Time domain studies of optical transitions in insulating transition metal oxides

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Aknowledgement

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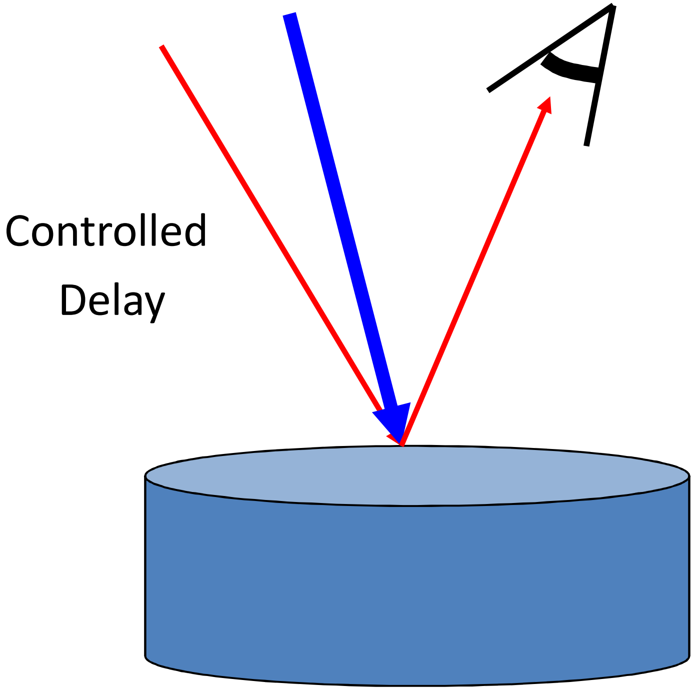
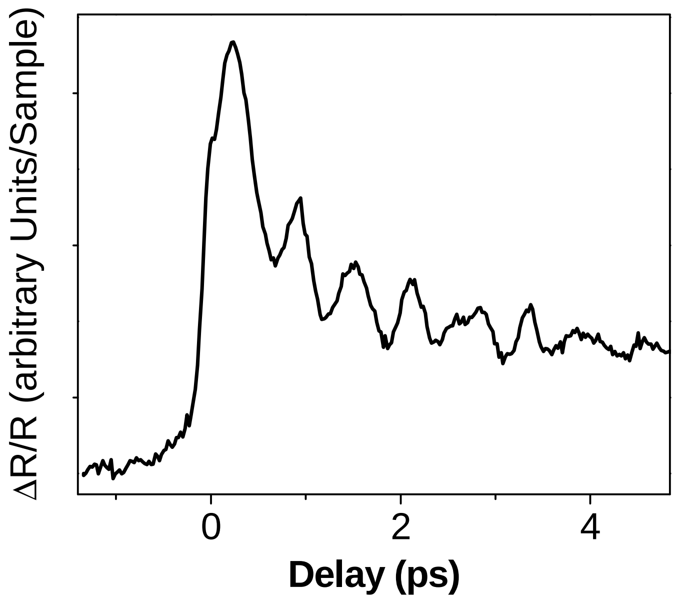
- ✓ Claudio Giannetti (Department of Physics, Università Cattolica del Sacro Cuore , Brescia)
- ✓ Julia Reul and Markus Gruninger (University of Koln, De)
- ✓ Paul van Loosdrecht, Tom T. Palstra (Zernike institue for advanced material, Groningen, Nl)
- ✓ Simon Wall (Fritz Haber institute Berlin, De)
- ✓ Andrea Perucchi (Sissi, Elettra, Trieste)

Pump and Probe measurements

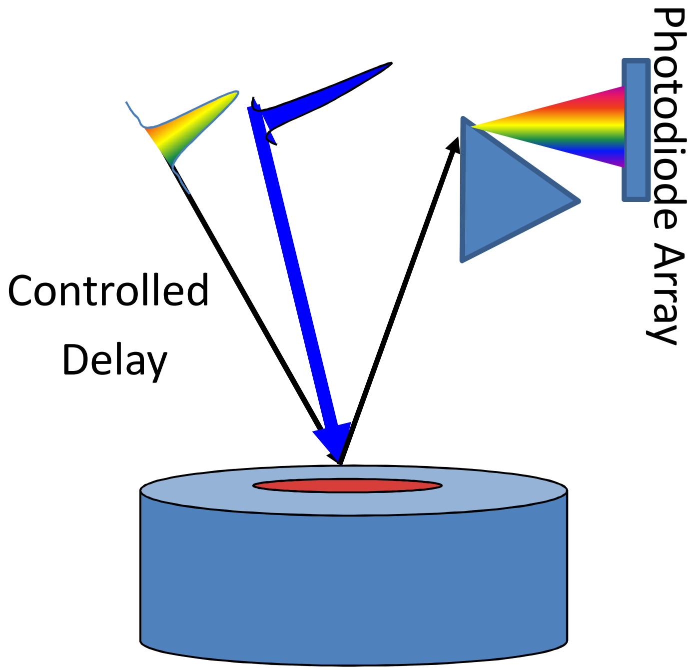


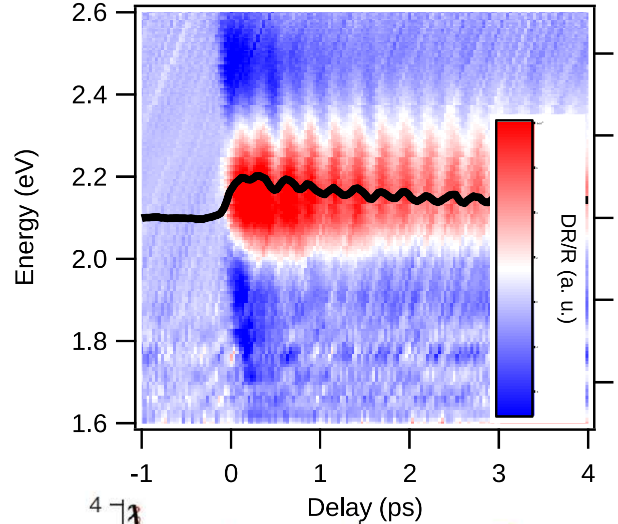


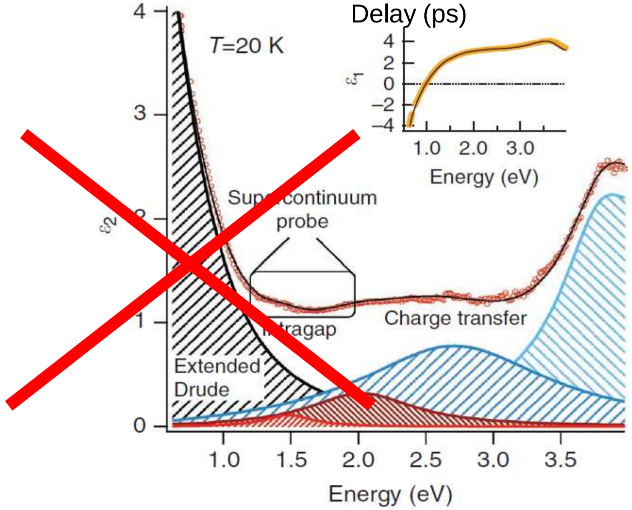
›**Low density excitation (Linear regime)**

›High density excitation (non-linear, optical control)

Pump and Probe Spectroscopy







Nat. Comm. 1354,1, 2011

- ✓ Differential models based on equilibrium measurements

Outline

- ✓ Pump&Probes spectroscopy and broadband white-light probes on insulating transition metal oxides
- ✓ Hubbard exciton revealed by time-domain optical spectroscopy in YVO_3
- ✓ Charge transfer excitation in La_2CuO_4

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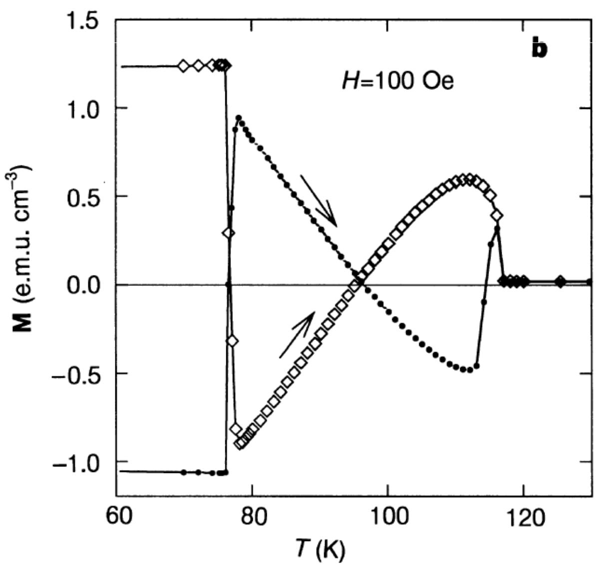
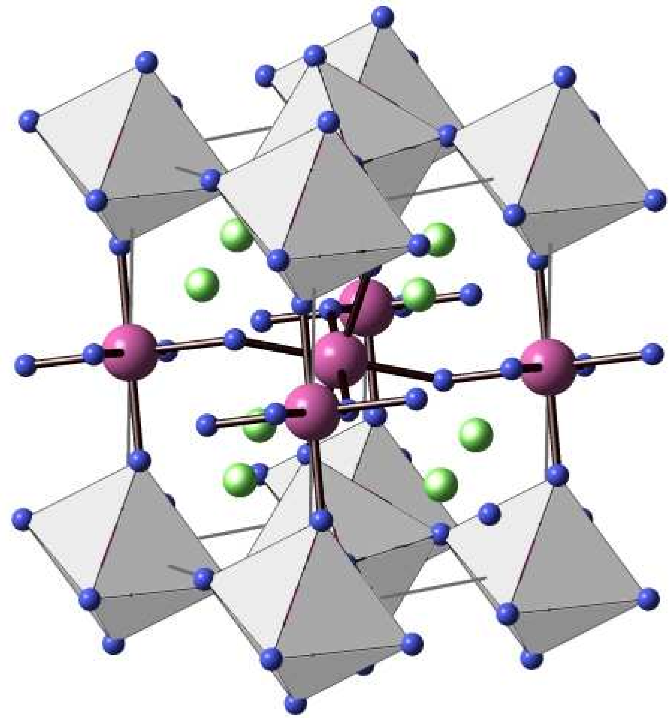
T-ReX Research Group

YVO_3: Properties

- ✓ Layer compound of BaTiO3-like distorted/tilted octahedra (Pbnm)
- ✓ $V^{3+} \rightarrow 3d_2$
- ✓ Mott insulator
- ✓ Magnetization reversal
- ✓ Orbital/Spin orderings



Y. Ren, Nature 396, 401,1998

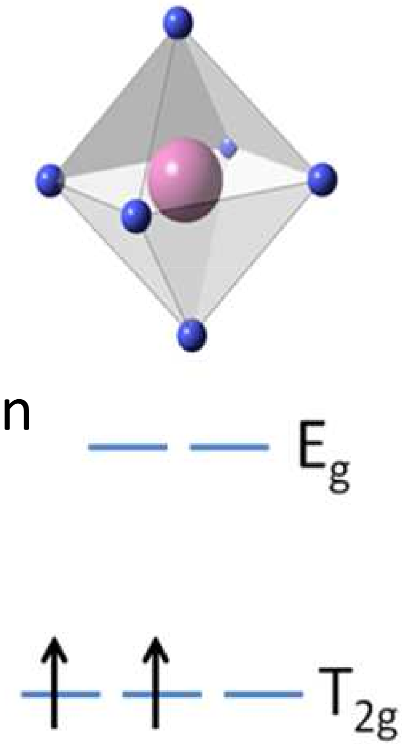


FERMI @elettra

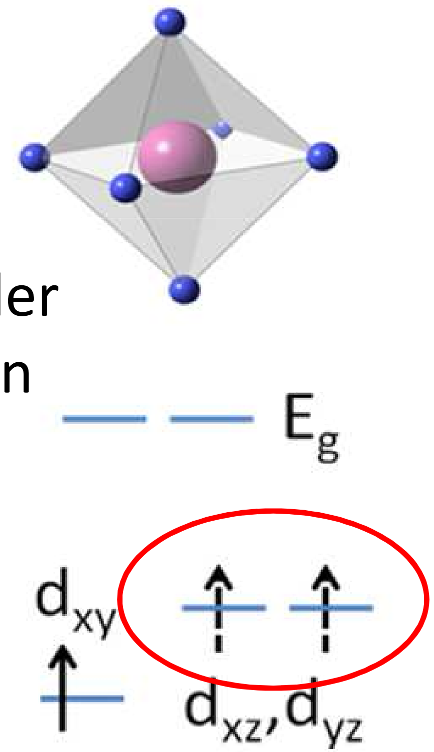
YVO_3: Properties

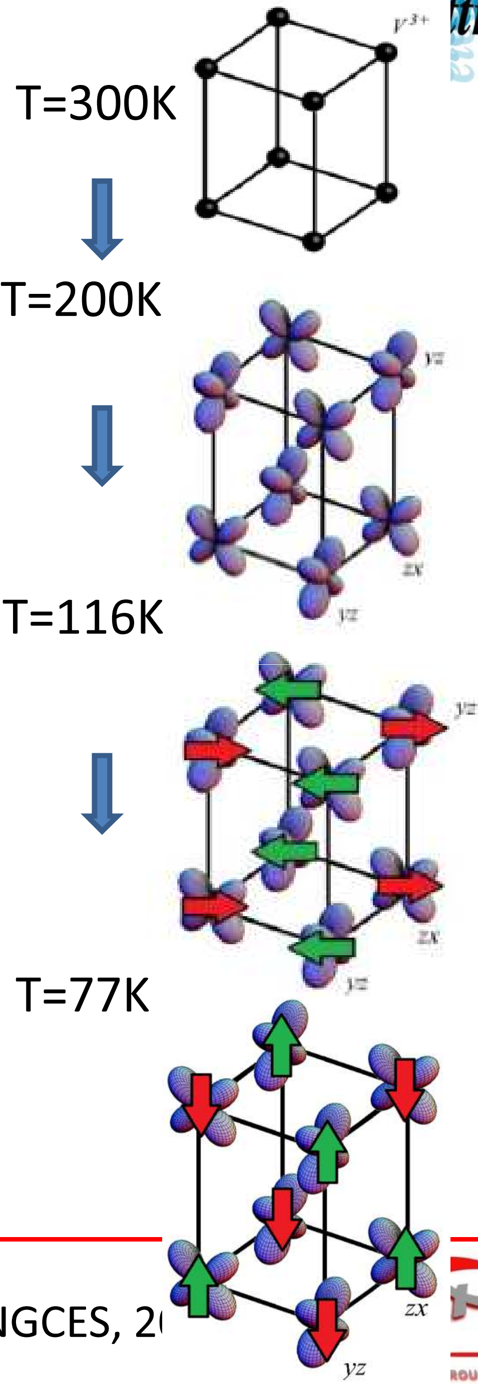
Orbital Physics in V^{3+} ($3d_2$)

Oxygen octahetron



Jahn-Teller distortion

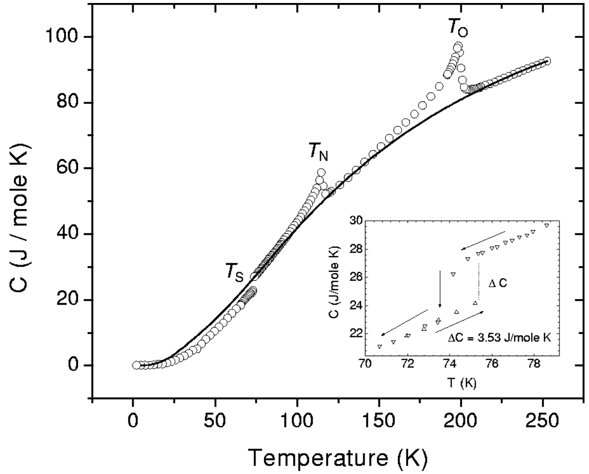


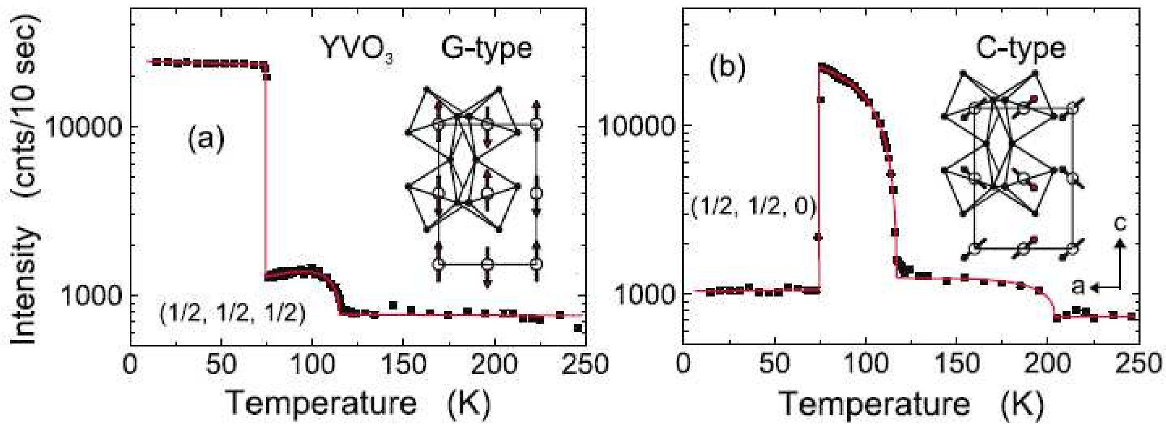


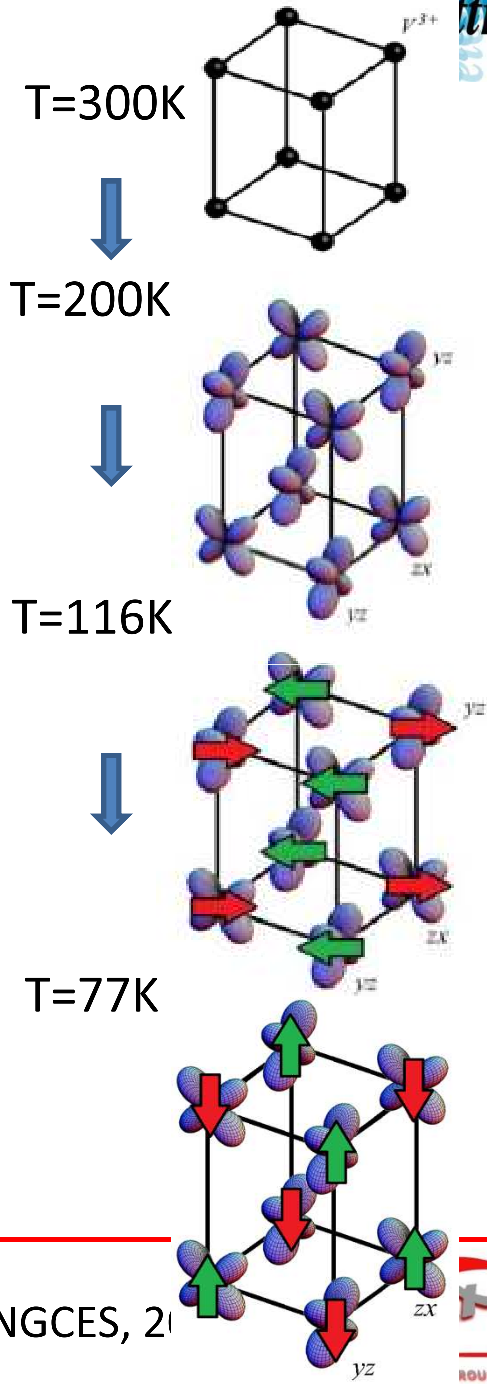
YVO_3: Properties

- ✓ Mott insulator

  Mott gap ~1.2eV
- ✓ Crystal field determined "mainly" by JT
- ✓ No Quantum fluctuation



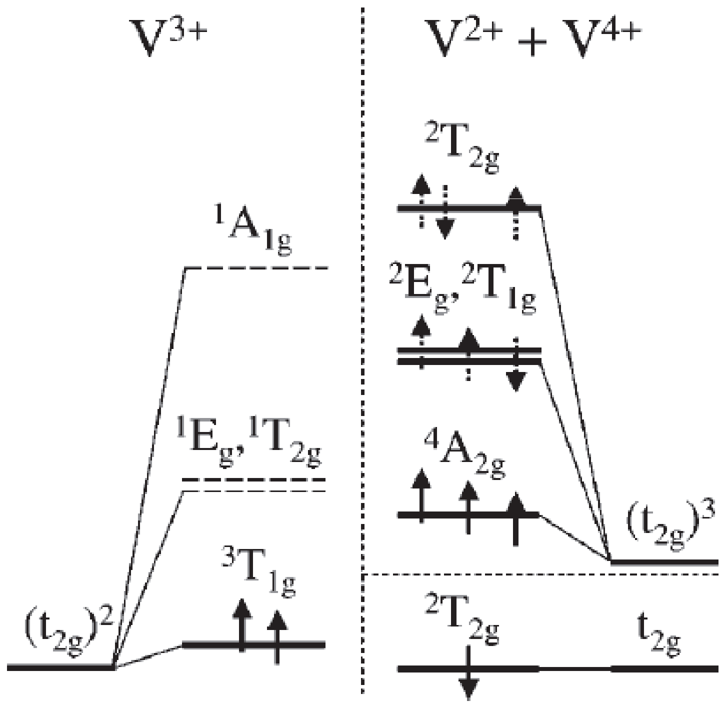




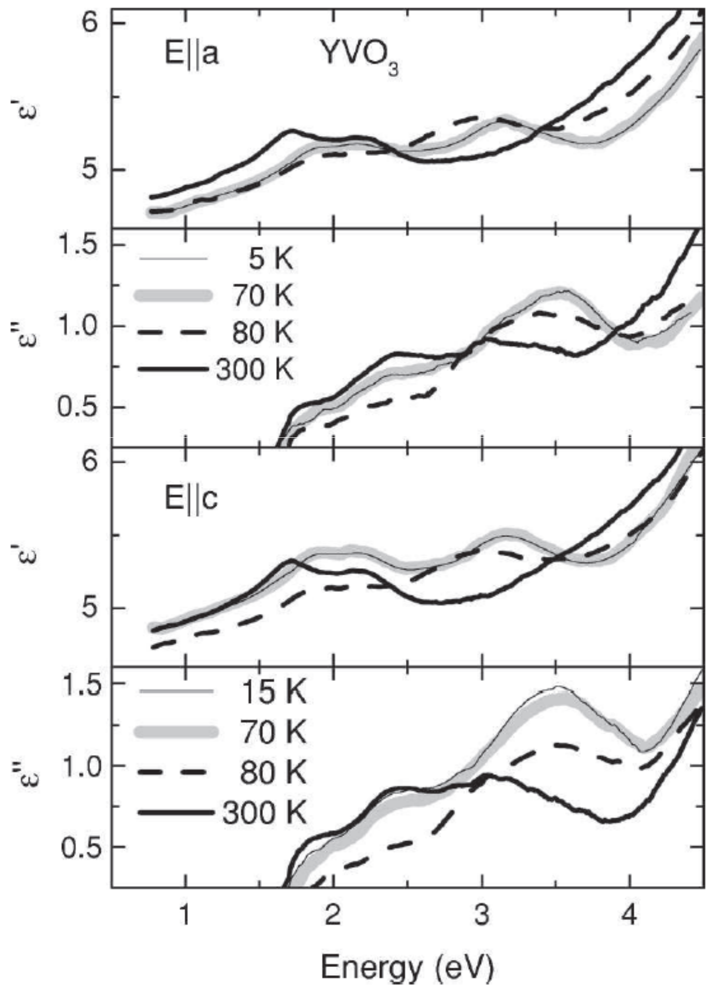
PRB, 65, 174112 (2002); PRL 91, 257202 (2003); PRL 99, 126402 (2007)

YVO_3: Optical Properties

- ✓ $d_2d_2 \rightarrow d_1d_3$
  $V^{3+}V^{3+} \rightarrow V^{2+}V^{4+}$
- ✓ Multiplet Calculations

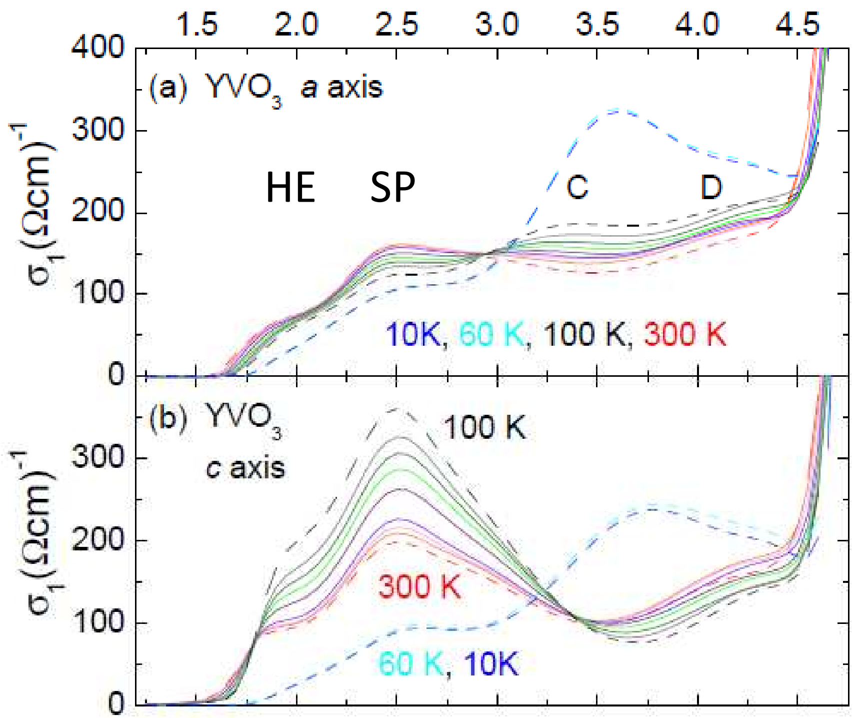


Tzvetkov A. & al. PRB, 69, 075110

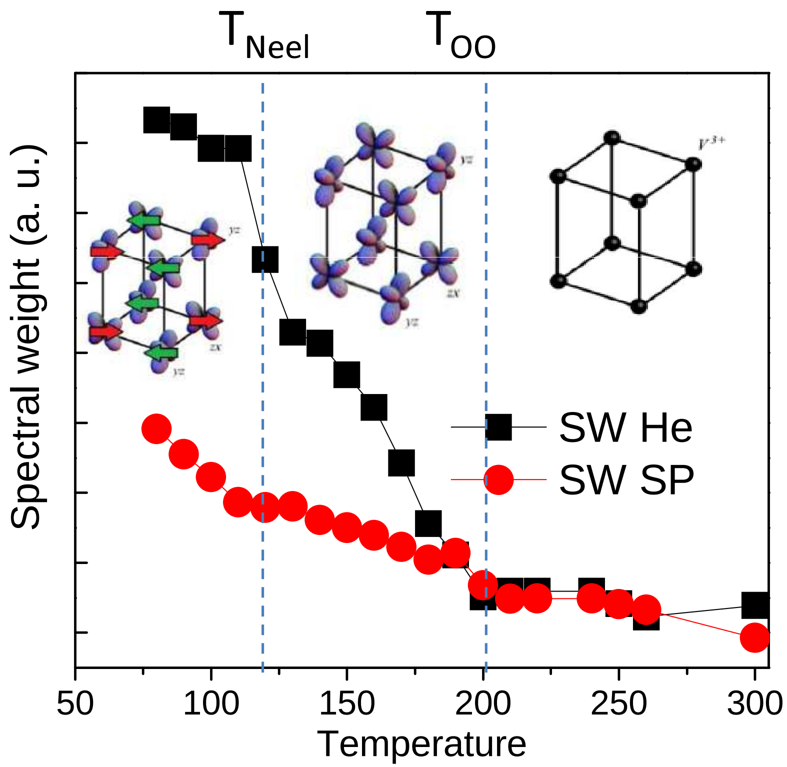


YVO_3: Optical Properties

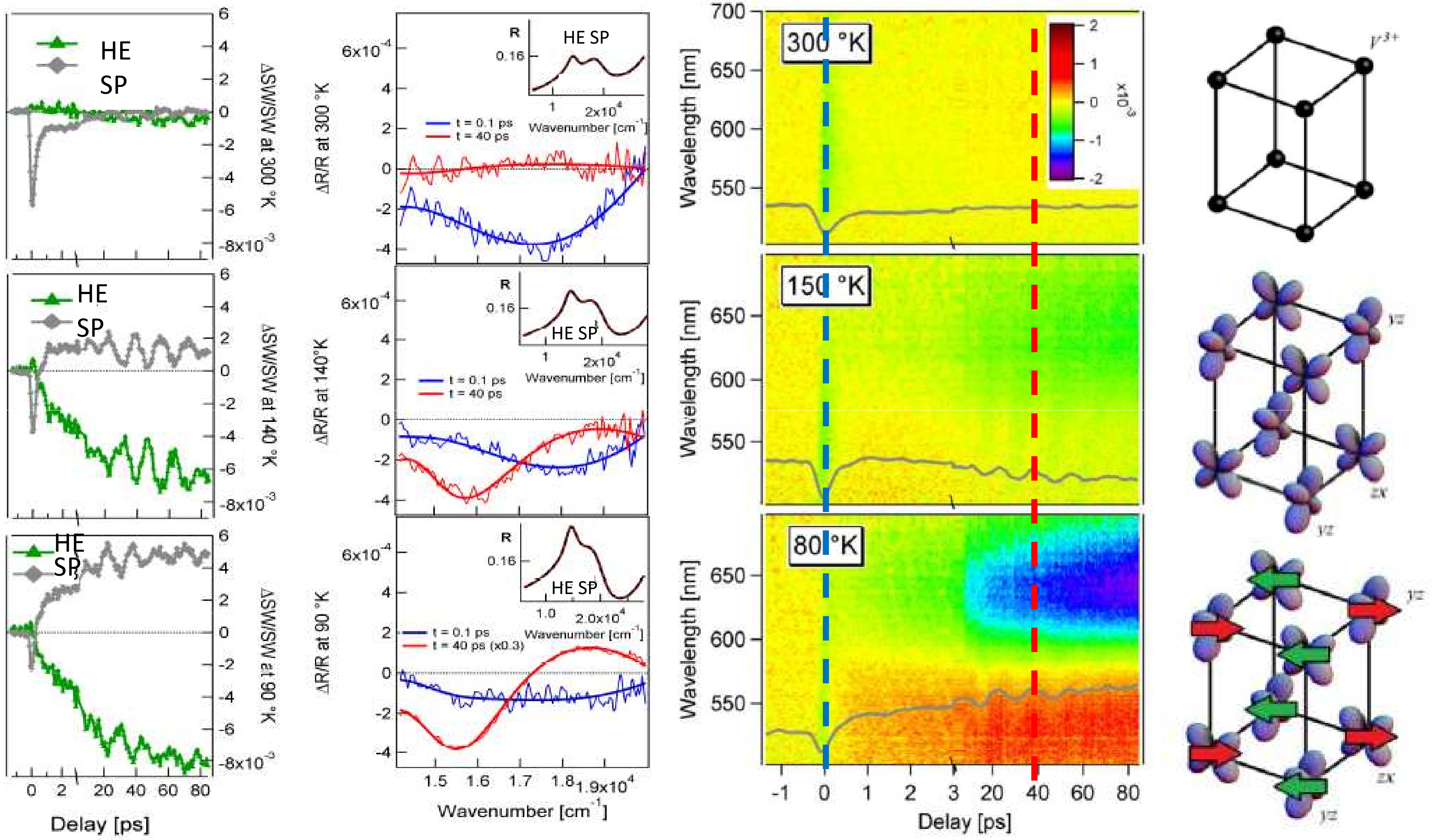
- ✓ 6 Gaussian and 1 Tauc-Lorentz oscillators
- ✓ Anomalous behavior of SW in the Spin and OO phases?!



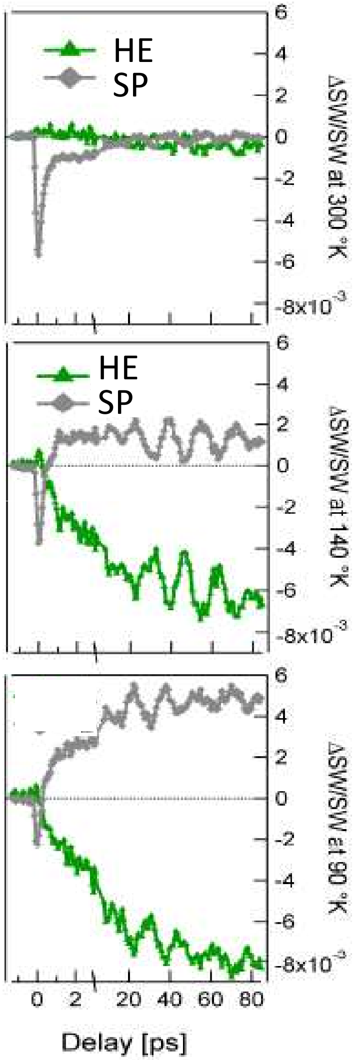
Reul. J, Gruninger M. & al. Preprint



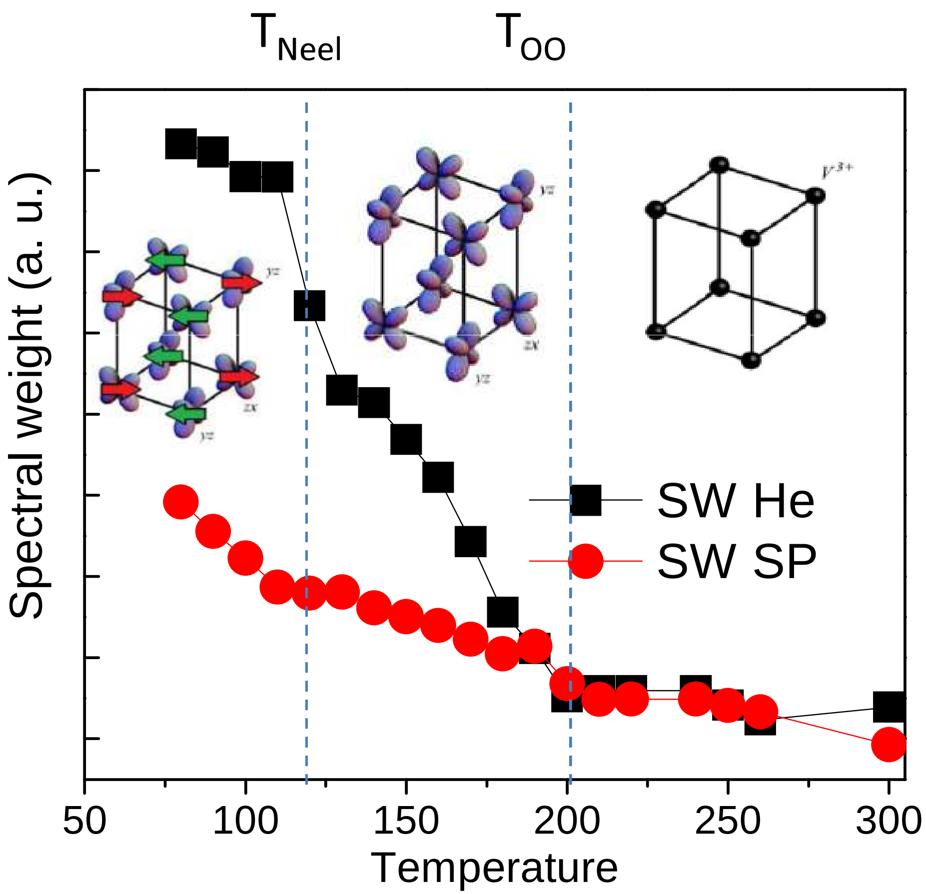
Transient Optical Properties



Thermal Vs. non-Thermal SW



- ✓ Thermal benchmark
- ✓ Non-Thermal Thermal contribution

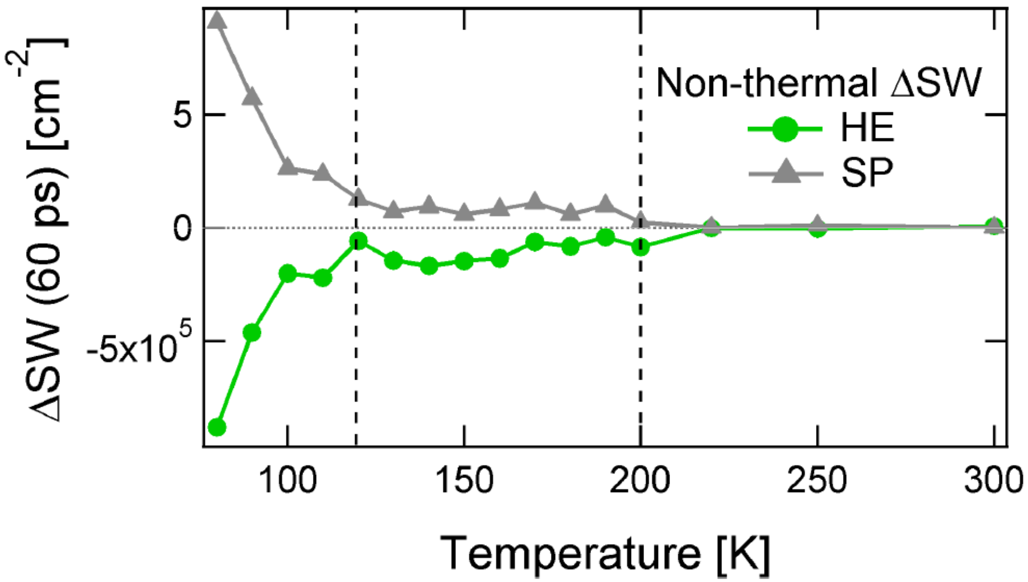


Thermal Vs. non-Thermal SW

- ✓ Thermodynamic estimate of the temperature variation in the photo-excited state(ΔT)

$$\Delta T[K] = \frac{Q_{abs} \cdot N_A \cdot V}{S \cdot d \cdot u \cdot C_{mol}}$$

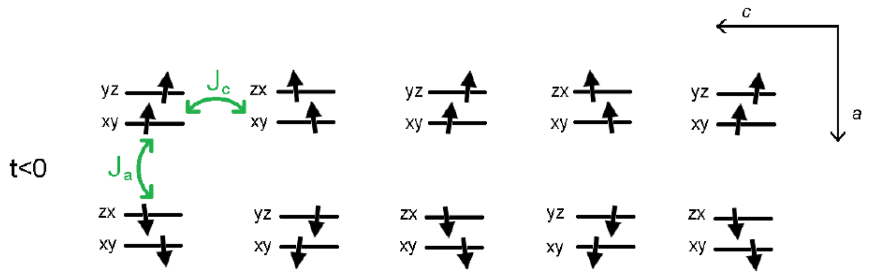
- ✓ Extrapolation of static optical properties (T_x+ΔT)



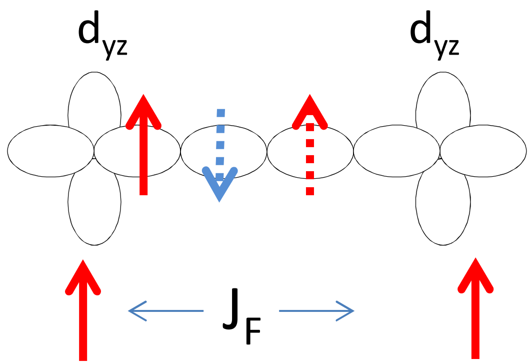
✓Direct Exchange of SW between the two bands

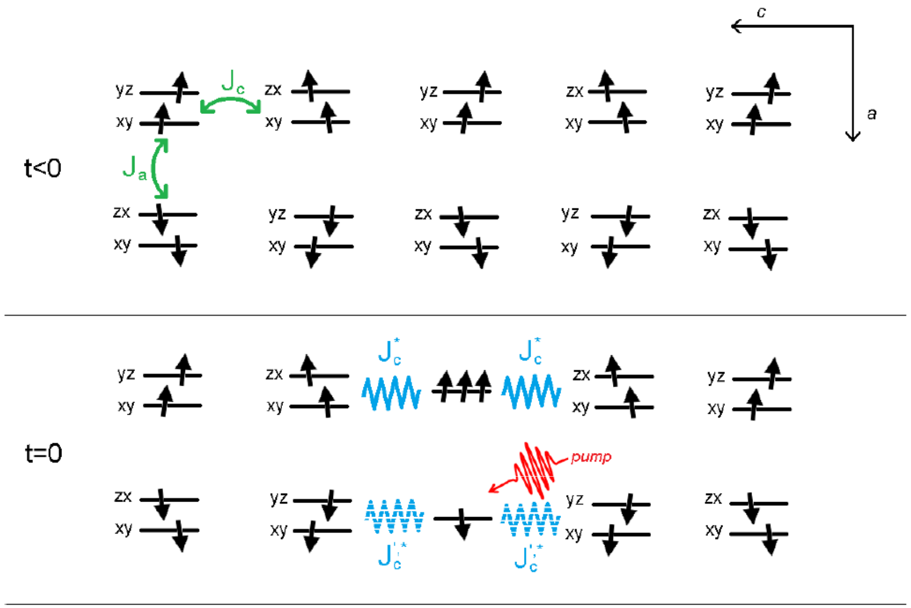
i.e. It is the same band!

Spectral weight exchange

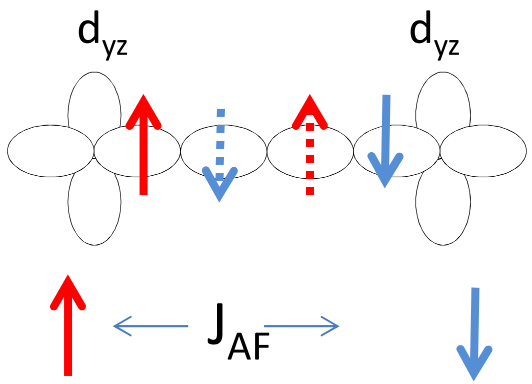


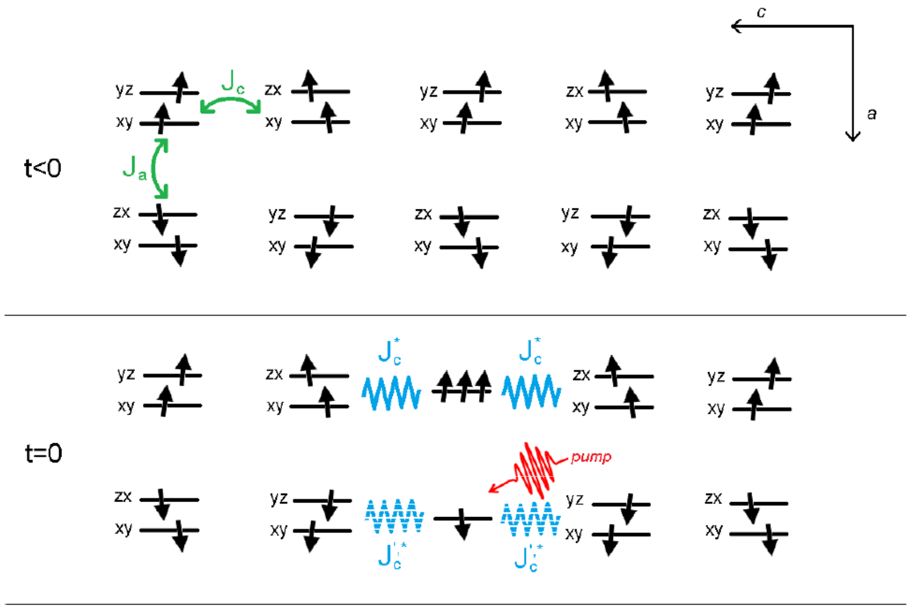
Spectral weight exchange



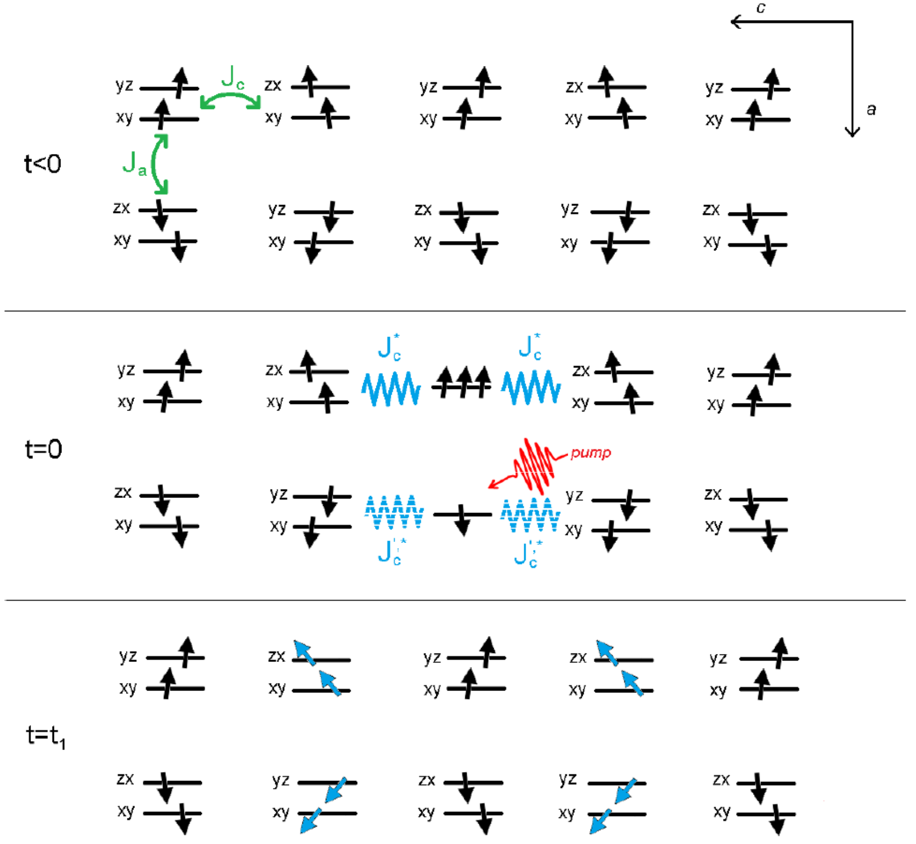


Spectral weight exchange





Spectral weight exchange

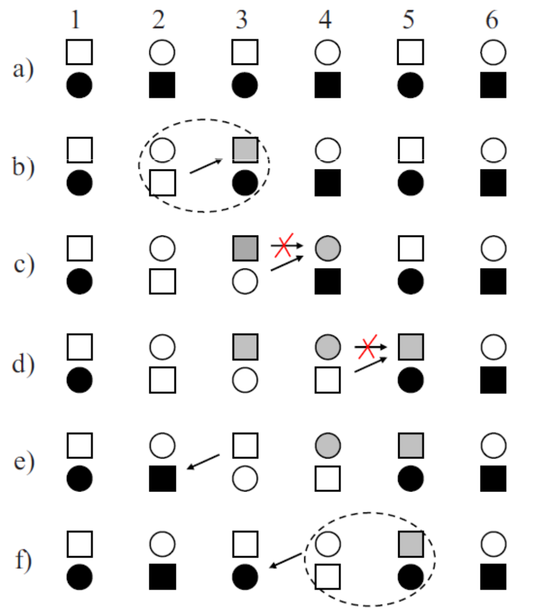


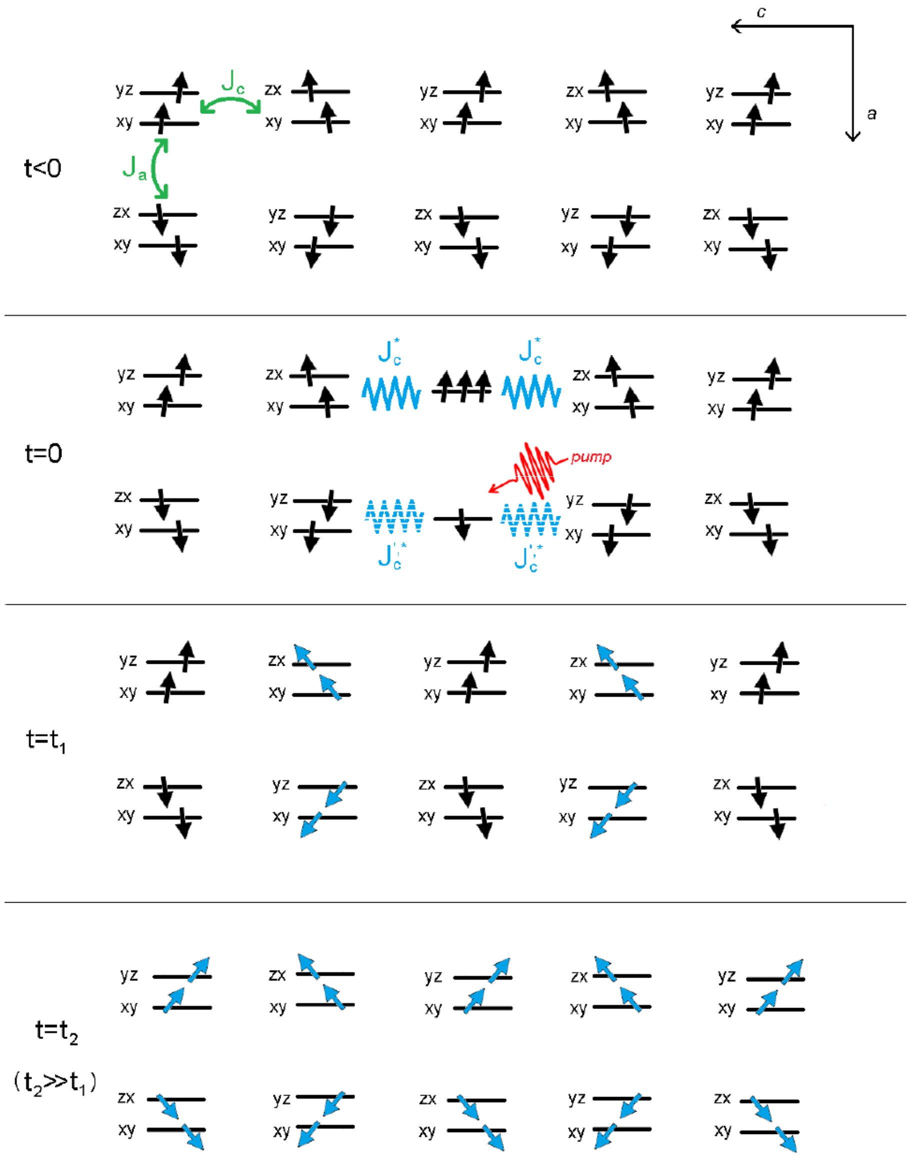
Spectral weight exchange

- ✓ Holon and Doublon are localized excitation

  Vs

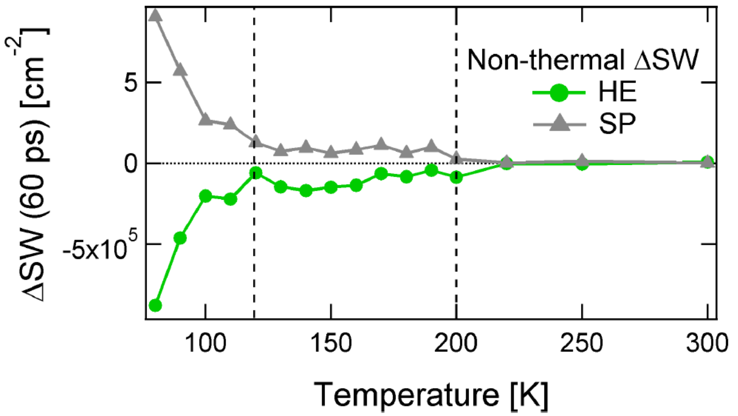
- ✓ Holon + Doublon (Exciton!)

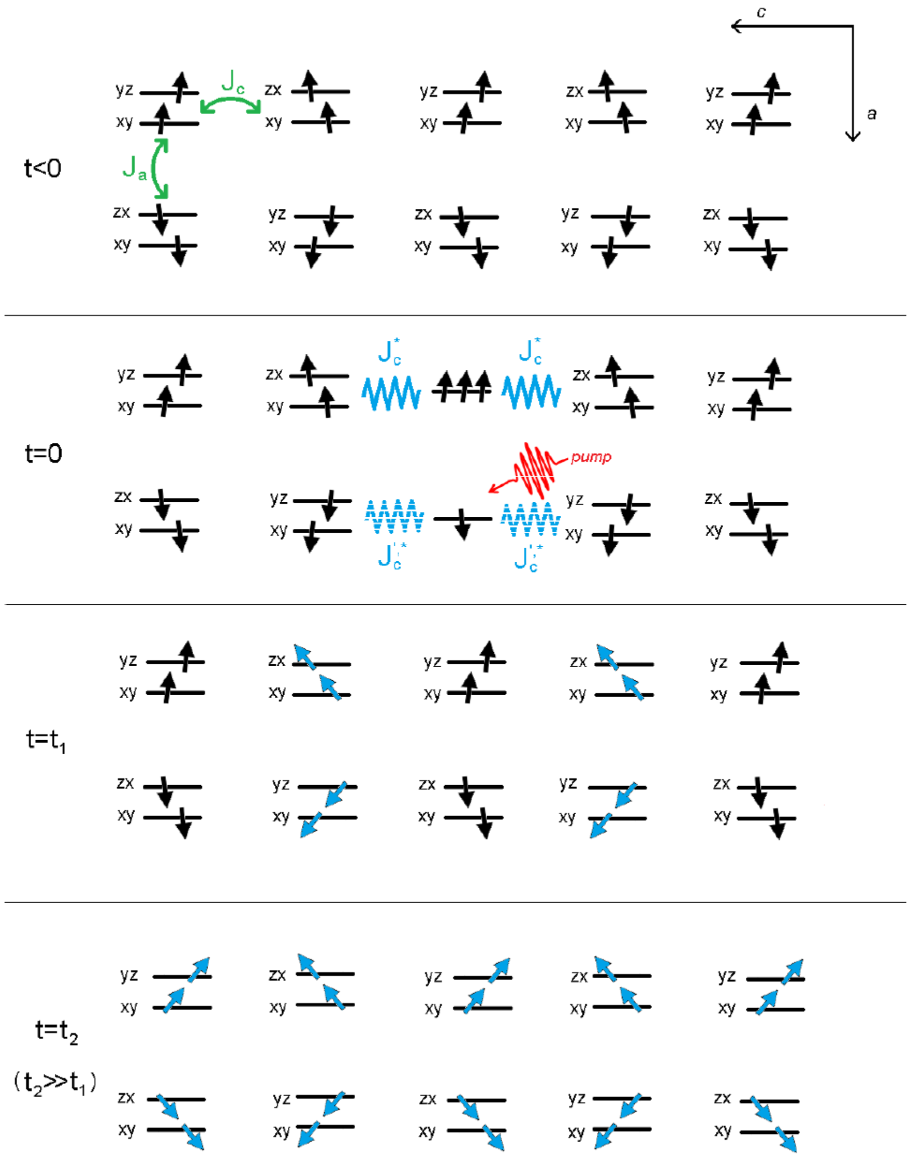




Phys. Rev. B, 65, 174112 (2008), PRB, 66, 035111 (2002).

Spectral weight exchange





Magnetic contribution to the kinetic energy of a bound state between "doublon" and "holon"!!

Conclusion 1

- ✓ Excitonic state on Hubbard band
- ✓ Quantify the magnetic contribution to the kinetic energy gain?!

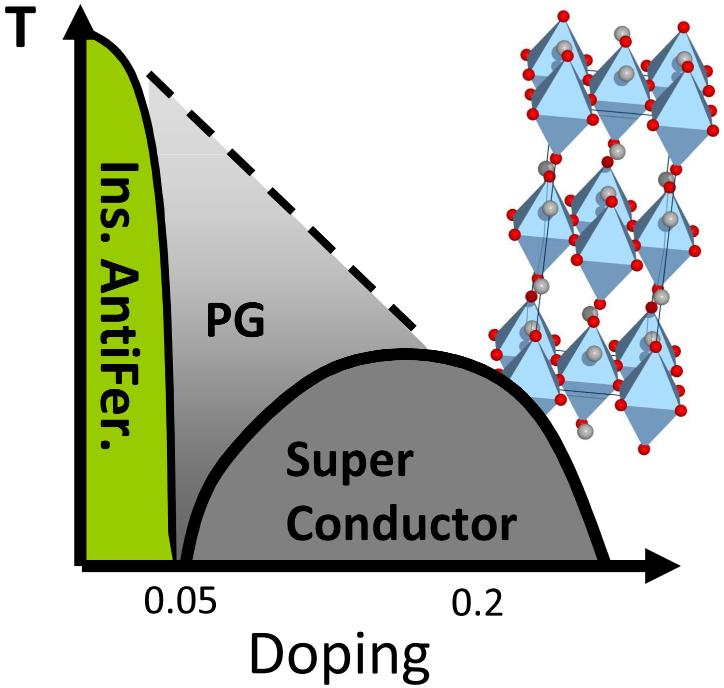
http://arxiv.org/pdf/1205.4609.pdf

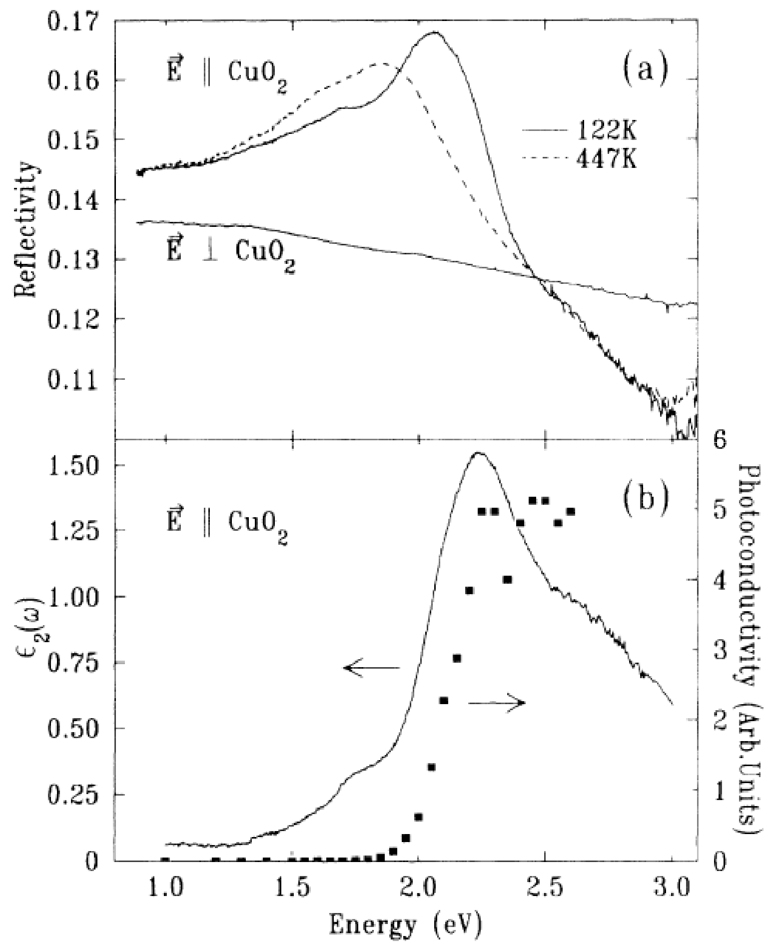
Outline

- ✓ Pump&Probes spectroscopy and broadband white-light probes on insulating transition metal oxides
- ✓ Hubbard exciton revealed by time-domain optical spectroscopy in YVO_3
- ✓ Charge transfer excitation in La_2CuO_4

Charge Transfer excitation in La_2CuO_4

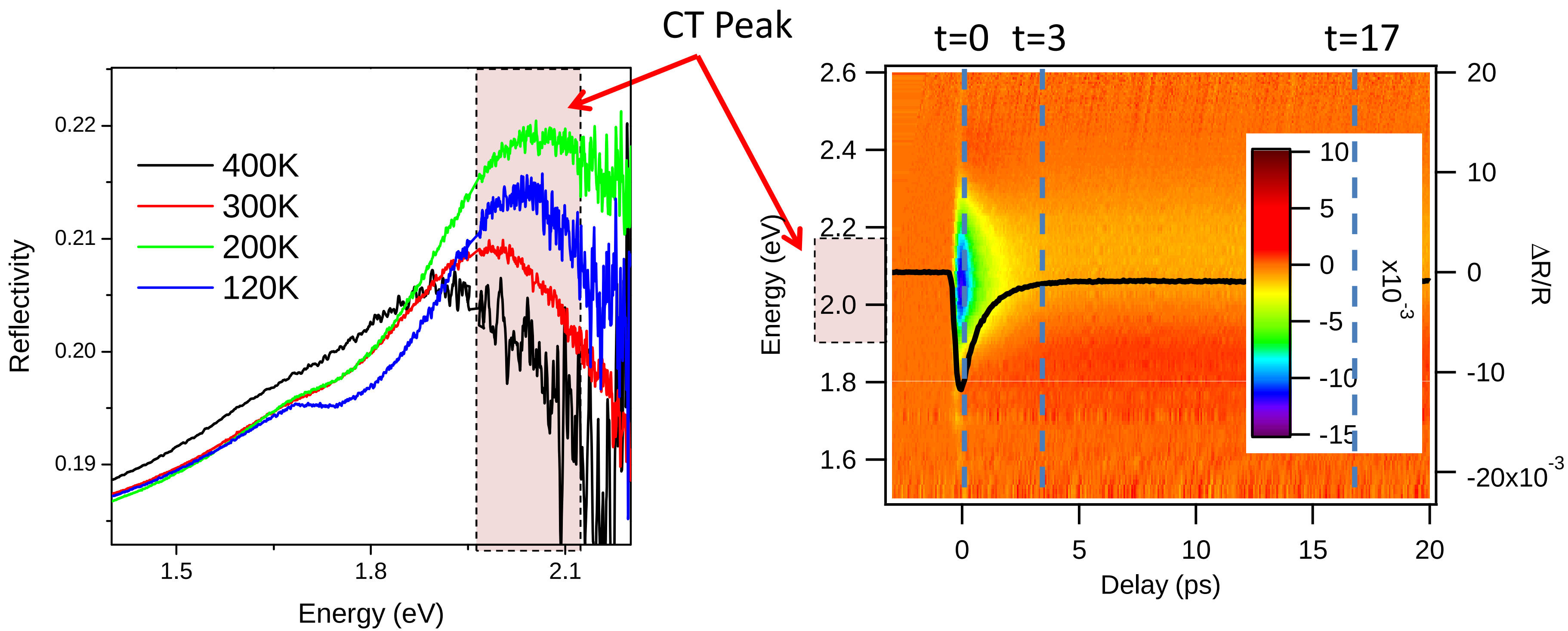
- ✓ Layered Structure
- ✓ $Cu^{2+} \rightarrow 3d^9$
- ✓ Doped 2D Spin ½ System





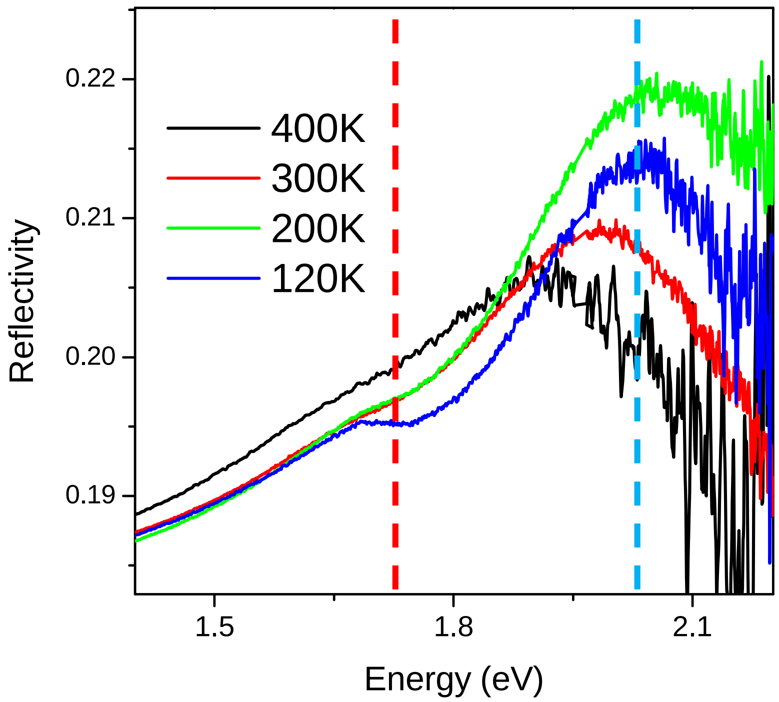
Phys. Rev. Lett. 69, 1109 (1992)

Static and Time domain Ref. in La_2CuO_4

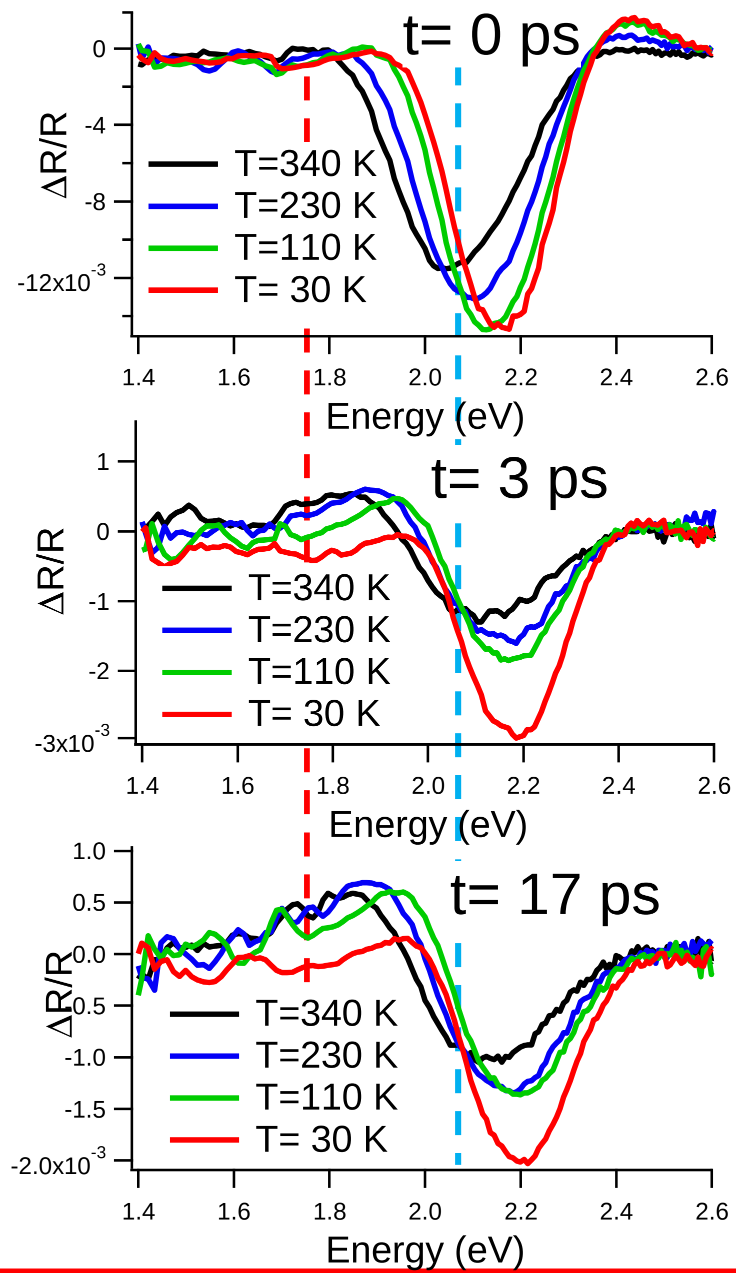


- ✓ Charge Transfer is strongly perturbed in the time domain
- ✓ Dynamical measurements of the charge transfer

- ✓ Charge Transfer is strongly perturbed within 1 ps (Mott gap collapse)
- ✓ The sideband is perturbed much late



- ✓ **Pump 1 eV = Pump 3eV!?**



Proposed Approach

Short Range interaction for Frenkel “like” excitons

$$\epsilon_2(\omega) \propto |1 - gF(\omega)|^{-2} \mathrm{Im}F(\omega) \qquad F(\omega) = -\lim_{\eta \to 0} \int_0^\infty \frac{D(\omega')d\omega'}{\omega - \omega' + i\eta}$$

Electrons and holes form Frohlich polarons so that the T dependence of the polaron self-energy causes a shift of the band edge:

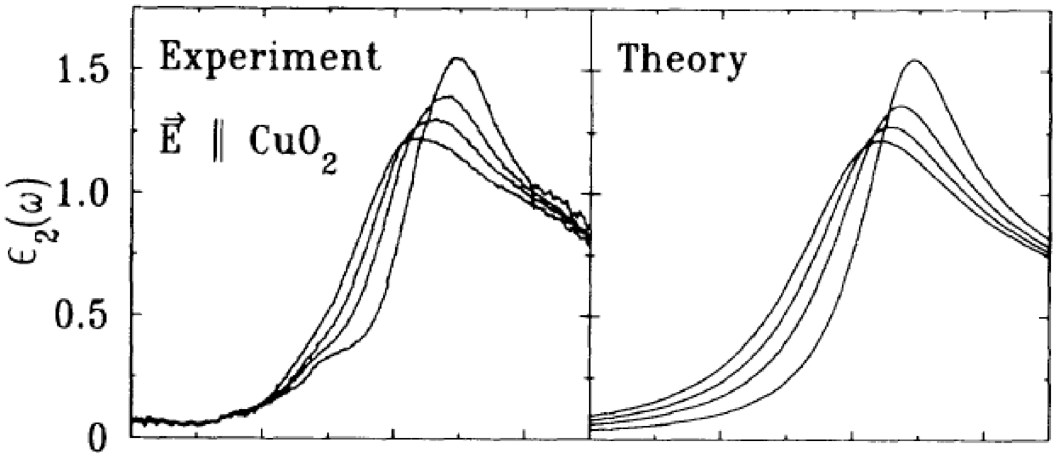
$$E_g(T) = E_g^0 - 2\hbar\omega_0\alpha_p[n(\hbar\omega_0/k_B T) + 1]$$

Polaron Coupling constant

$$\alpha_p = \frac{1}{2}\frac{e^2}{2\hbar\omega_0}\left(\frac{1}{\epsilon_\infty} - \frac{1}{\epsilon_s}\right)\left(\frac{2m\omega_0}{\hbar}\right)^{1/2}$$

Polaron Relaxation Rate

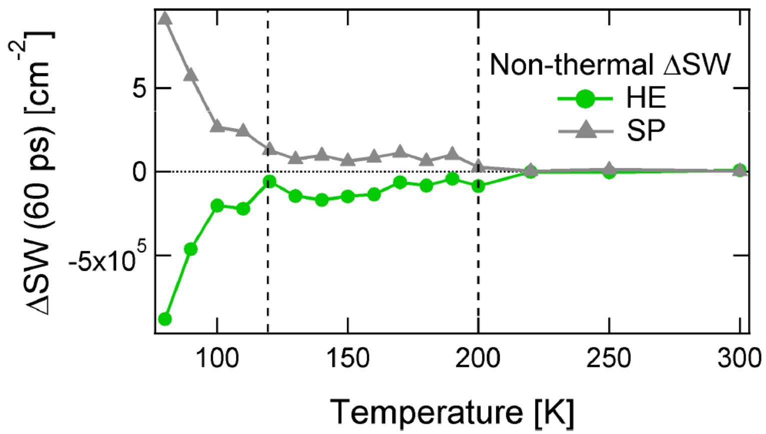
$$\gamma(T) = 2^{3/2}\omega_0(\hbar\omega_0/E)^{1/2}\alpha_p[2n(\hbar\omega_0/k_B T) + 1]$$



Phys. Rev. Lett. 69, 1109 (1992)

Conclusions

- ✓ In absence of free carriers time domain spectroscopy can be used to address the nature of high energy optical excitations
- ✓ YVO_3: Unveiled the nature of the twofold band, Hubbard exciton, and magnetic contribution to kinetic energy



- ✓ La_2CuO_4: Preliminary study of charge transfer excitation