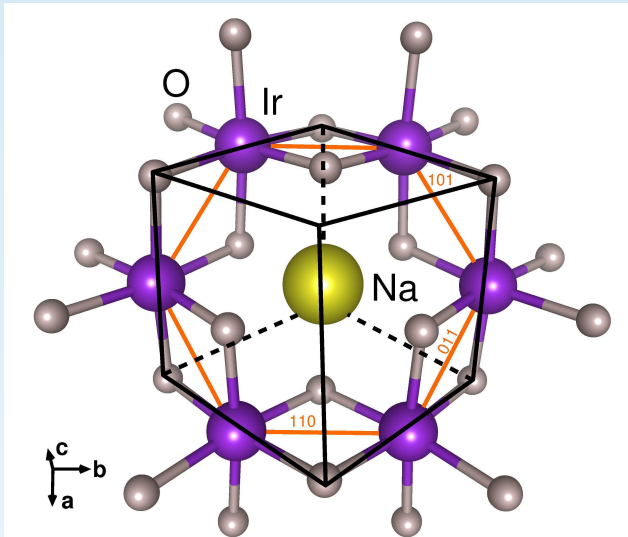
- More delocalized orbitals (5d) $\longrightarrow U \sim 1-2$ eV

- large weight ($SO \sim 1-2$ eV)

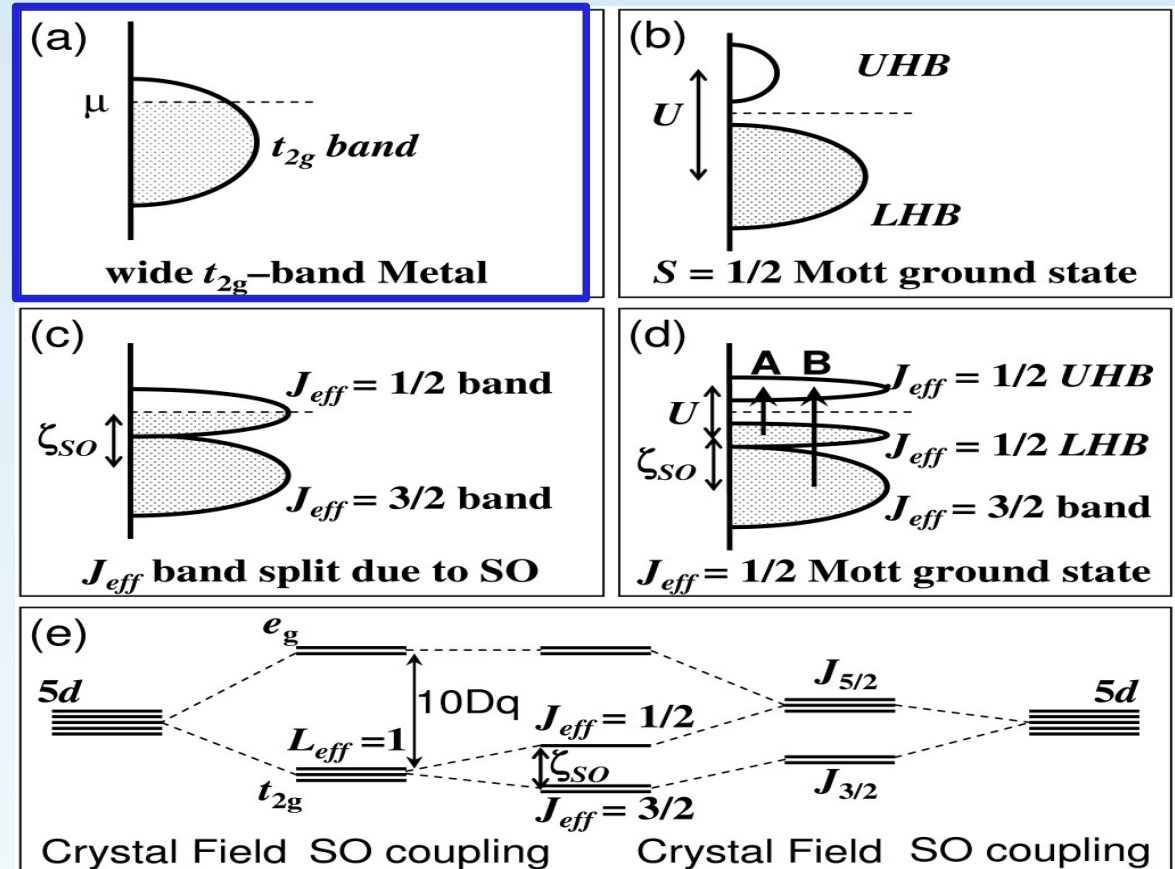
$$H_{s.o.} = \frac{1}{2m^2c^2r} (\hat{S} \cdot \hat{L}) \frac{dV}{dr} \longrightarrow \boxed{Z}$$

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

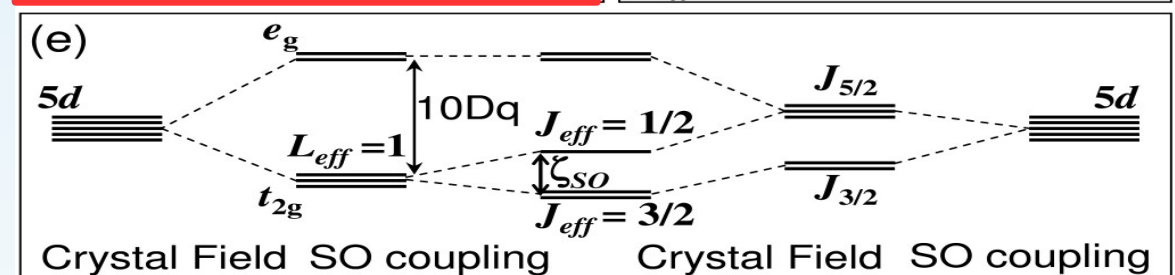
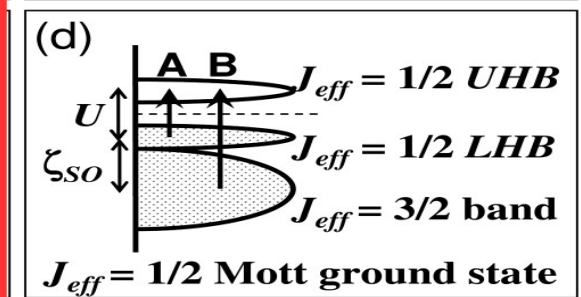
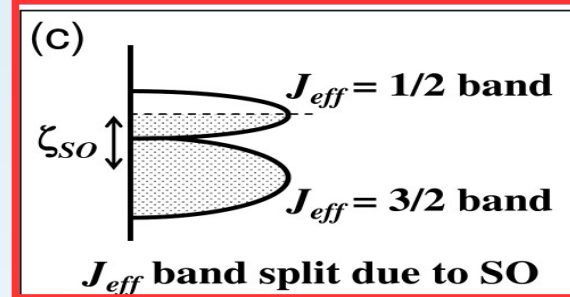
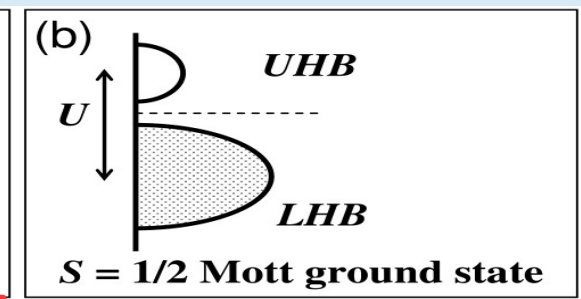
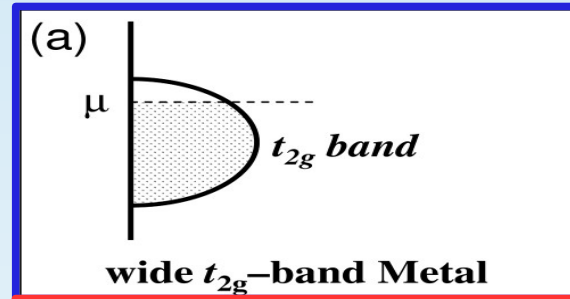
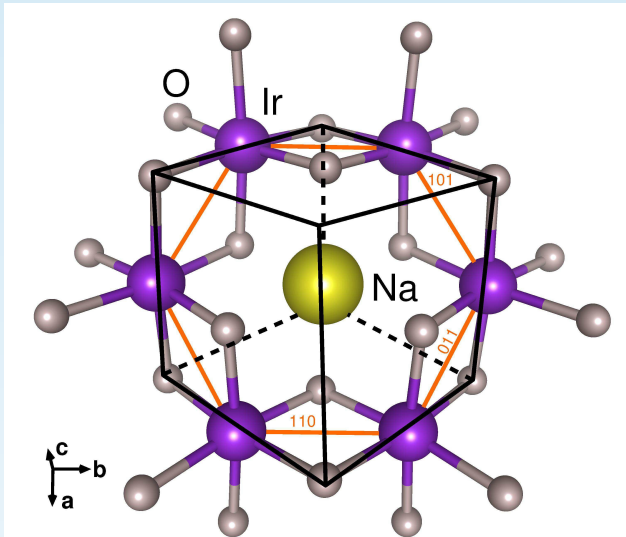
Properties of Na_2IrO_3 at equilibrium



- Crystal field
- Spin orbit coupling
- Coulomb repulsion



Properties of Na_2IrO_3 at equilibrium

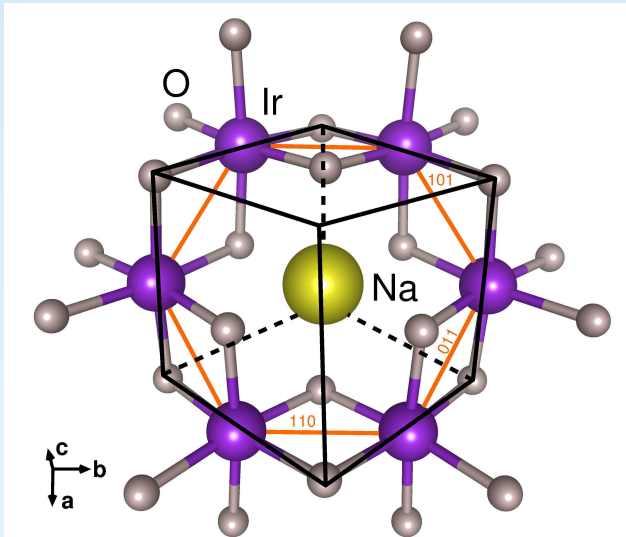


- Crystal field

- Spin orbit coupling

- Coulomb repulsion

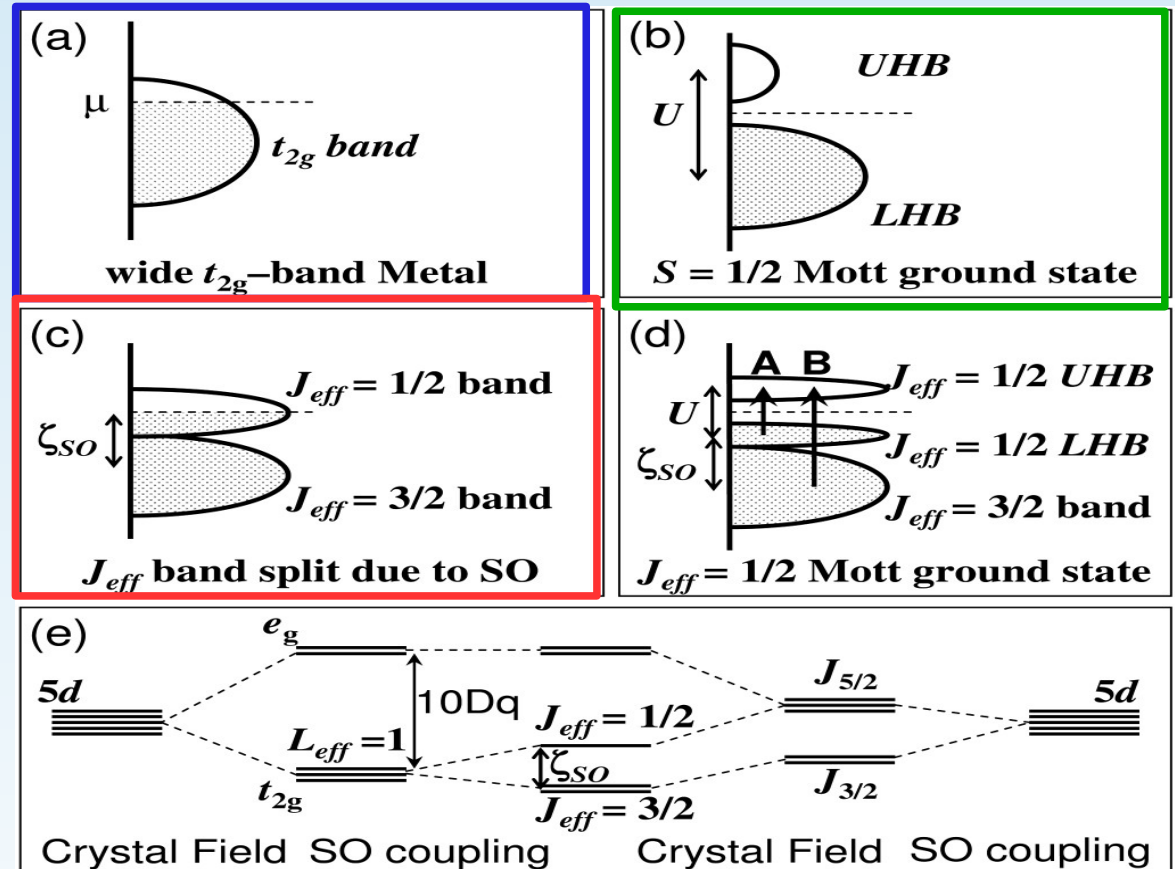
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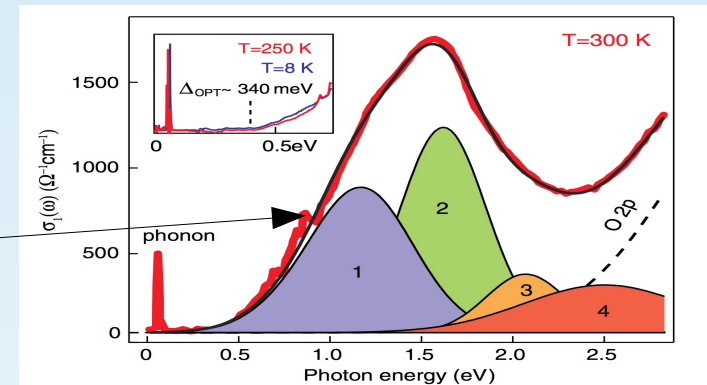
Electronic band structure?

Physical properties of Na_2IrO_3 at equilibrium

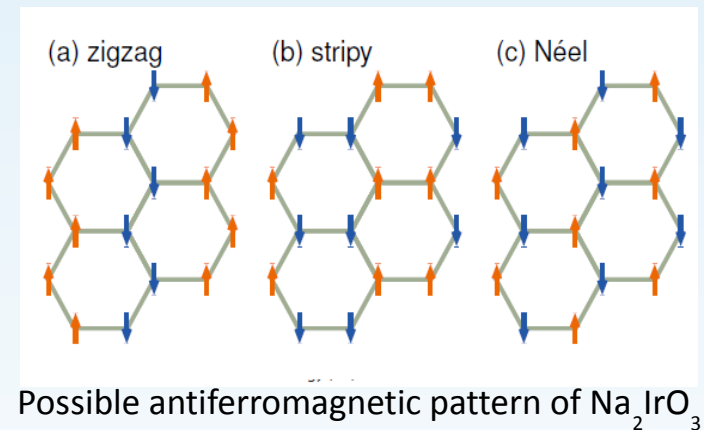
- At $T = 300\text{K}$ is a **Mott Insulator** (340meV)

Optical conductivity data (red line)

[R. Comin, A. Damascelli et al.]



- At $T = 15\text{K}$ present an **antiferromagnetic order**



How W , SO and U determine these properties?

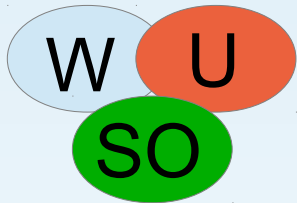
Time-resolved optical spectroscopy

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

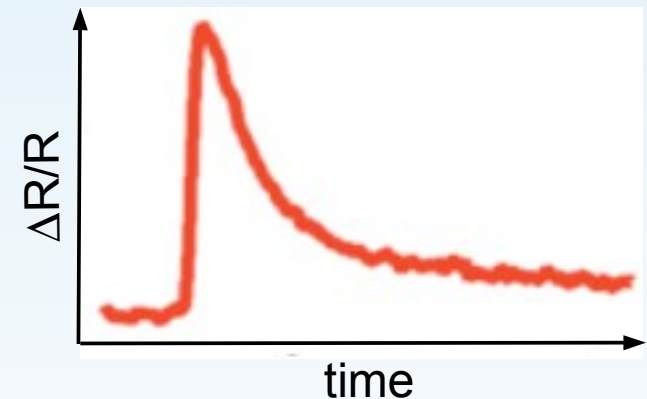
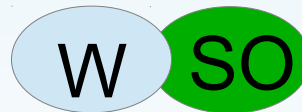
At equilibrium comparable scale of energy



Typically in time scale

$$U \ll SO \text{ and } W$$

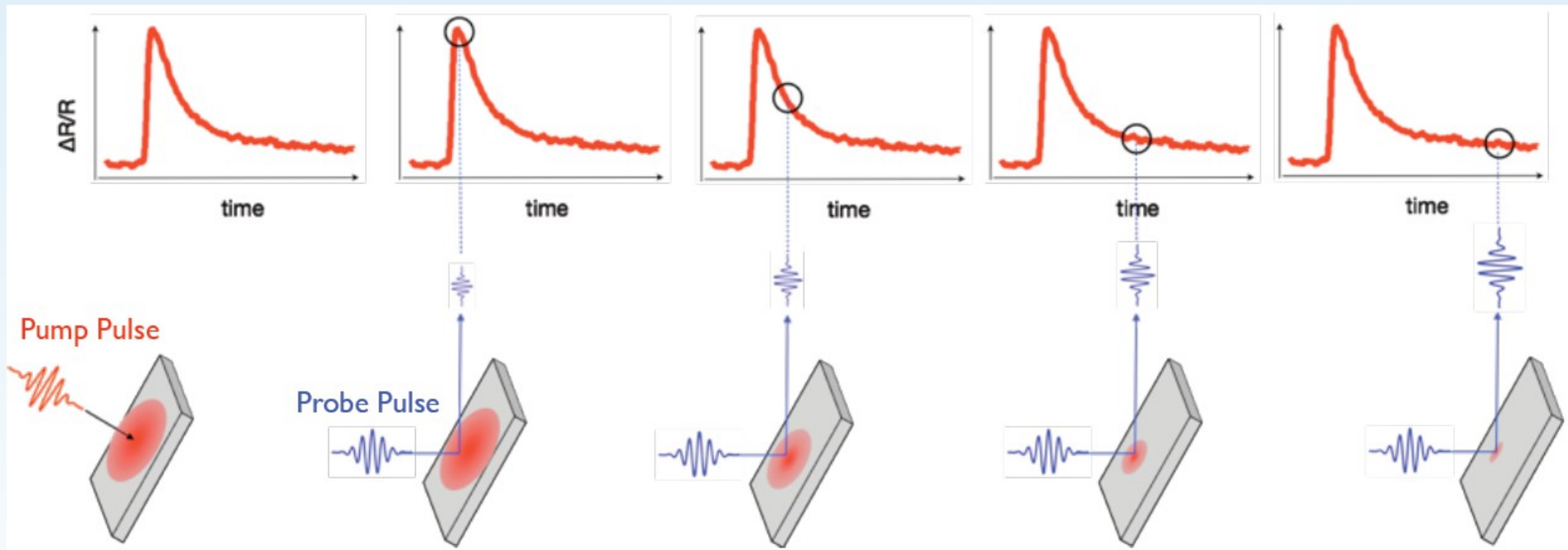
Out of equilibrium: different time scale



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

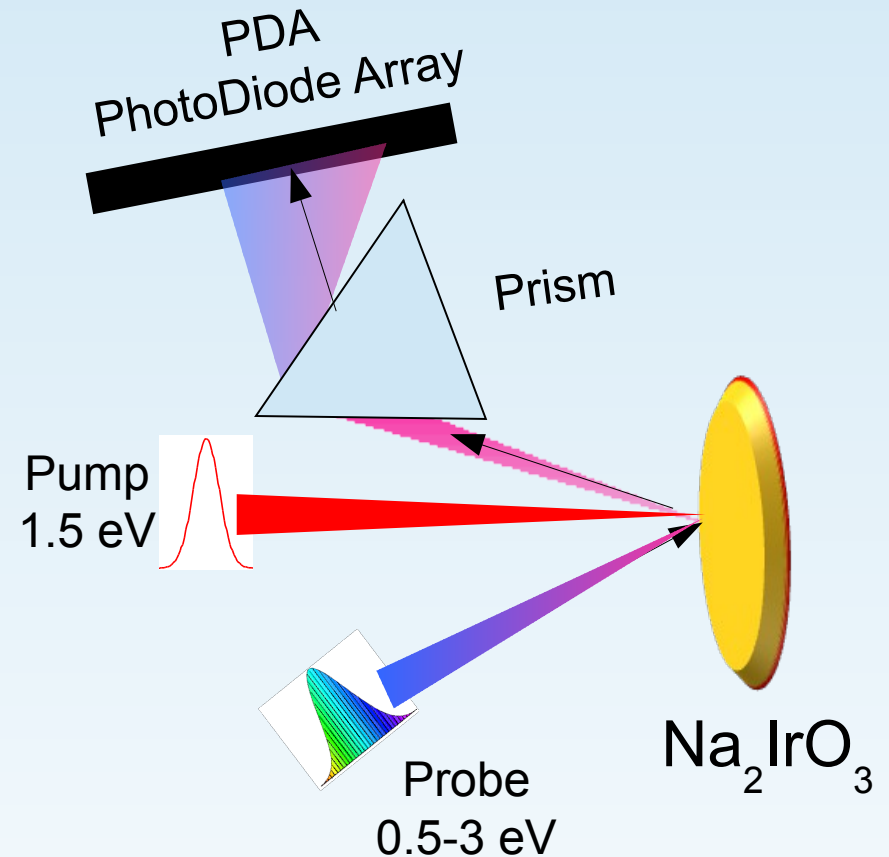
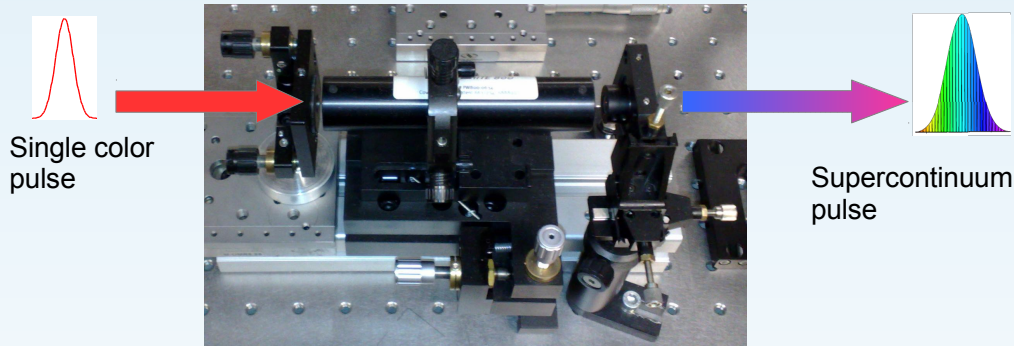


- Investigate dynamics of several ps (~ 10 ps)
- High time resolution of fs (~ 100 fs)

Time resolved spectroscopy Pump & Probe

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

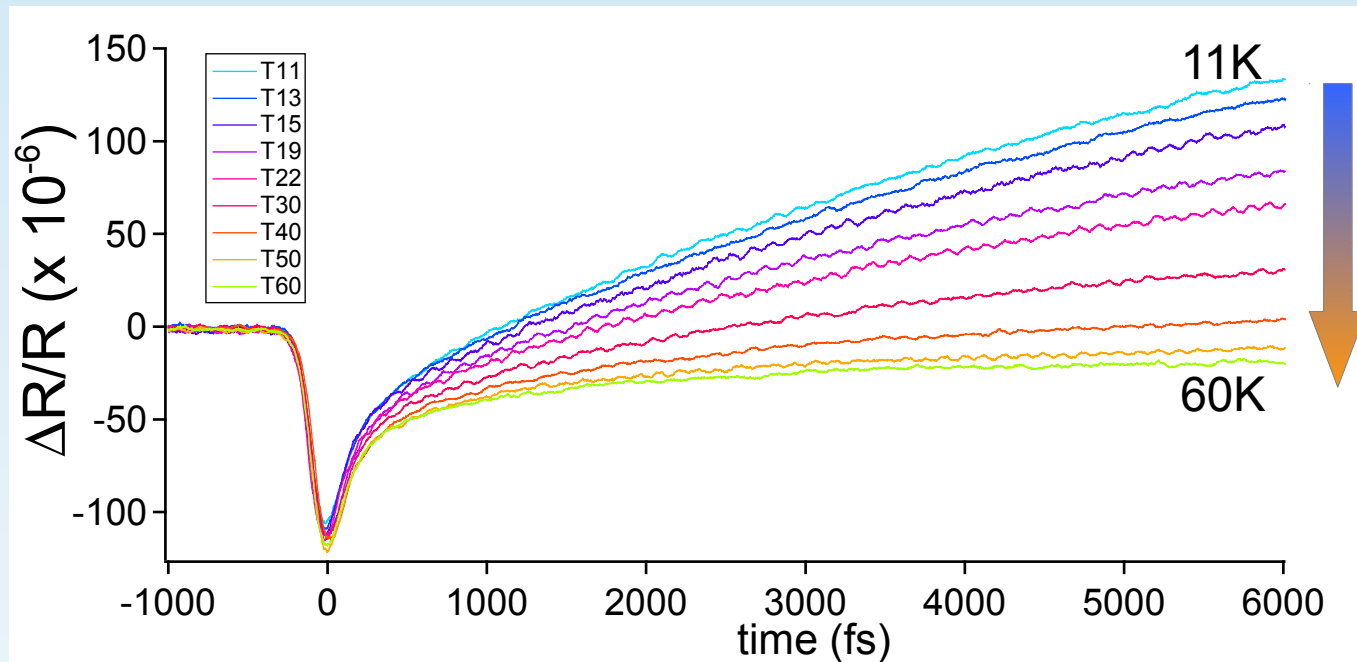
- Time-resolved measurements
- Frequency-resolved measurements



Experimental $\Delta R/R$

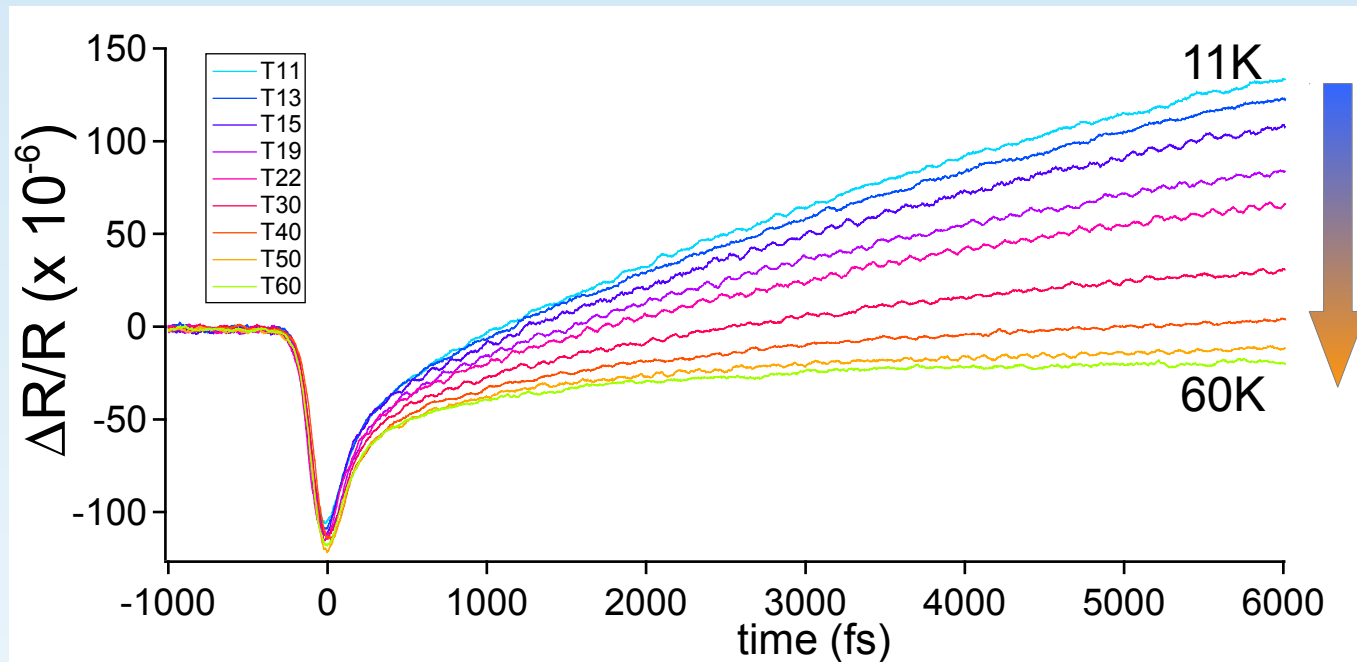
$$\frac{\Delta R}{R}(\omega, \tau) = \frac{R_{ex}(\omega, \tau) - R_{eq}(\omega)}{R_{eq}(\omega)}$$

Single color measurements as a function of T

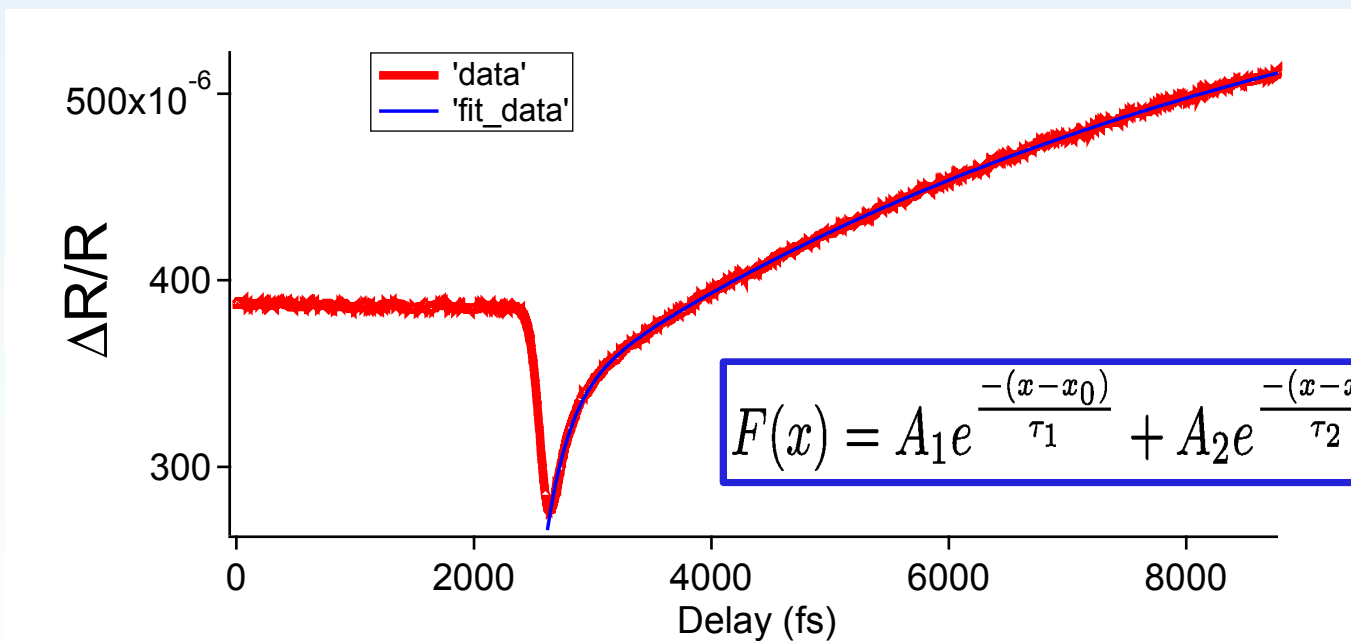


increasing temperature

Single color measurements as a function of T

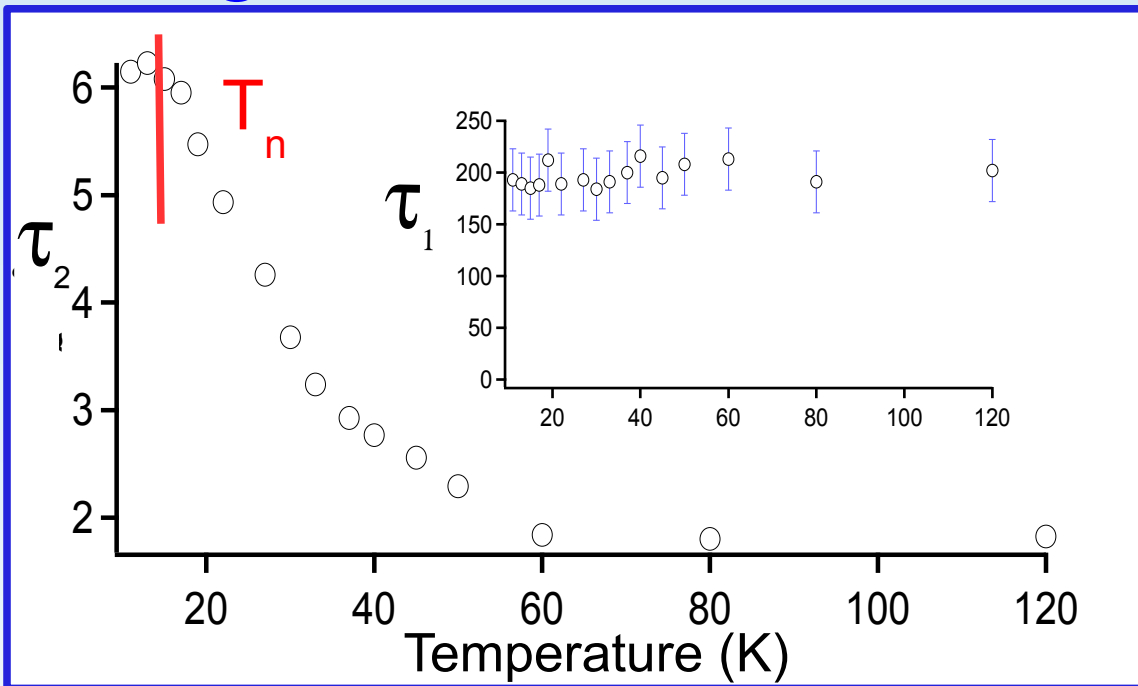


increasing temperature



It is used a double exponential fit function

Single color measurements as a function of T



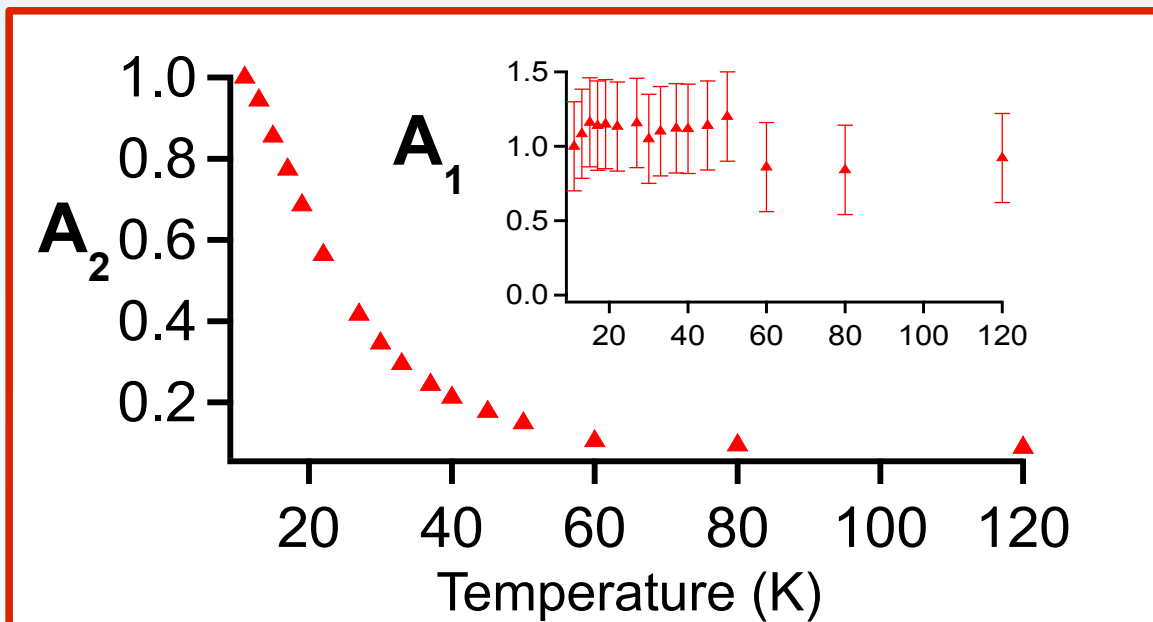
The fit's results evidences that on T onset:

$\tau_1 \sim$ constant at 200 fs

A_1 constant

τ_2 variation 6 \rightarrow 2 ps

A_2 diverges at T_n

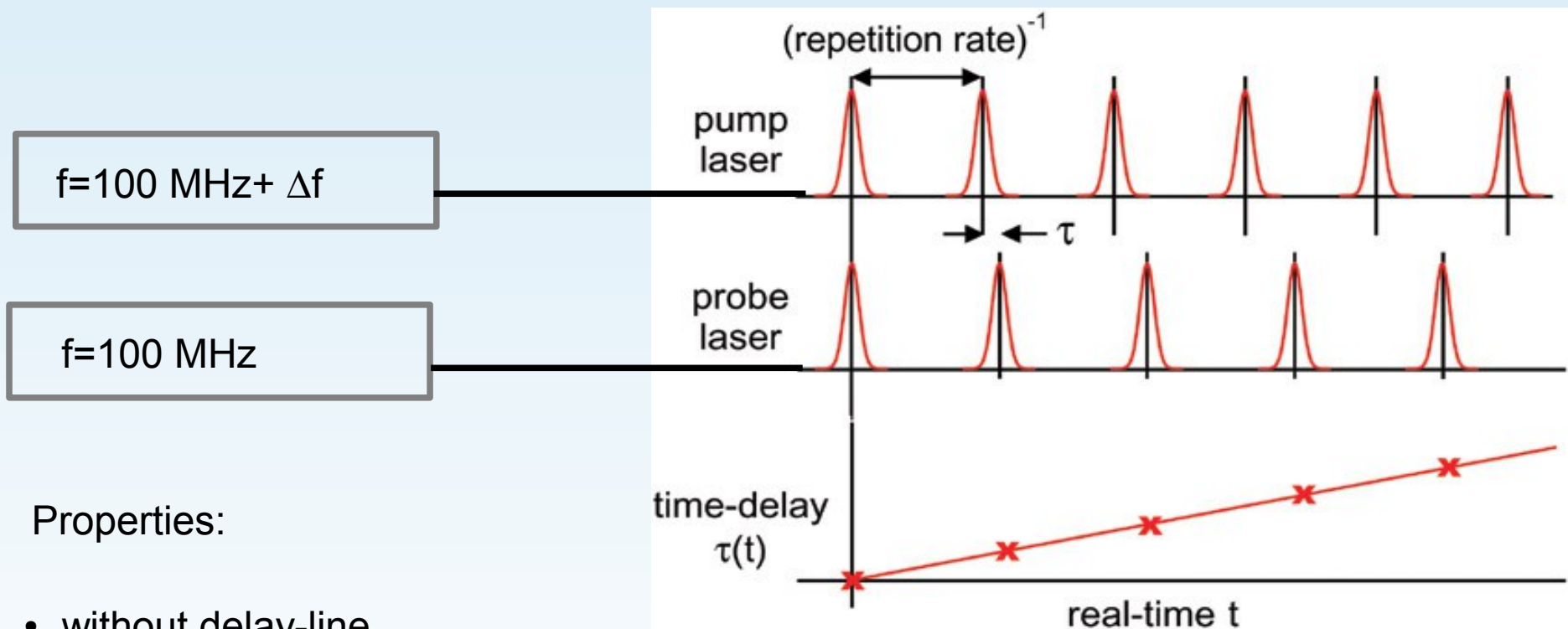


The experiment shows a temperature dependence and a divergence at T_n

The dynamics on ns scale?

ASOPS technique

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



$f=100 \text{ MHz} + \Delta f$

$f=100 \text{ MHz}$

Properties:

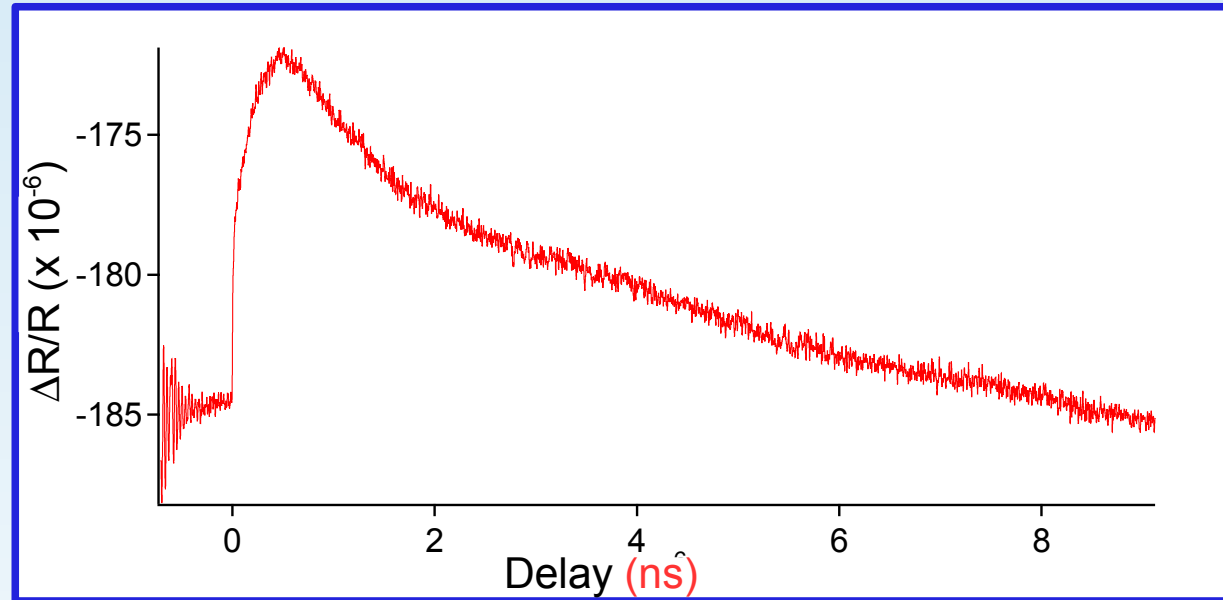
- without delay-line
- Dynamics of several ns

ASynchronous **OP**tical **S**ampling technique

ASOPS measurements

Difference between single color measurements:

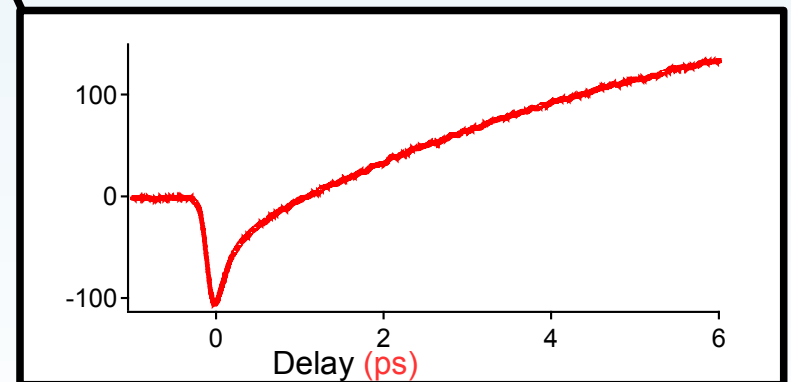
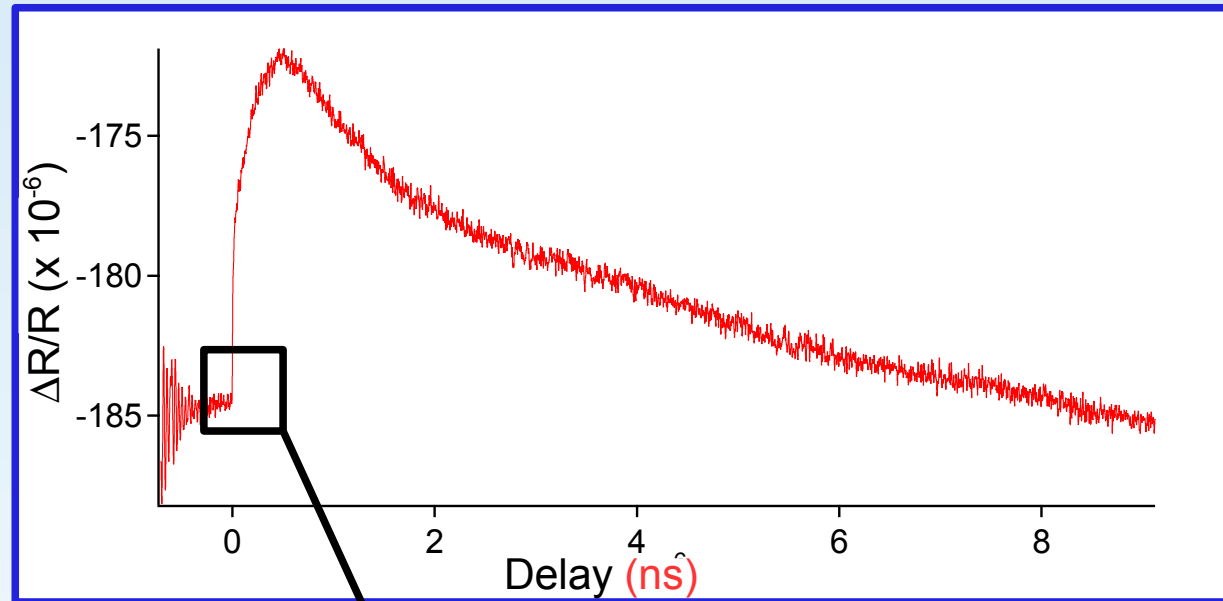
- Pump pulse energy is 0.7 eV
- Time windows of ns
- Lower time resolution (100 fs)



ASOPS measurements

Difference between single color measurements:

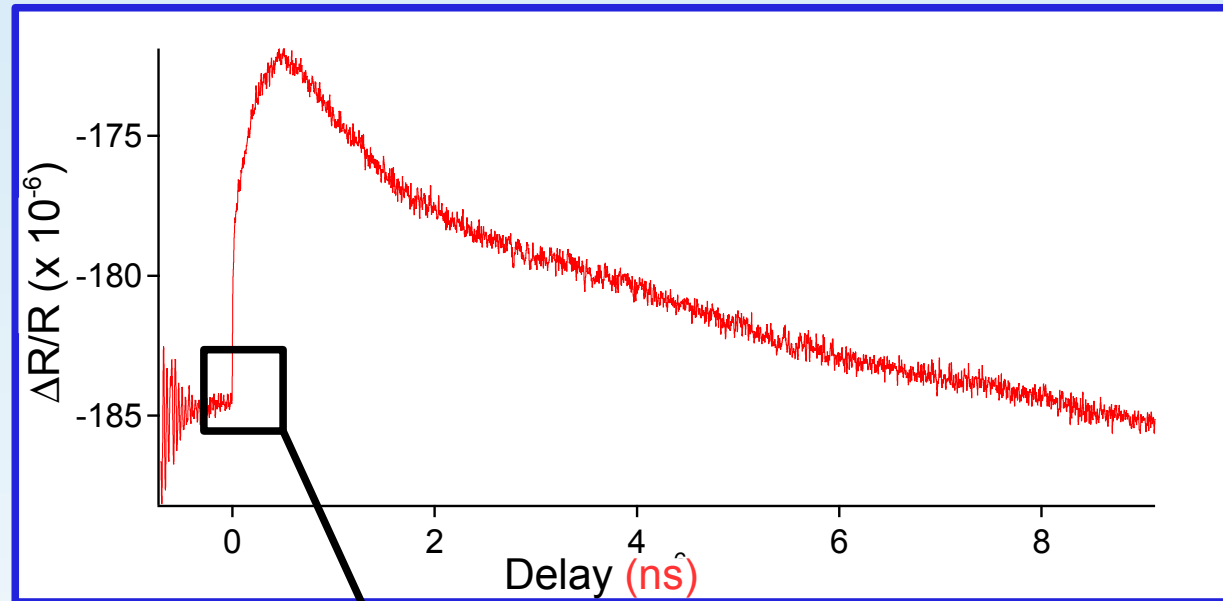
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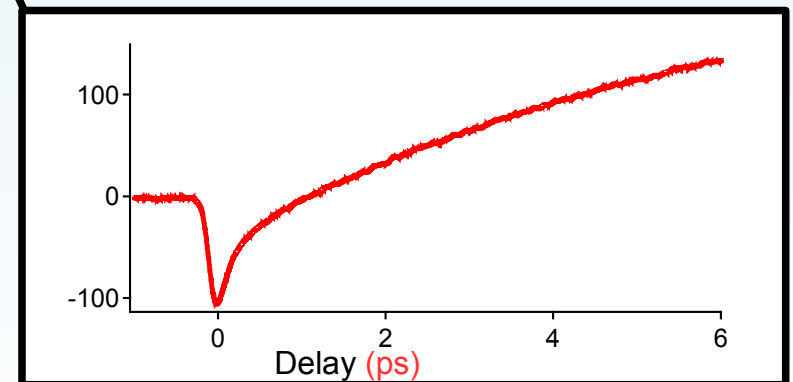
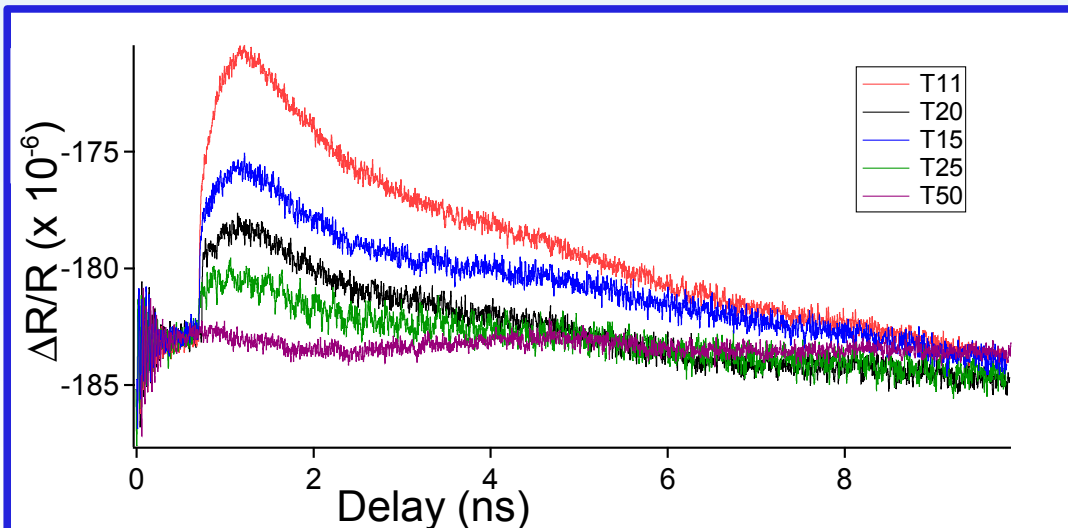
ASOPS measurements

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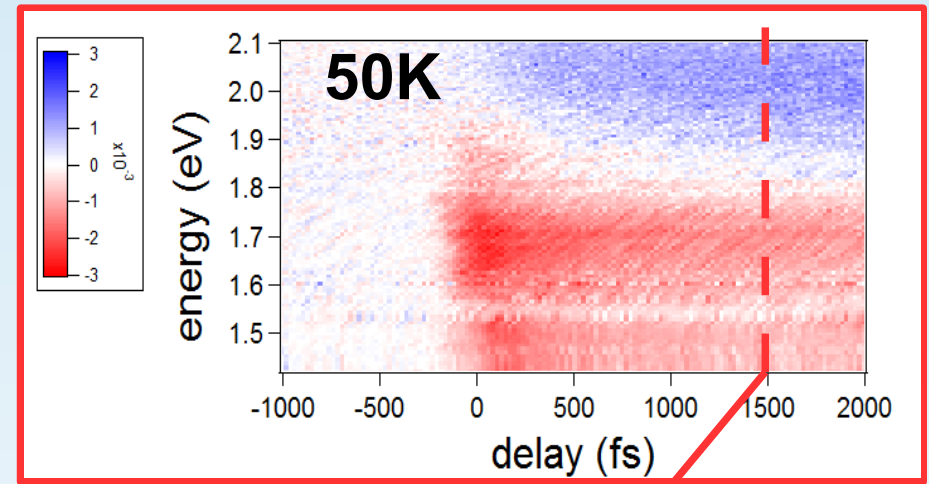
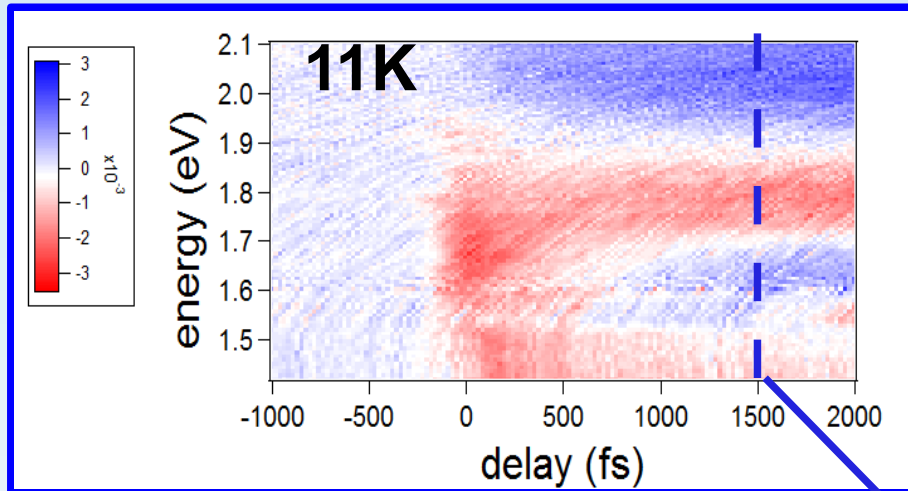
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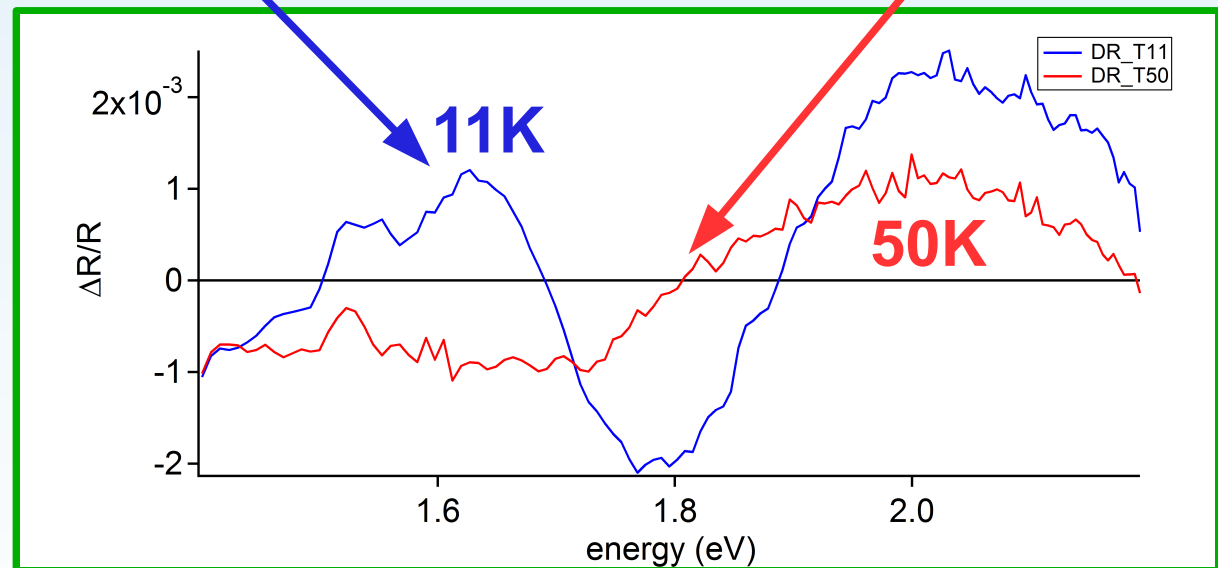
Measurements at different T



Supercontinuum measurements

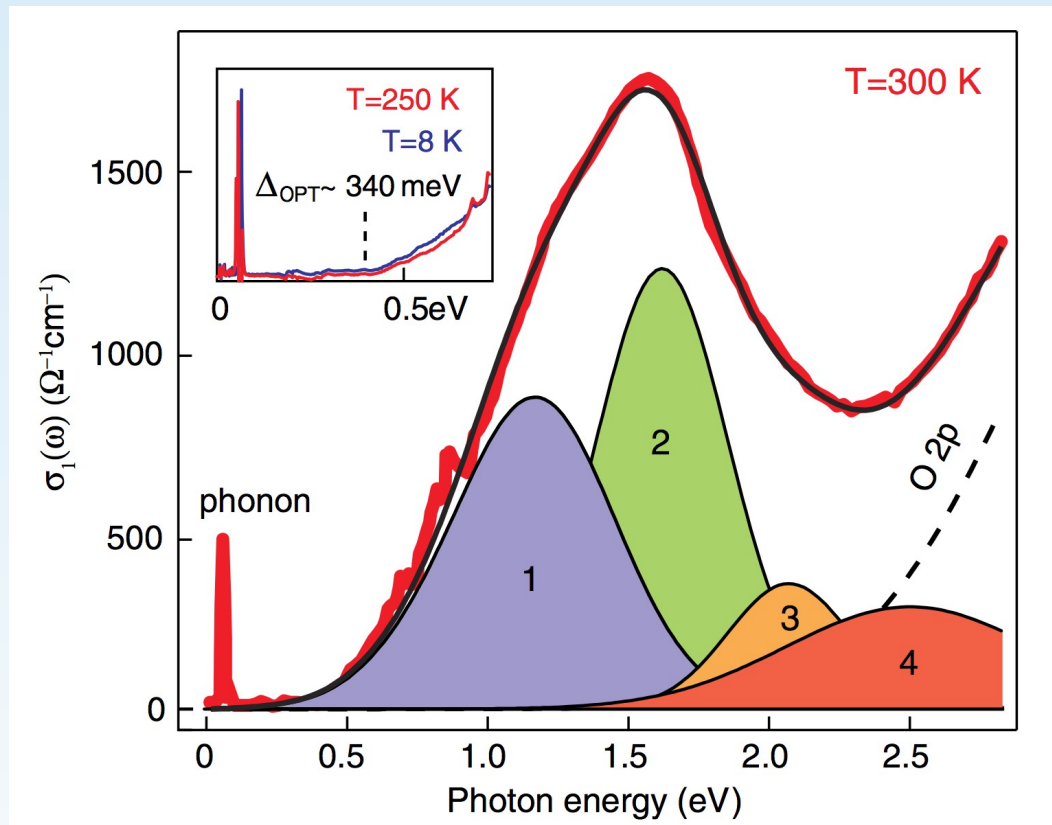


This variation of optical properties is related to antiferromagnetic transition



Preliminary results and open problems

Differential fit to describe the variation of reflectivity



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

Lorentz oscillator

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

Oscillator weight

Oscillator position

Oscillator width

Is possible to reproduce DR/R?

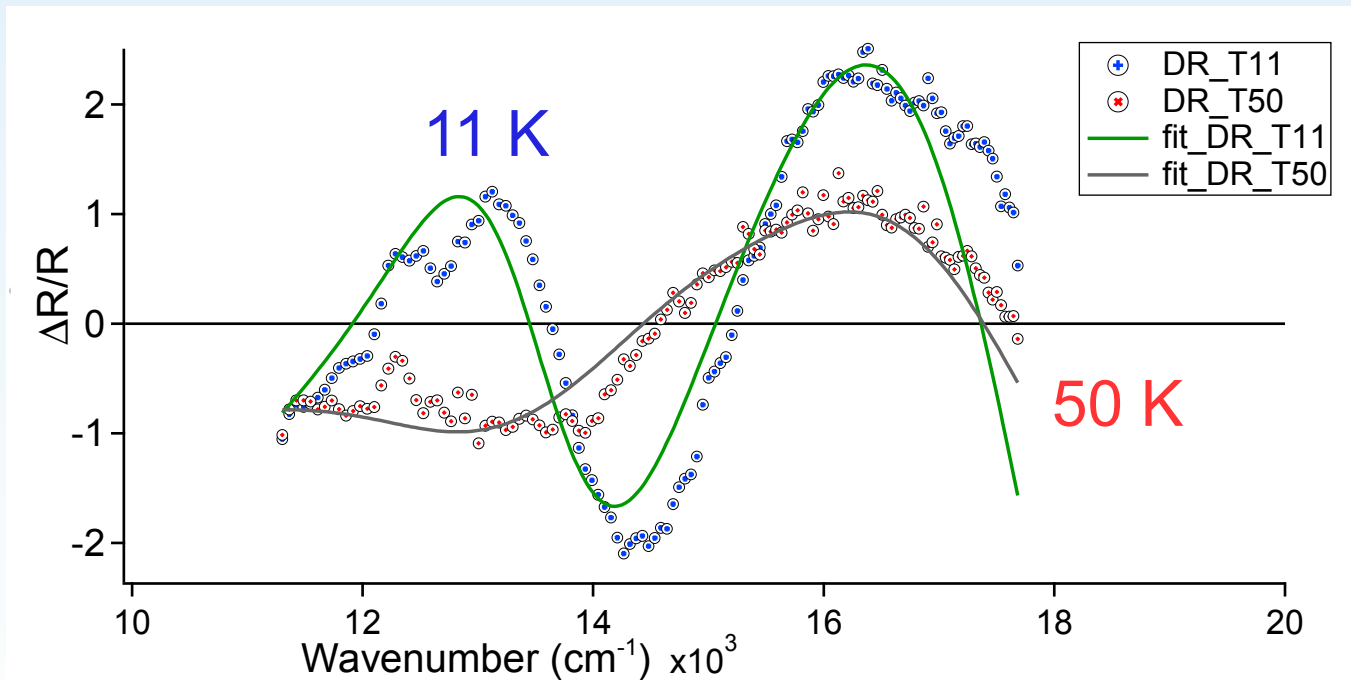
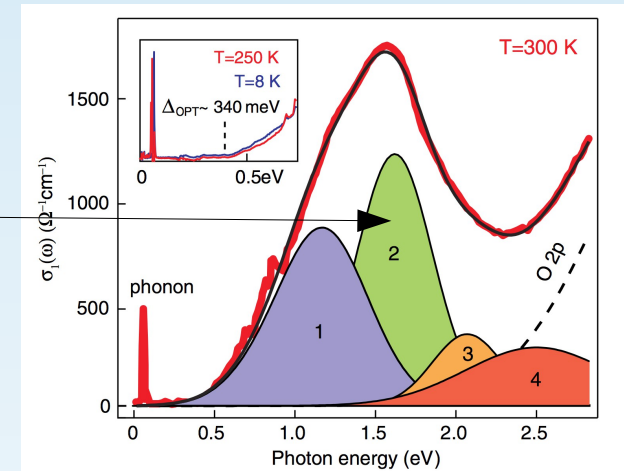
What are the parameter that changes?

Preliminary results and open problems

Differential fit to describe the variation of reflectivity

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 decouple 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!