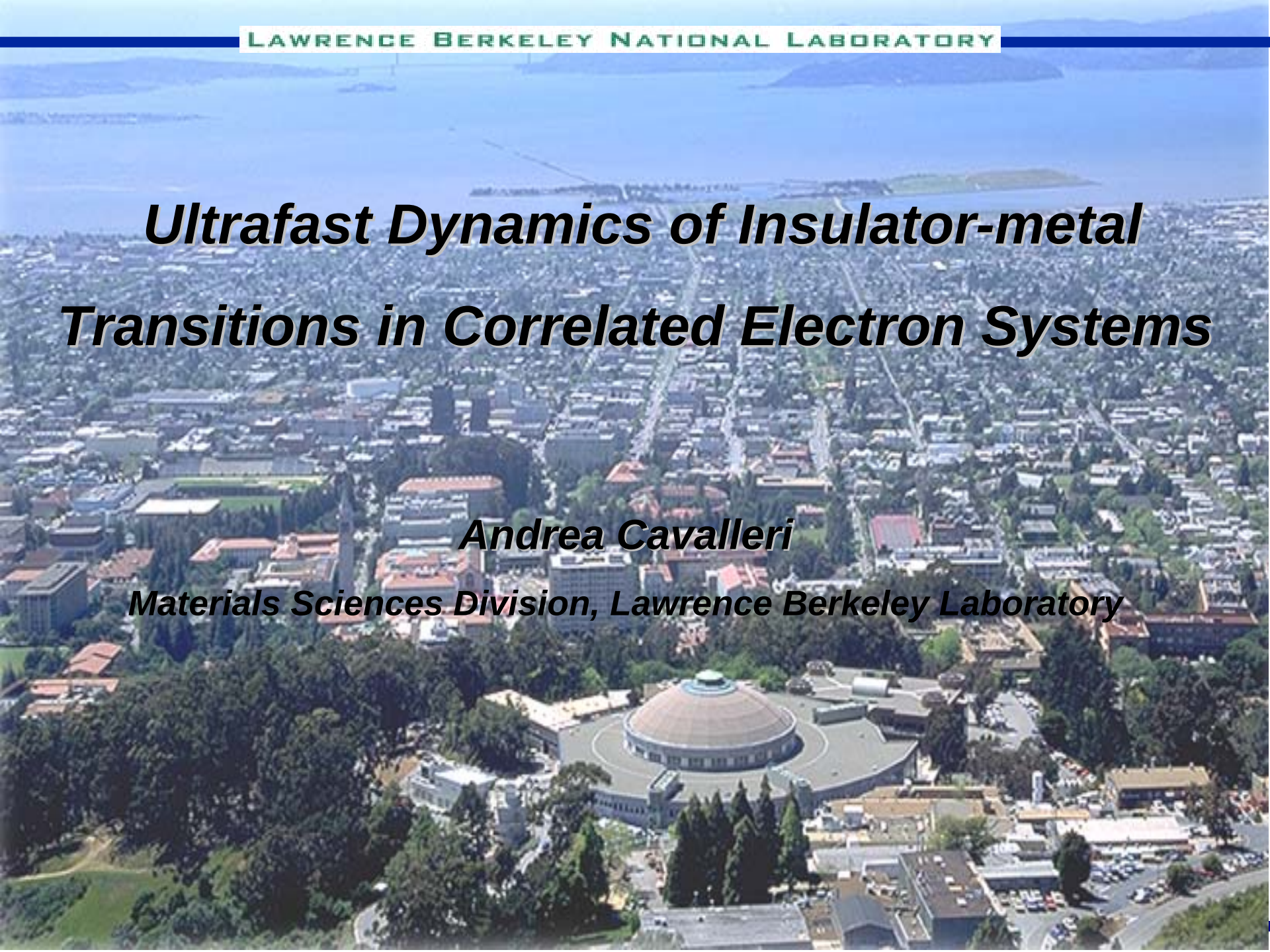


Ultrafast Dynamics of Insulator-metal Transitions in Correlated Electron Systems

Andrea Cavalleri

Materials Sciences Division, Lawrence Berkeley Laboratory

Contributors

M. Rini, H.H.W. Chong, R.W. Schoenlein
Lawrence Berkeley National Laboratory

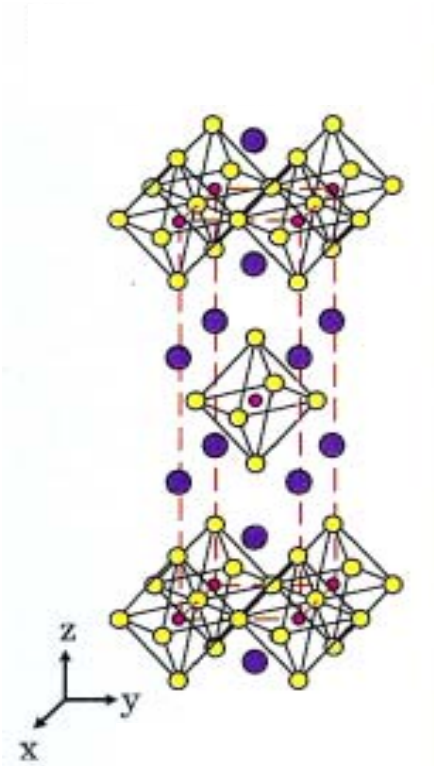
Th. Dekorsy
Forschungszentrum Rossendorf

S. Fourmaux, J.K. Kieffer
University of Quebec

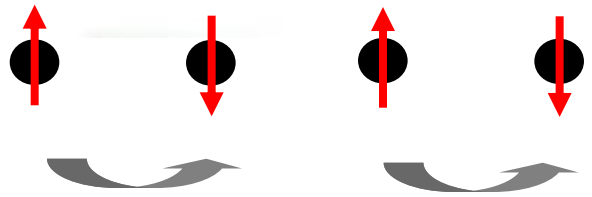
Strongly Correlated Materials

Oxides of Transition Metals

(e.g. Cu, Mn, Ni, V…)



Electrons are strongly interacting



~ eV

1) Unconventional Phenomena (e.g. Mott Insulator, High-T_c superconductivity, Colossal Magnetoresistance, Metal-Insulator transitions,…….)

2) Interesting phenomena and phase transitions at high temperatures

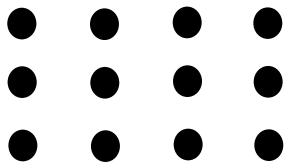
Ground States of Matter

$$F = E - TS$$

Low T

Minimum energy, low symmetry, order

e.g. crystalline



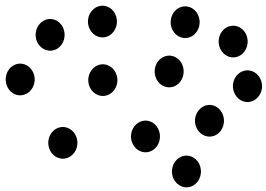
E minimum

$F = \textcircled{E} - TS$

High T

Maximum entropy, high symmetry, disorder

e.g. liquid

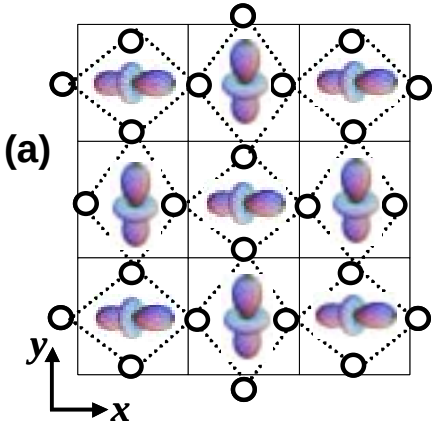


S maximum

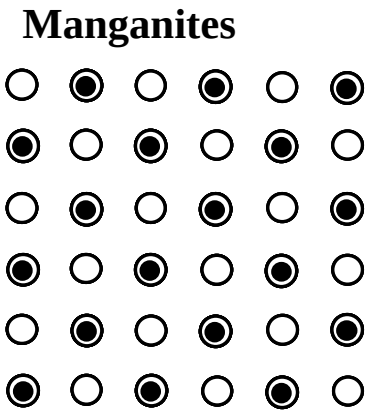
$F = E - T\textcircled{S}$

Exotic Ground States in Complex Solids

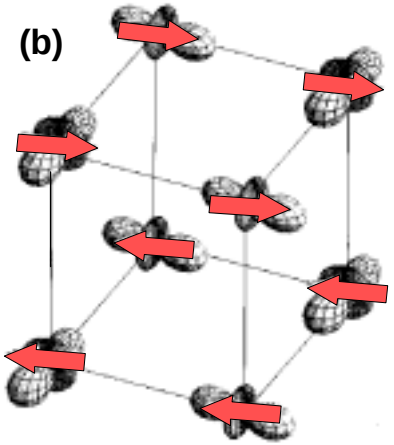
Jahn-Teller Instability
Orbital order



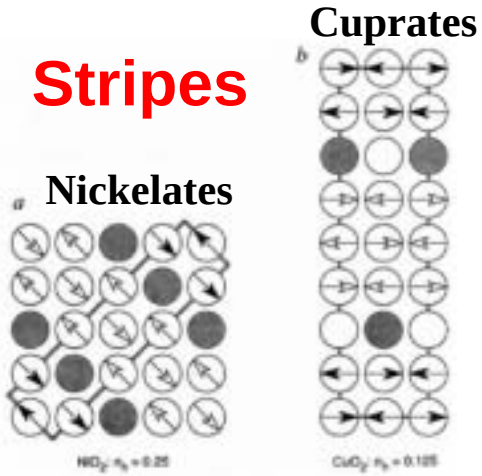
Charge order



Spin order



Stripes

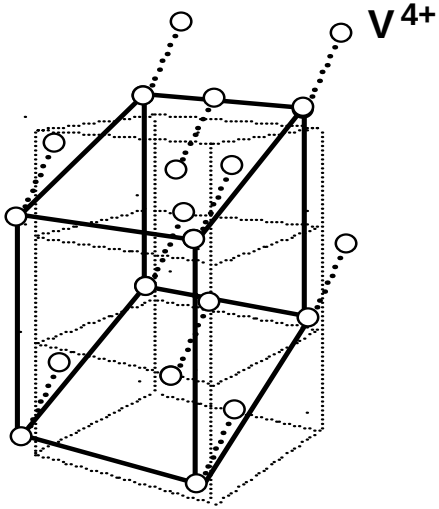


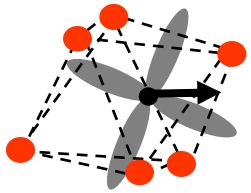
Understand interactions between

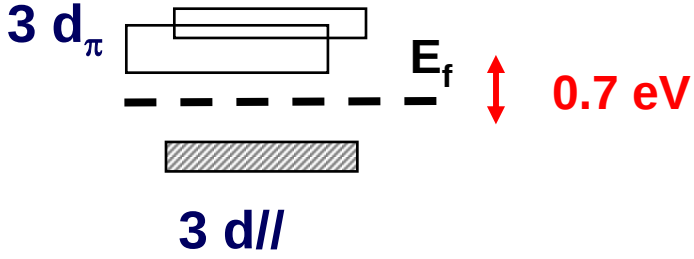
- ✓ **Atomic arrangements**
- ✓ **Carrier doping/ordering**
- ✓ **Magnetic ordering**

Metal-Insulator transition in VO_2: structure

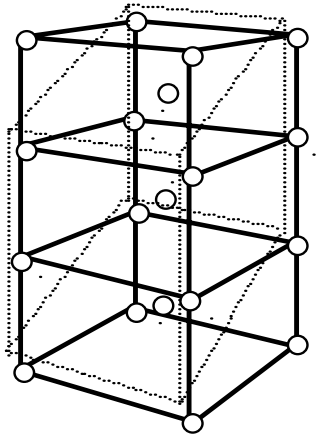
T < 340 K **Cell-doubled (monoclinic) insulator**

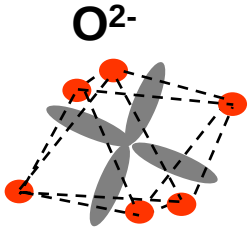


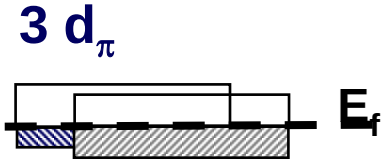




T > 340 K **Rutile - metal**

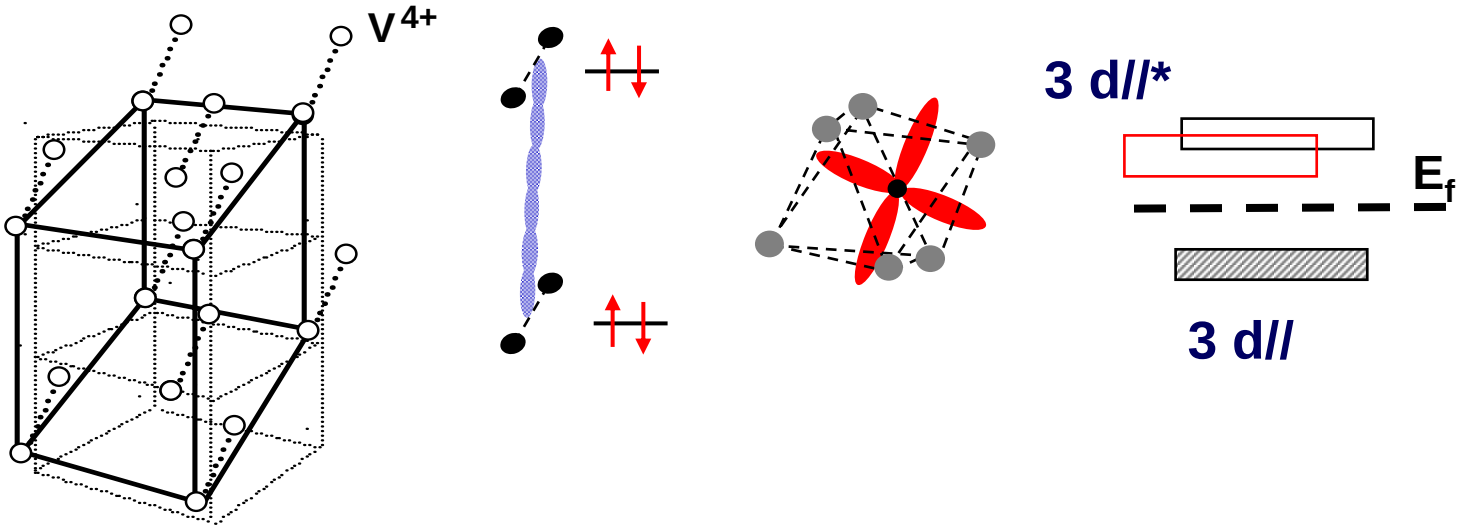




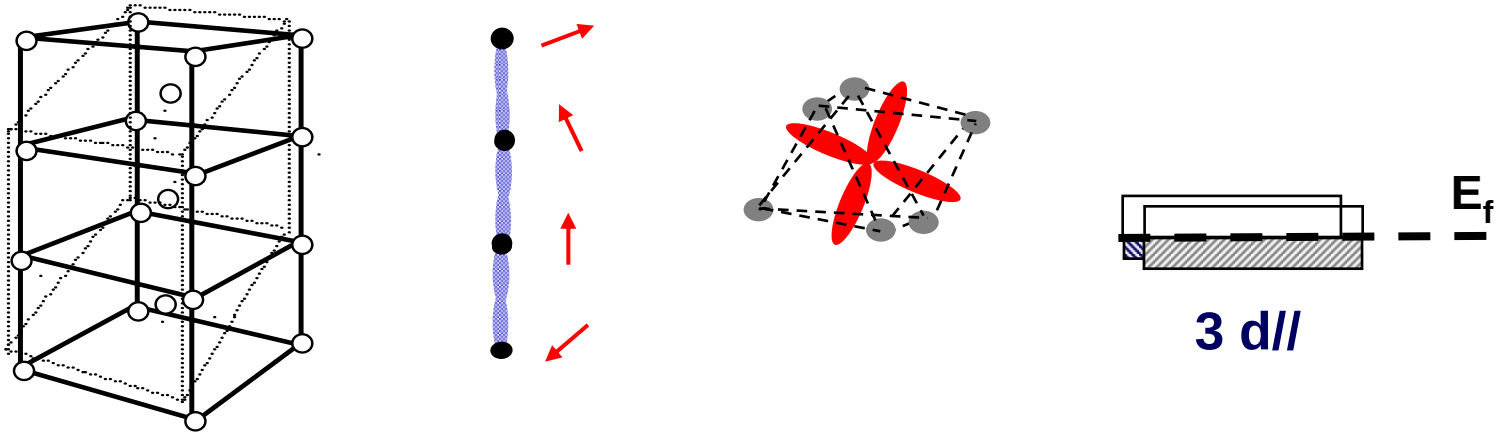


Metal-Insulator transition in VO_2: electrons

T < 340 K — **Singlets - Correlated Electrons - Insulator**

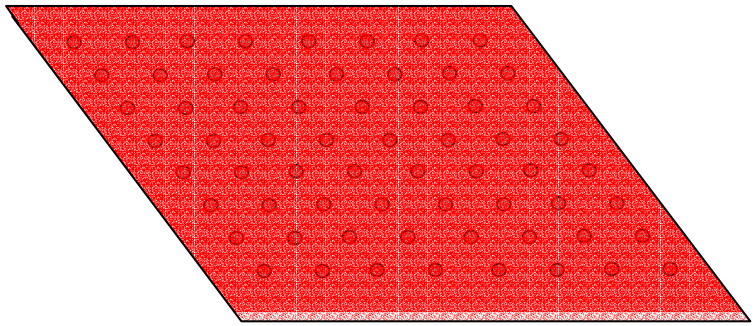


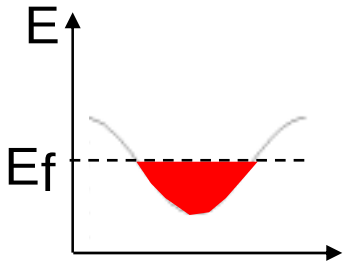
T > 340 K — **Delocalised - Paramagnetic - Metallic**



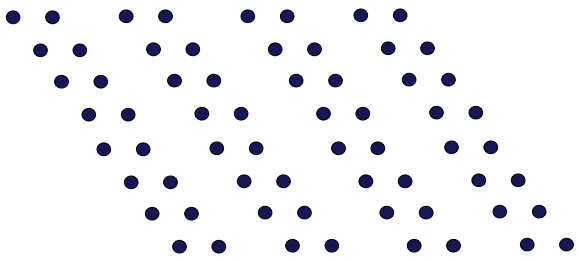
Peierls Transition and Metal-Insulator

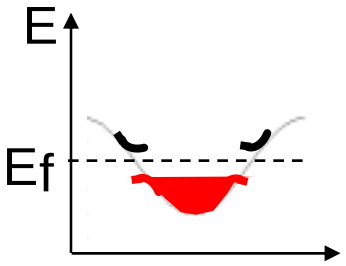
T > 340 K





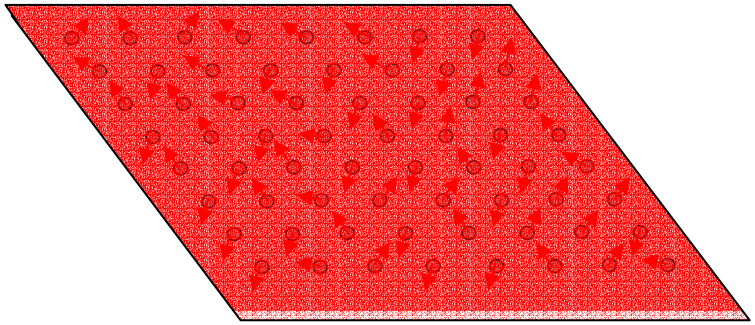
T < 340 K



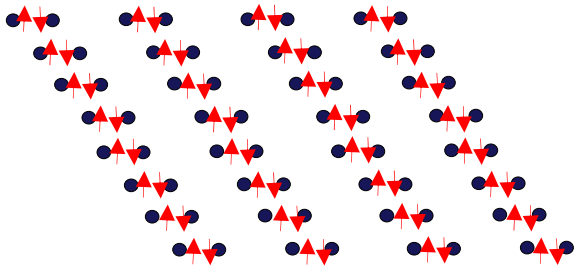


Spin Peierls Transition and Metal-Insulator

T > 340 K



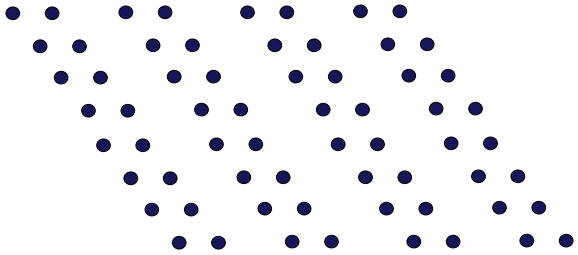
T < 340 K

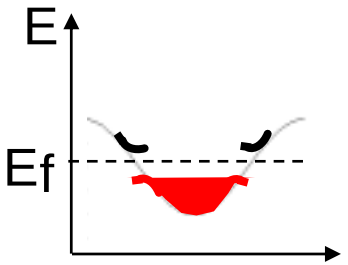


Singlet state: e-e minimum

Band Insulator or Mott Insulator?

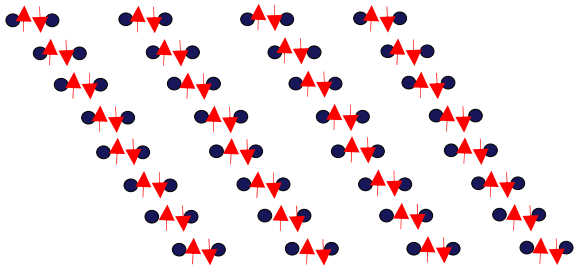
Band Insulator ?

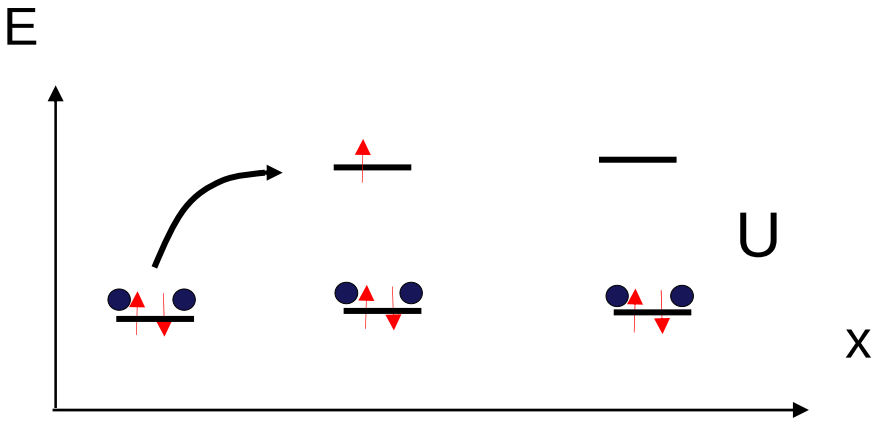




J.B. Goodenough *Phys. Rev. 117*, 1442 (1960); Wentzcowitch et al. *Phys. Rev. Lett. 72*, 3389 (1994)

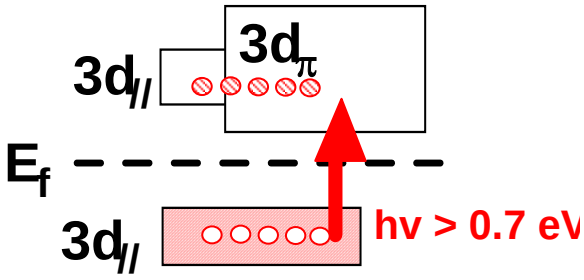
Mott Insulator ?





Zylbersztejn and N. Mott *Phys. Rev. B* 11, 4383 (1975)

Photo-induced Phase Transition



5 - 50% holes

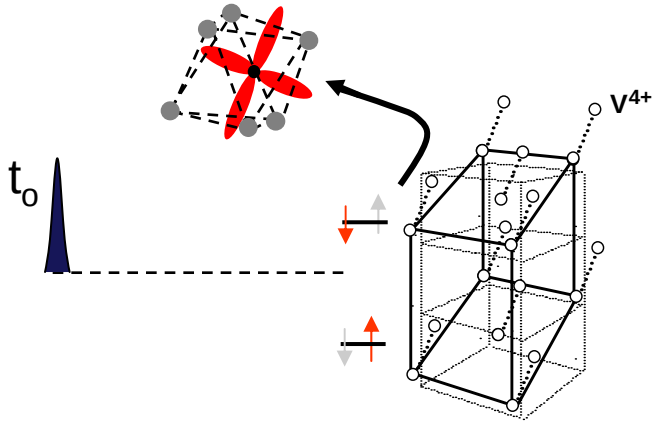
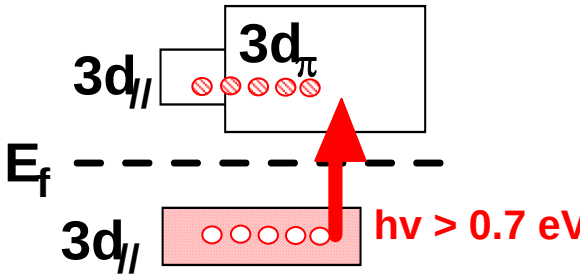
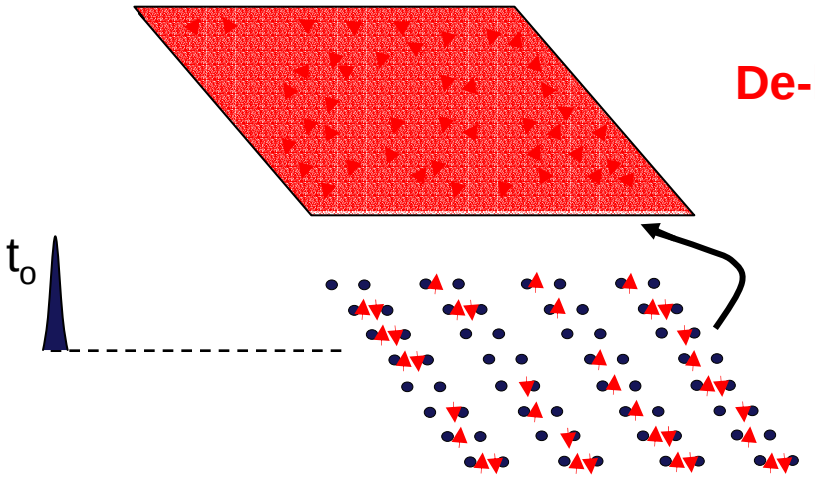


Photo-induced Insulator to Metal



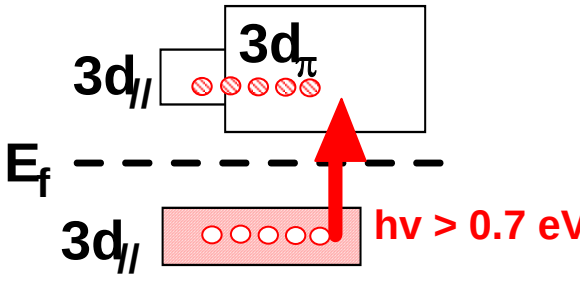
5 - 50% holes



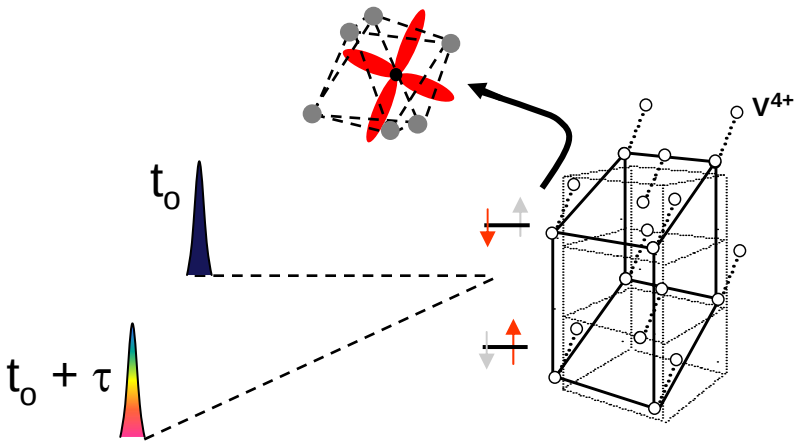
De-localised O2p-V3d bands

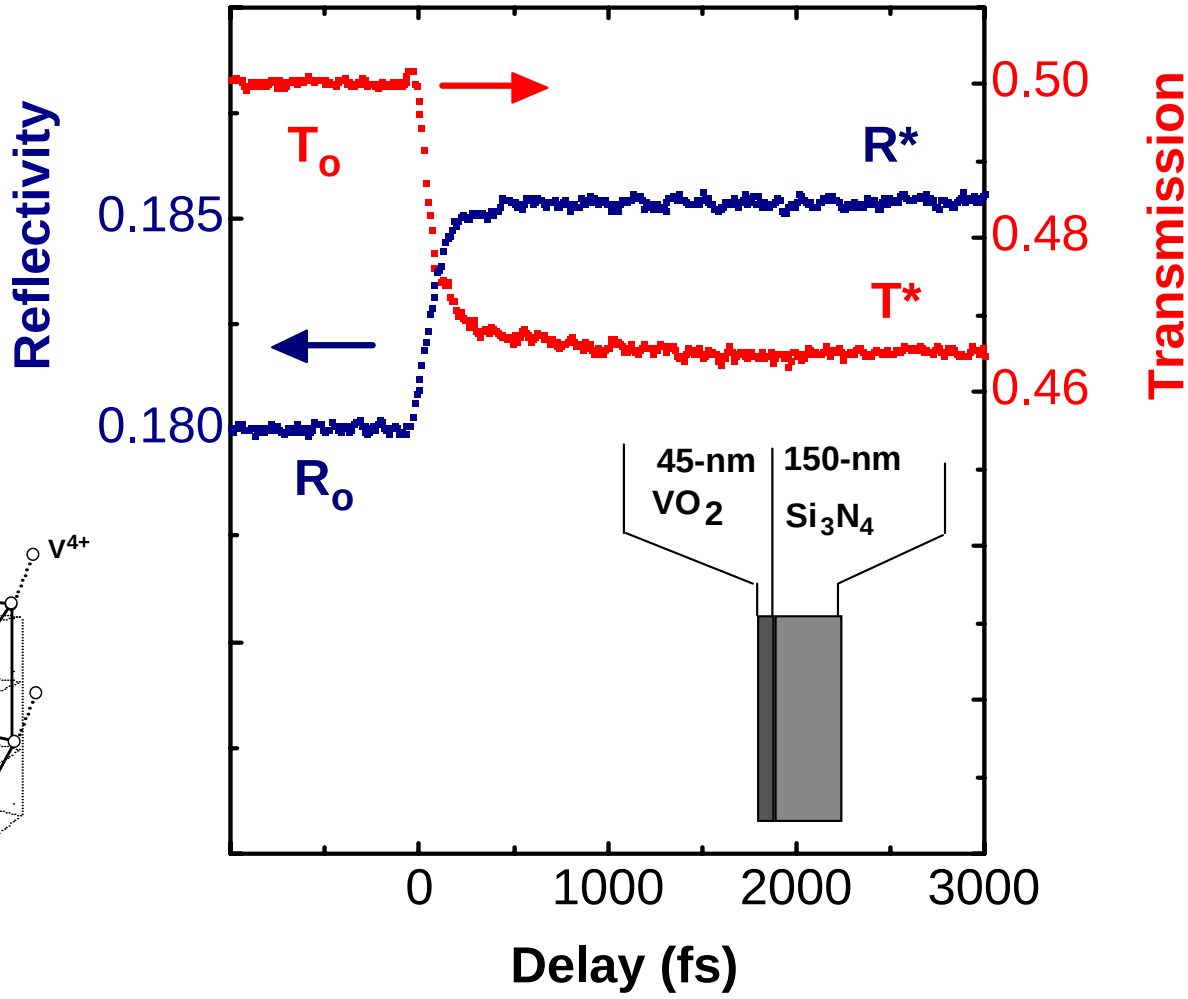
Charge-ordered localised Singlets

Photo-induced Phase Transition

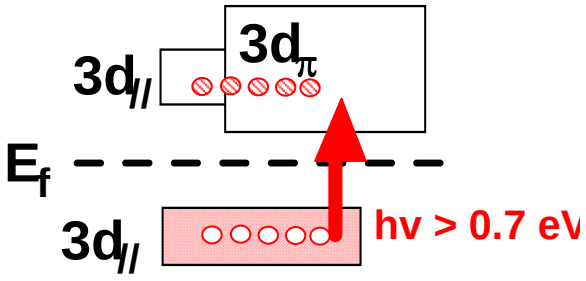


5 - 50% holes

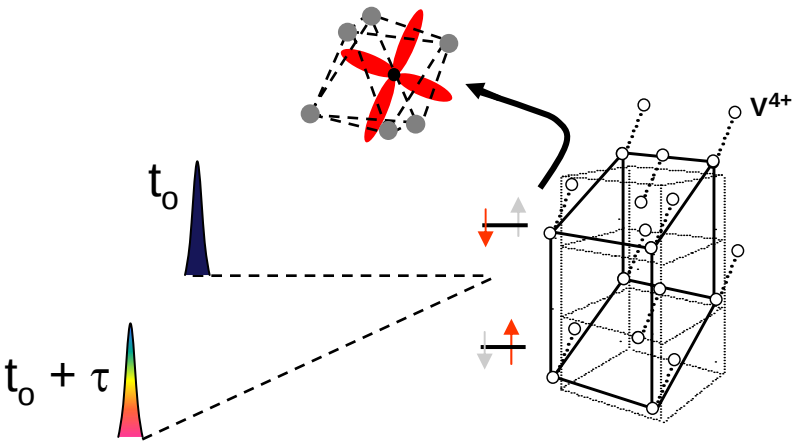


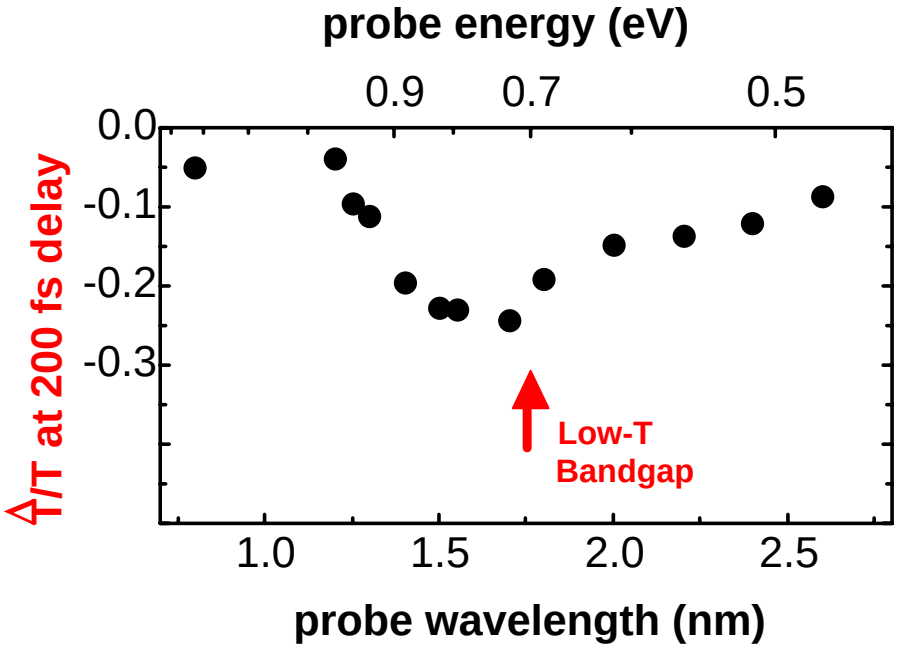


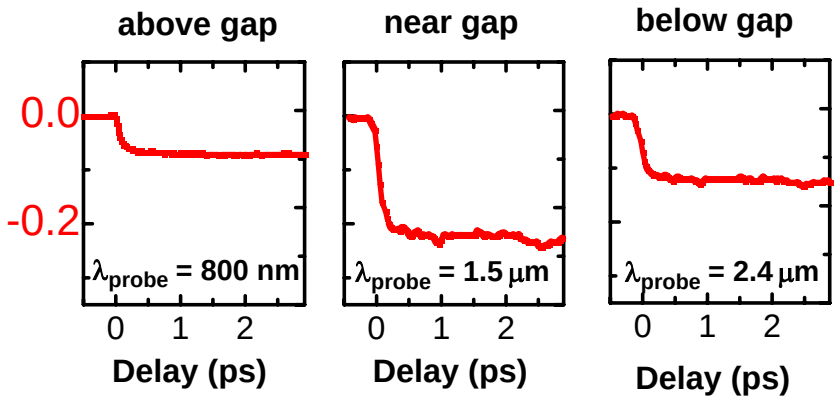
Mid-IR probing: Insulator to Metal



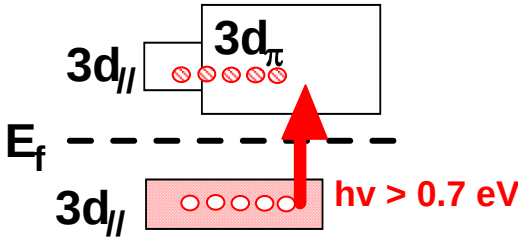
> 50% holes



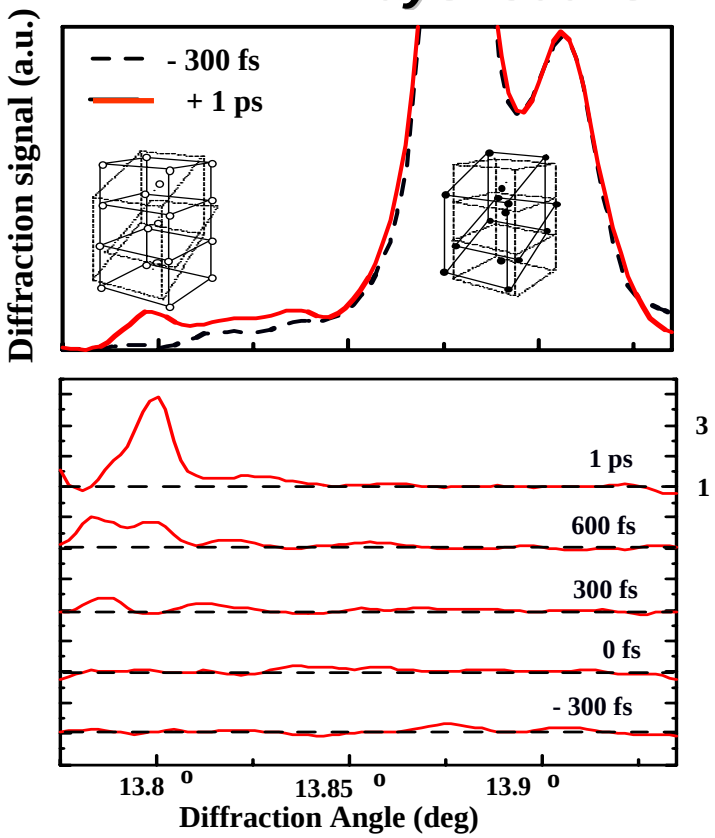




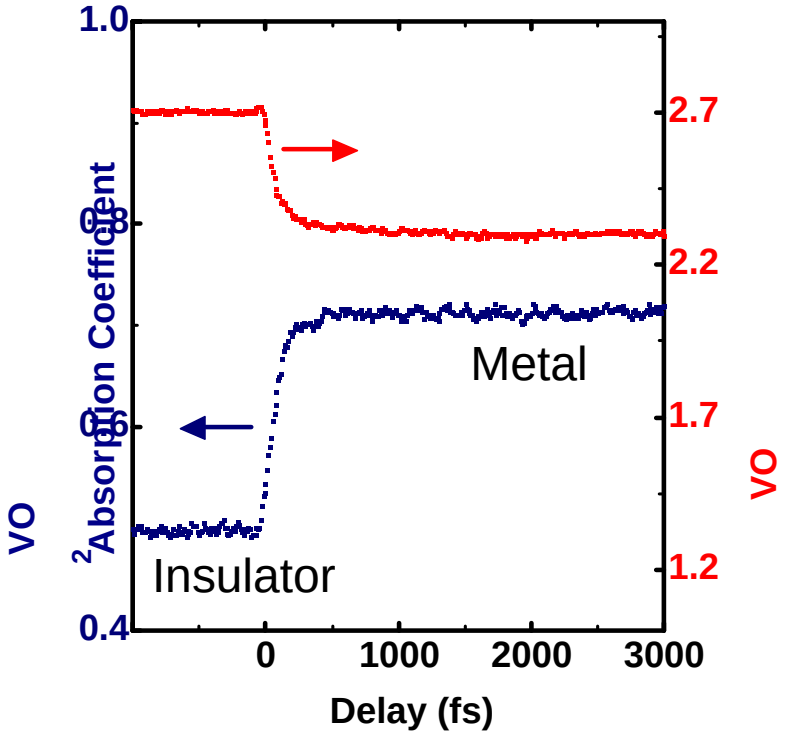
Structural and electronic transition



X-rays: 300 fs



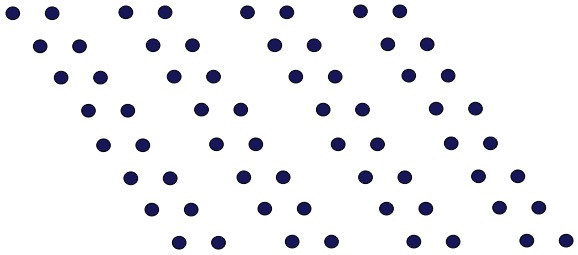
Optical: 100 fs

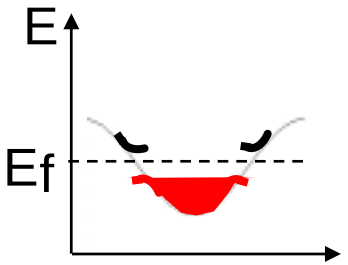


Cavalleri et al. *Phys. Rev. Lett.* 87, 237401 (2001)

Band Insulator or Mott Insulator?

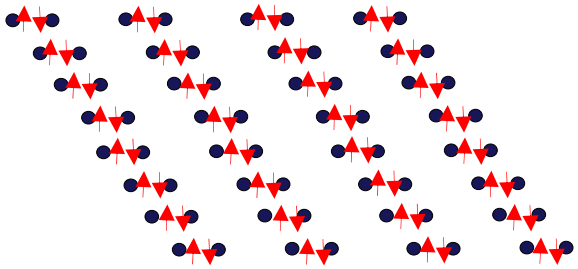
Band Insulator ?

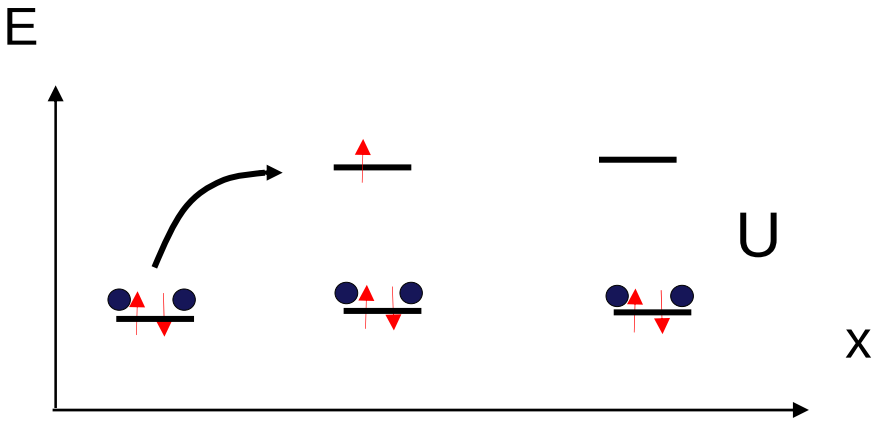




J.B. Goodenough *Phys. Rev. 117*, 1442 (1960); Wentzcowitch et al. *Phys. Rev. Lett. 72*, 3389 (1994)

Mott Insulator ?

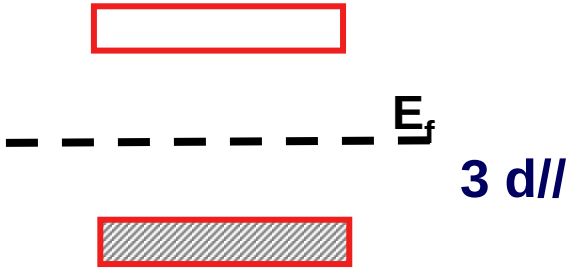


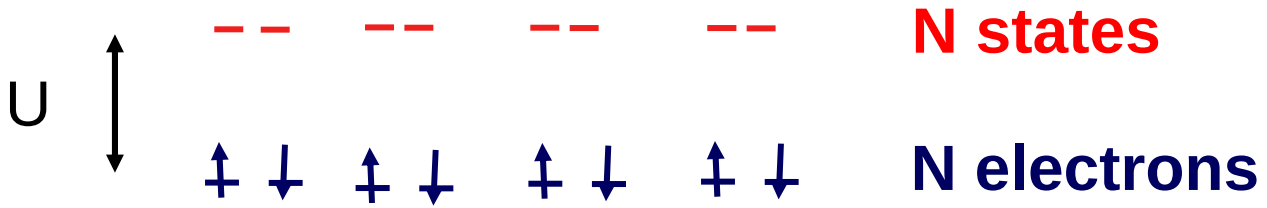


Zylbersztejn and N. Mott *Phys. Rev. B* 11, 4383 (1975)

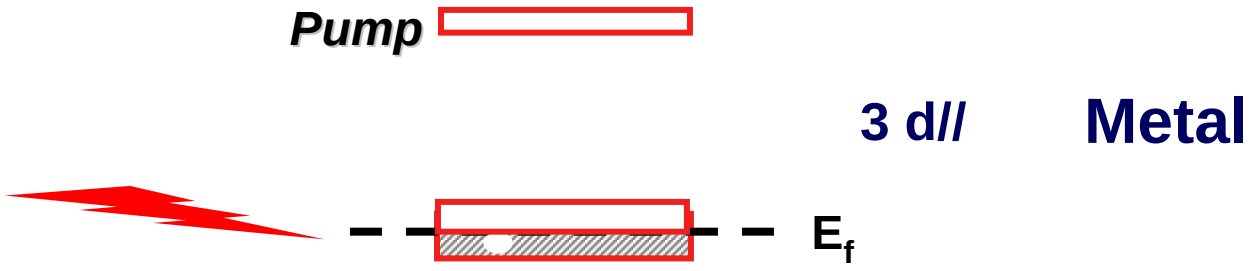
Low T Phase: Mott Insulator?

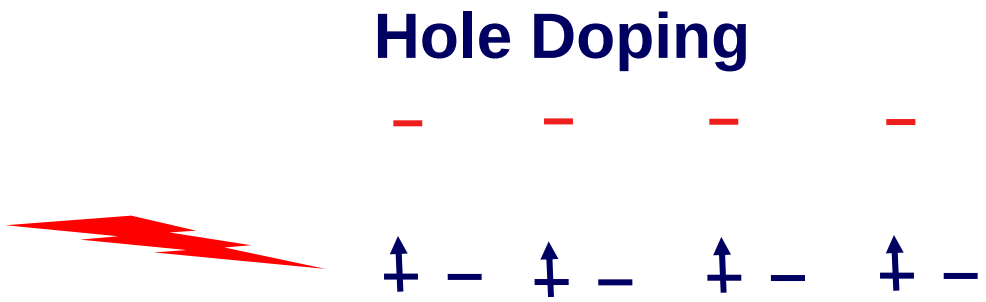
Mott Insulator ?



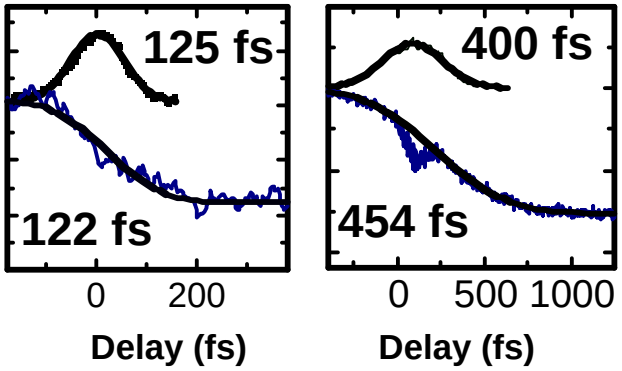


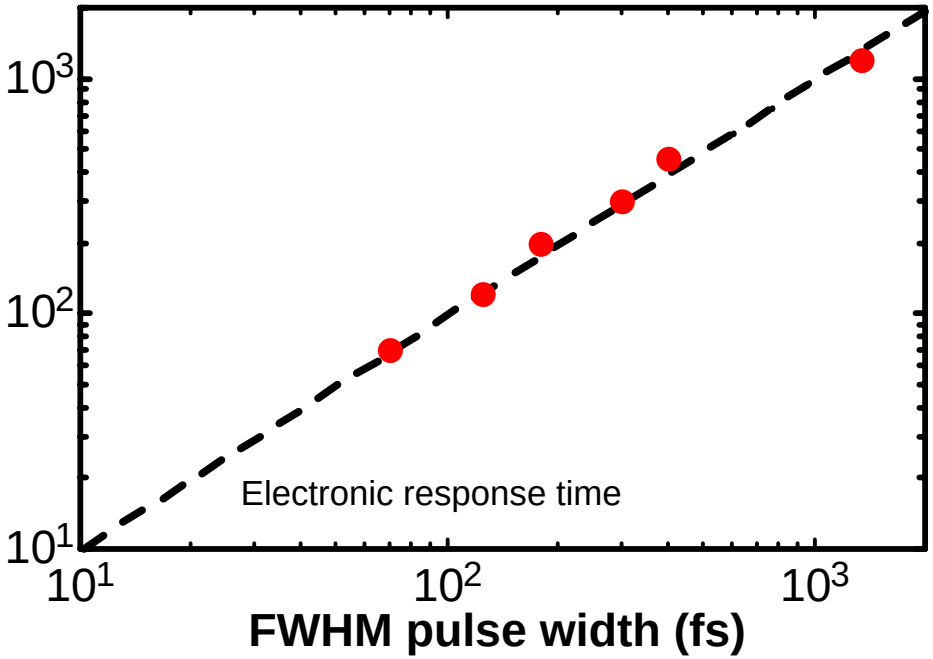
Mott Transition: Instantaneous



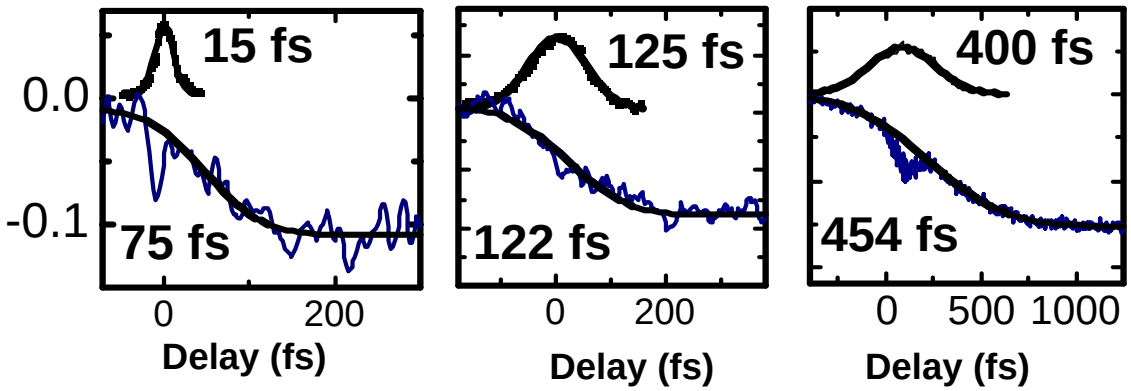


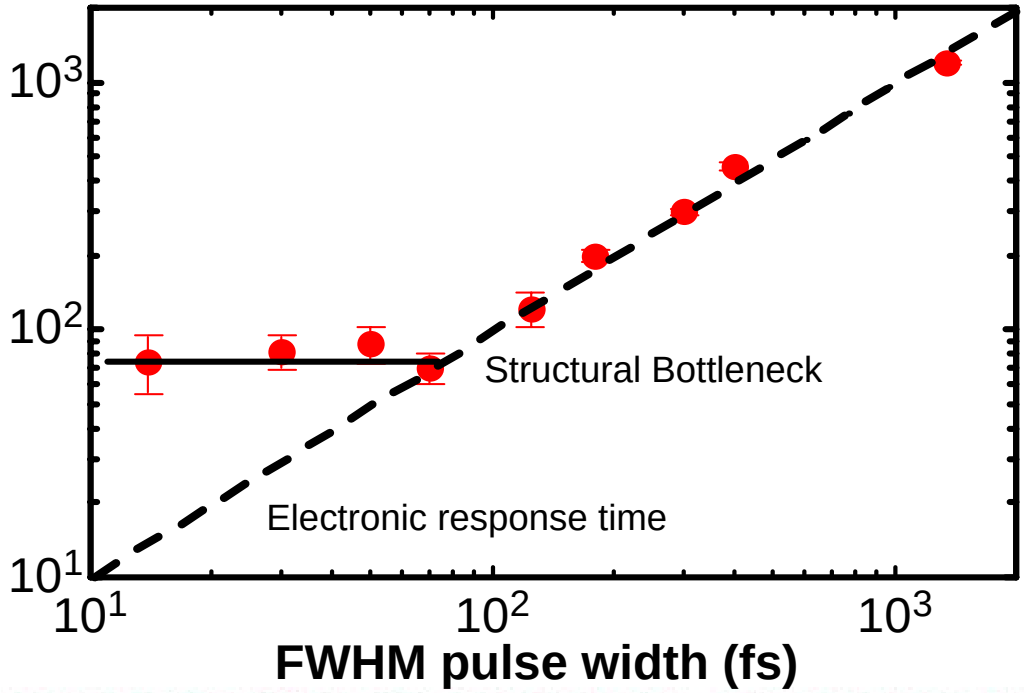
Phase Transition Time: 75 fs



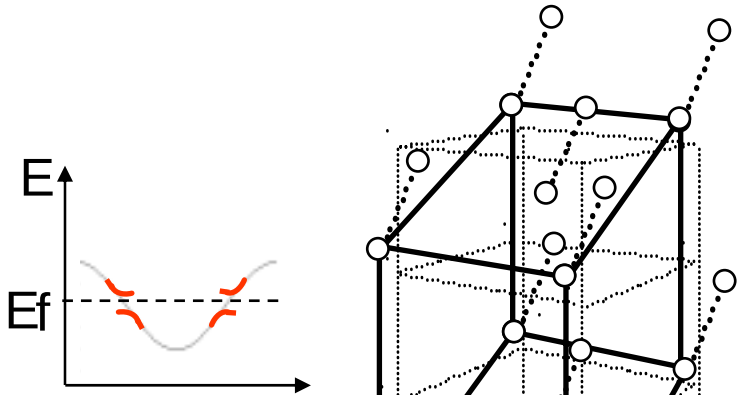


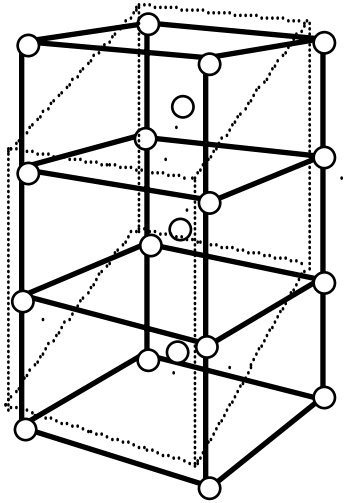
Phase Transition Time: 75 fs

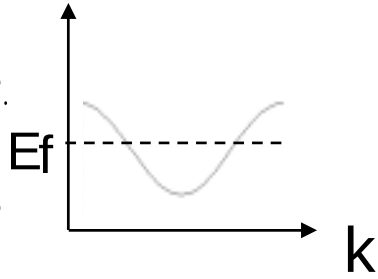




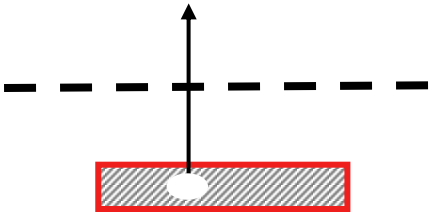
Reverse Peierls Transition: 100 fs

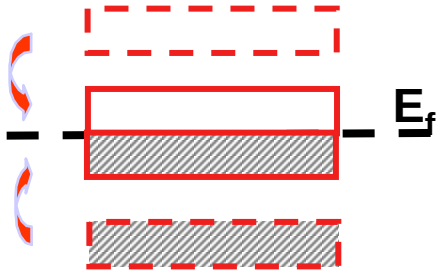






Optical Excitation



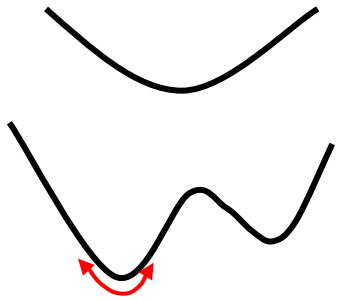


3 d//

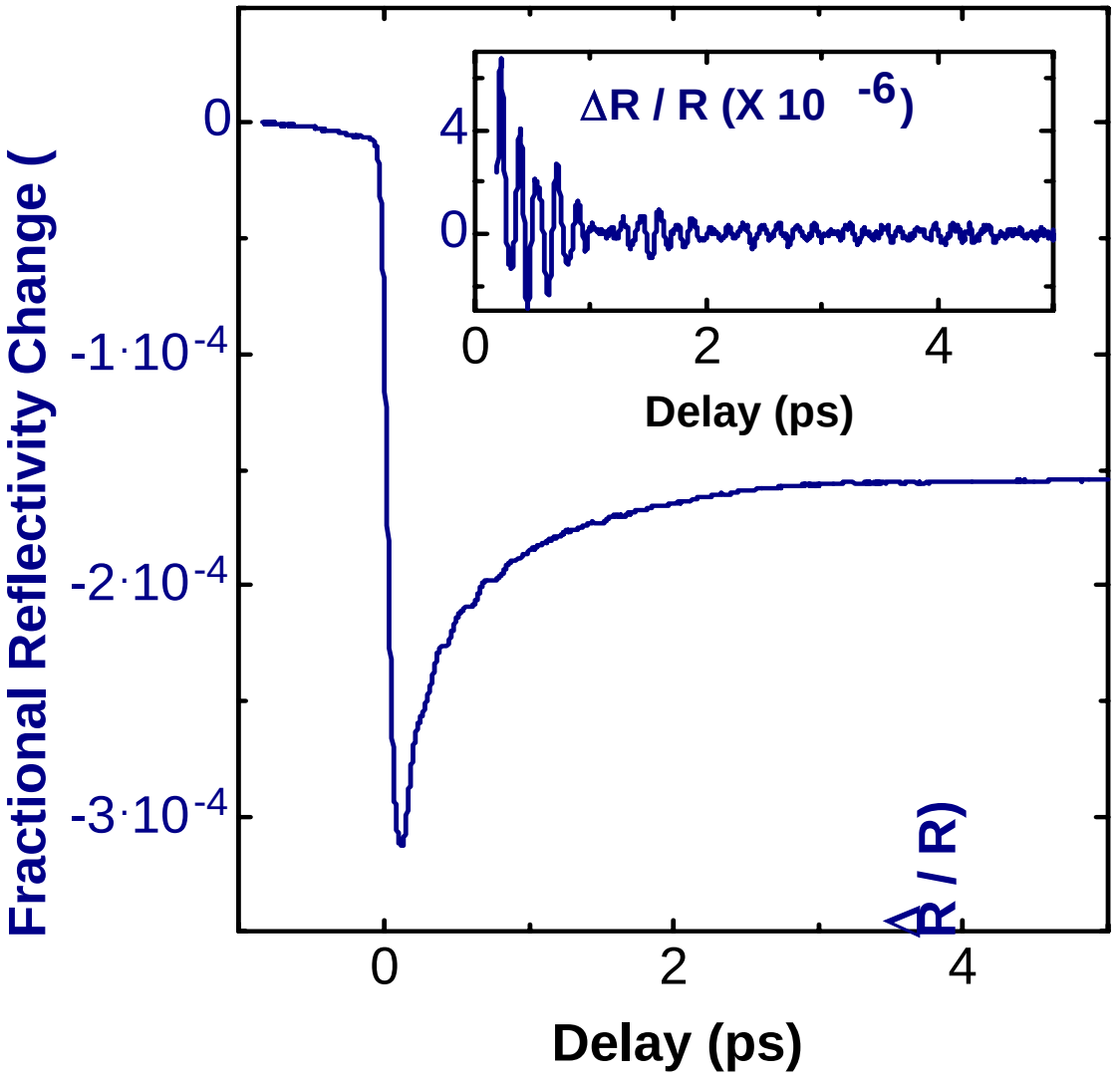
Impulsive excitation of Optical Phonons

Low fluence

No Phase transition

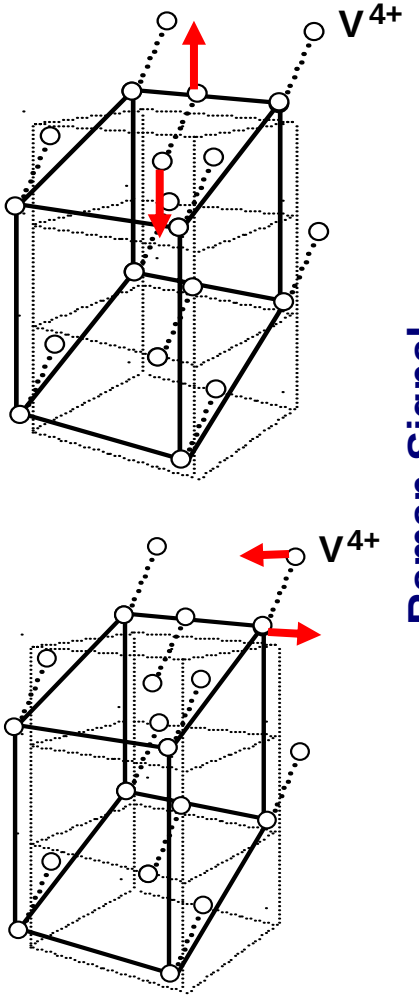


Monoclinic Rutile

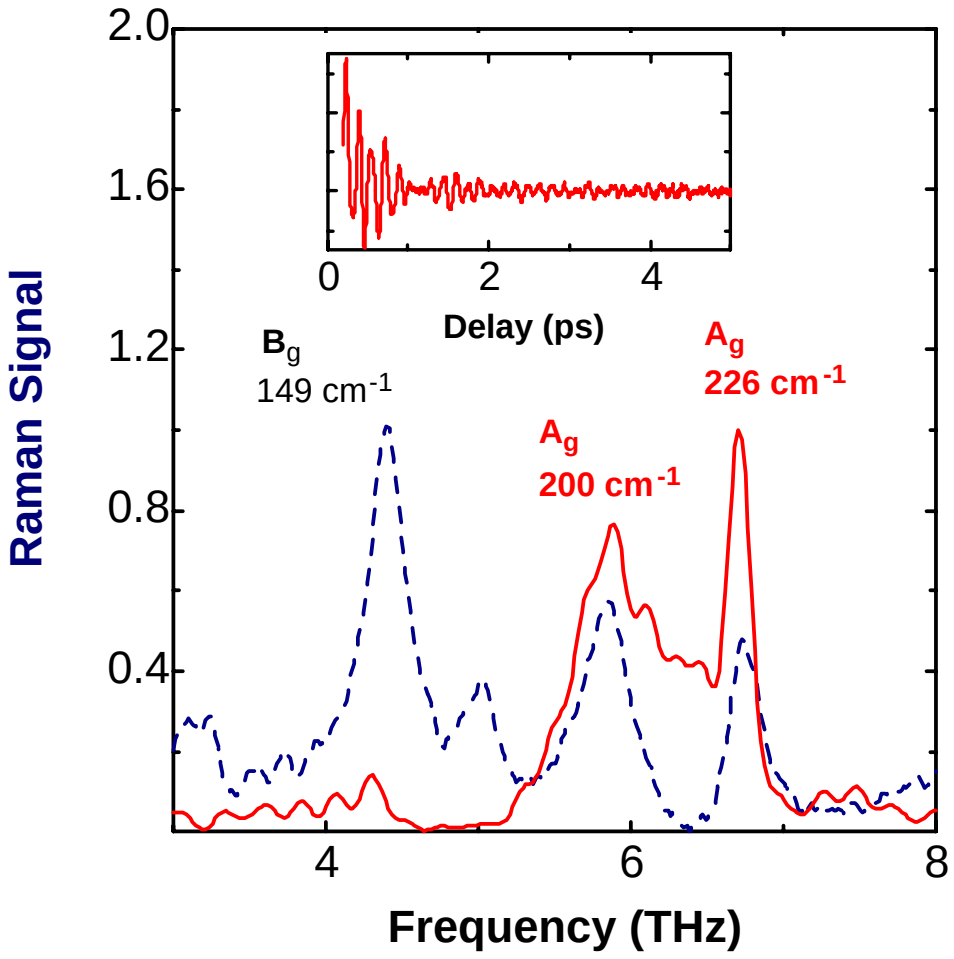


Excitation of symmetry-breaking modes

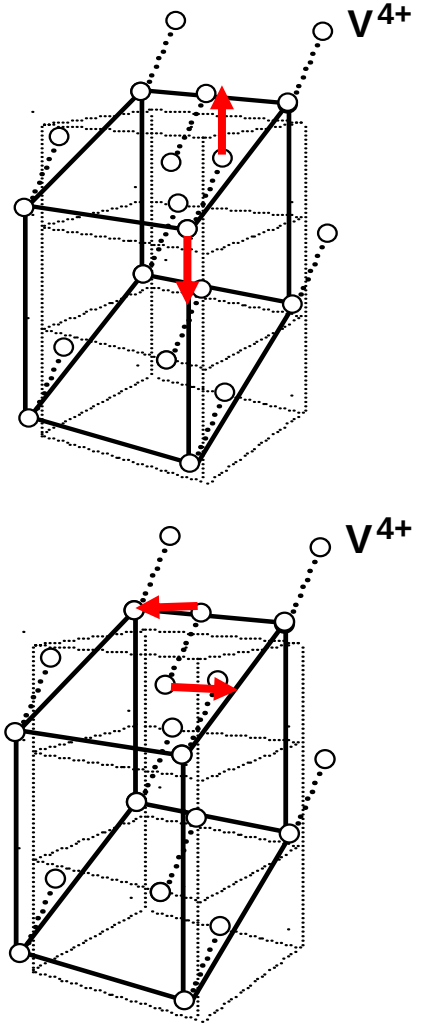
Mode 200 cm^{-1}



Impulsive Raman

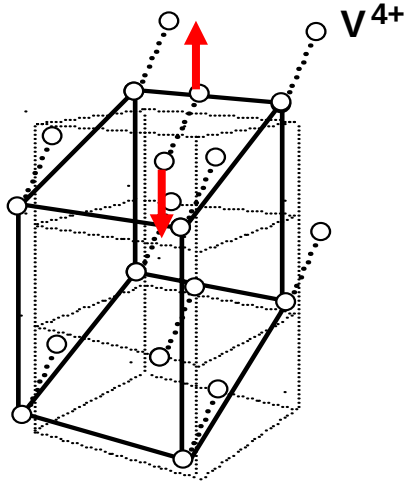


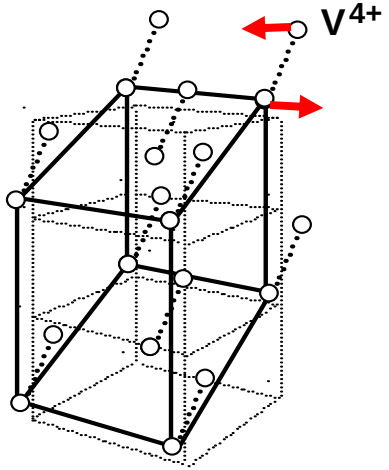
Mode 226 cm^{-1}

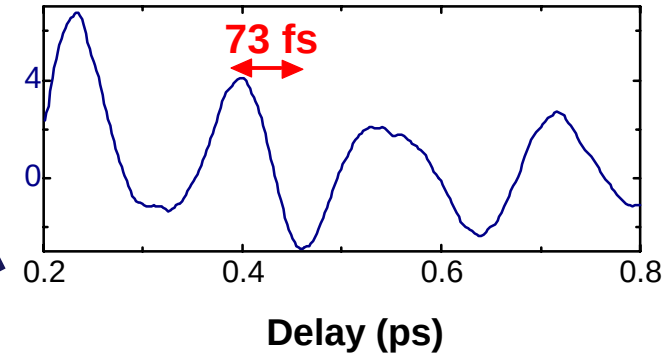


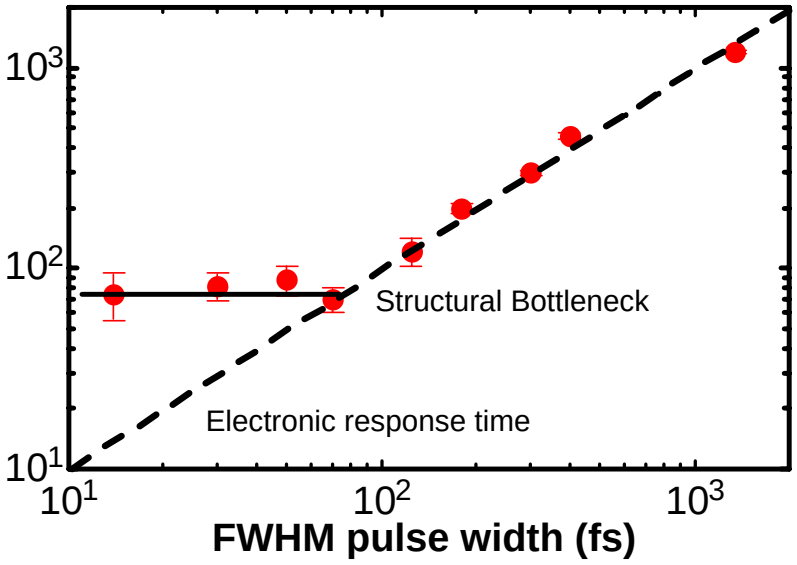
Excitation of symmetry-breaking modes

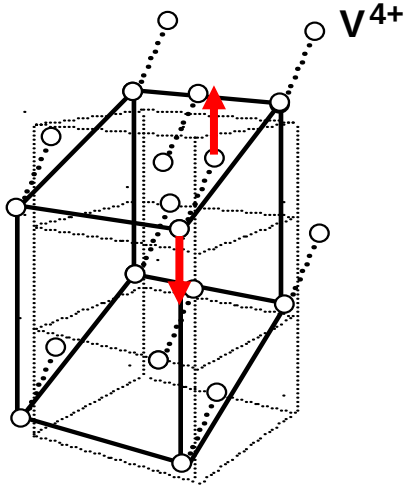
Impulsive Raman

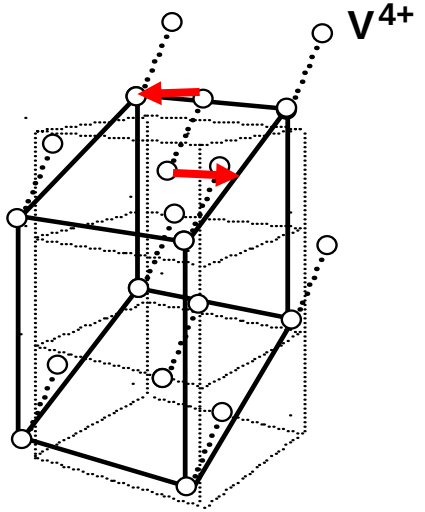






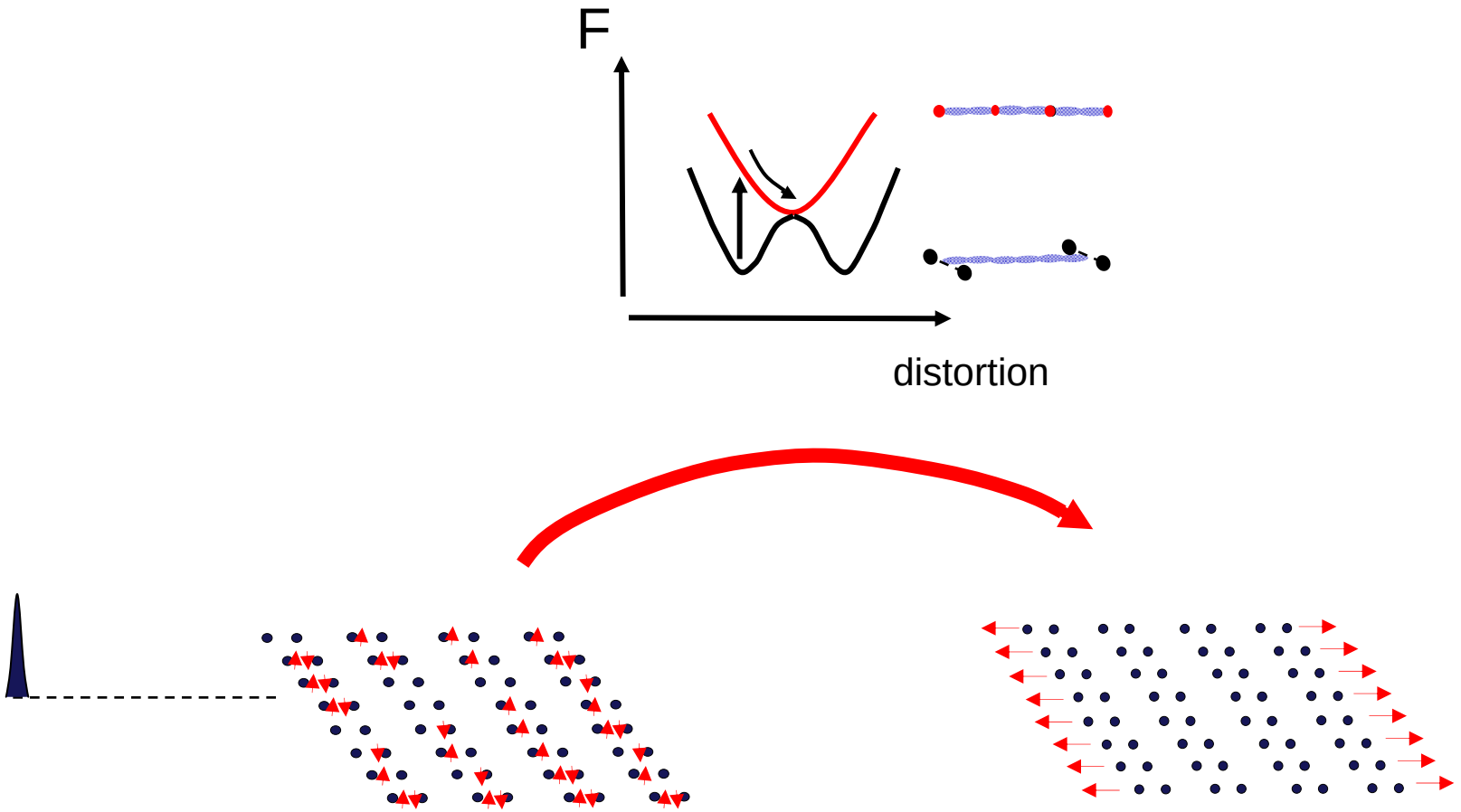






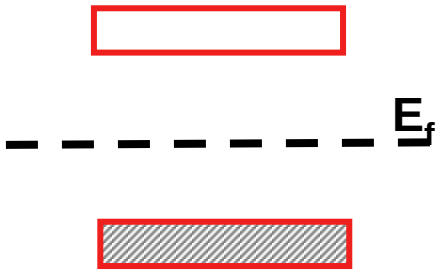
Cavalleri et al. cond-mat/0403214

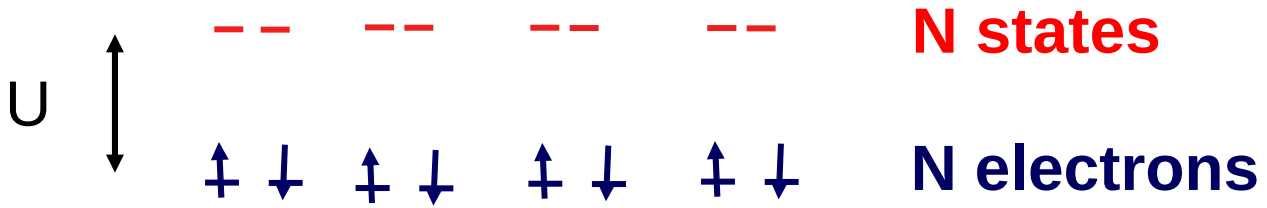
Photo-doping into a CES



Non-rigid Band Structure

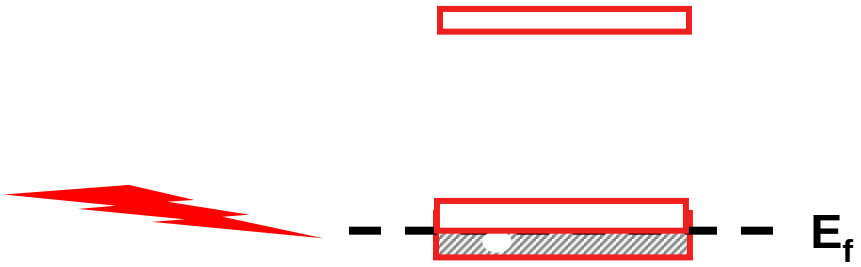
Correlated Electrons



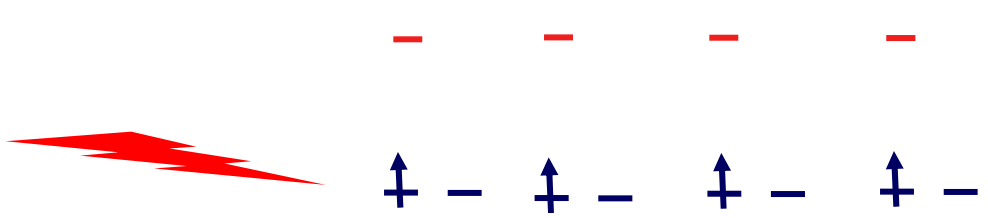


Non-rigid Band Structure

Correlated Electrons

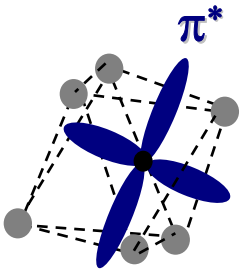


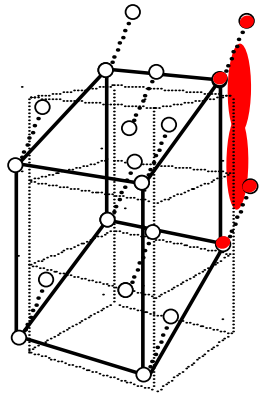
Hole Doping



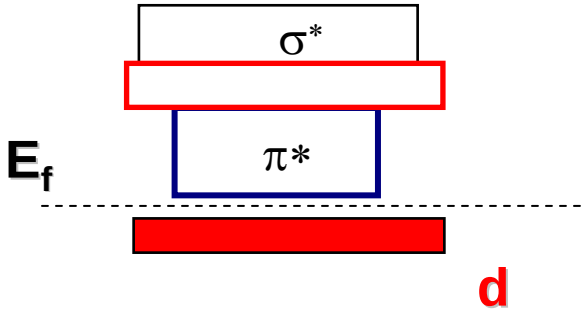
Electronic Structure: NEXAFS Spectrum

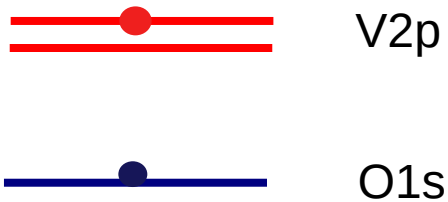
Orbitals



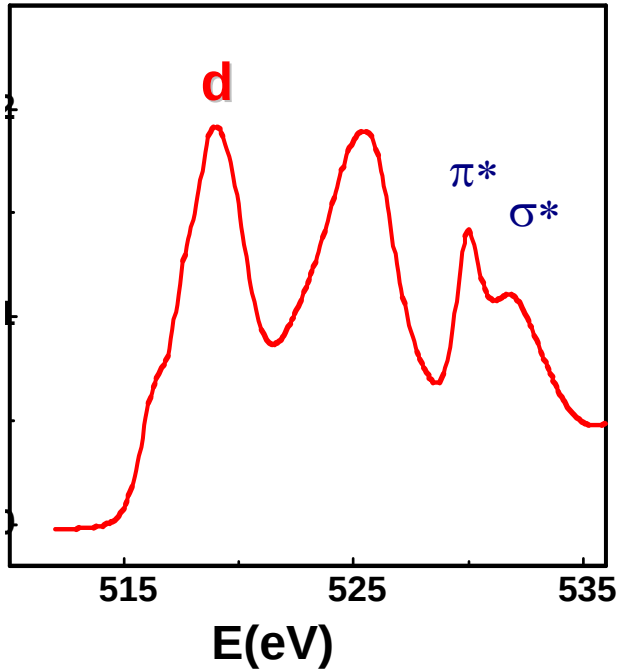


Band Structure



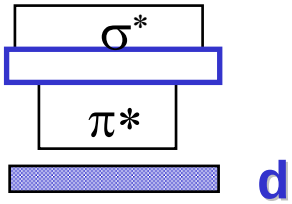


NEXAFS Spectrum

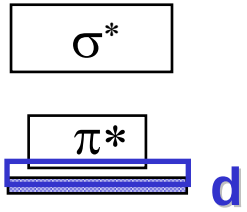


Temperature driven transition

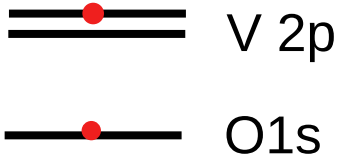
Insulator



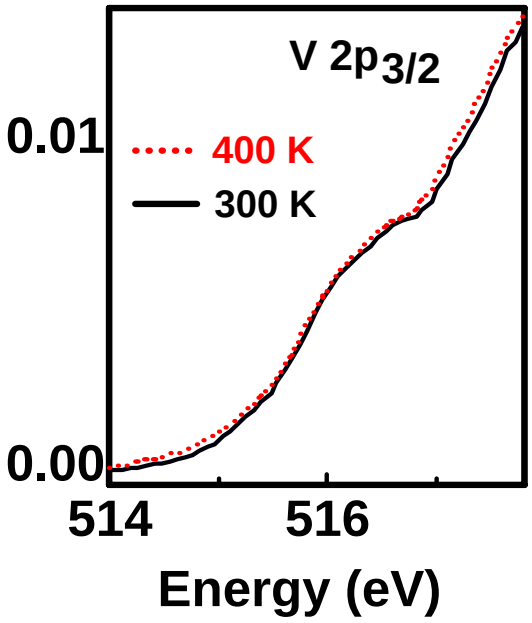
Metal



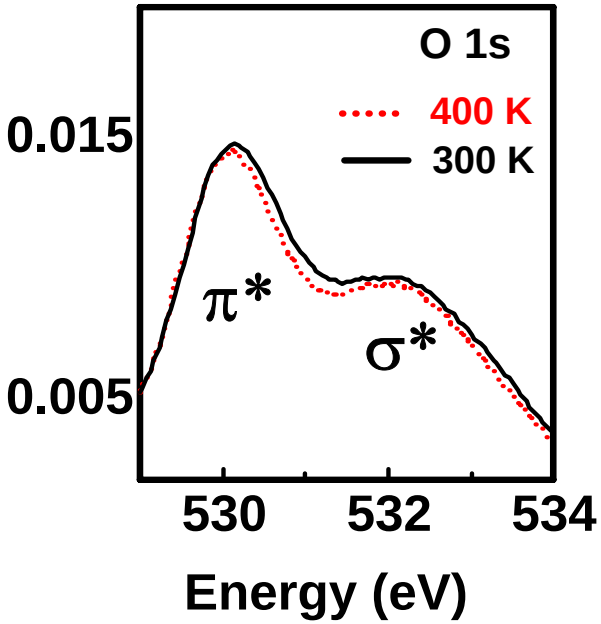
hν



d orbitals

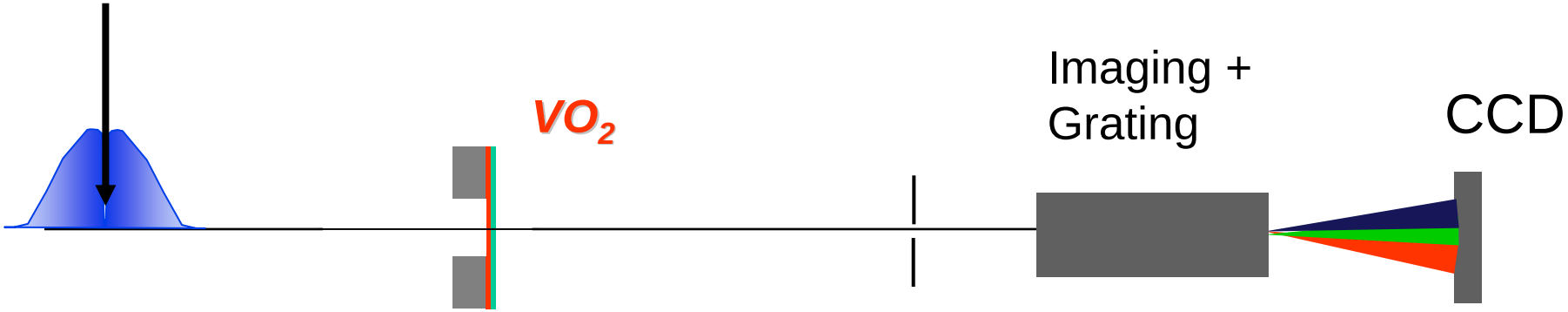


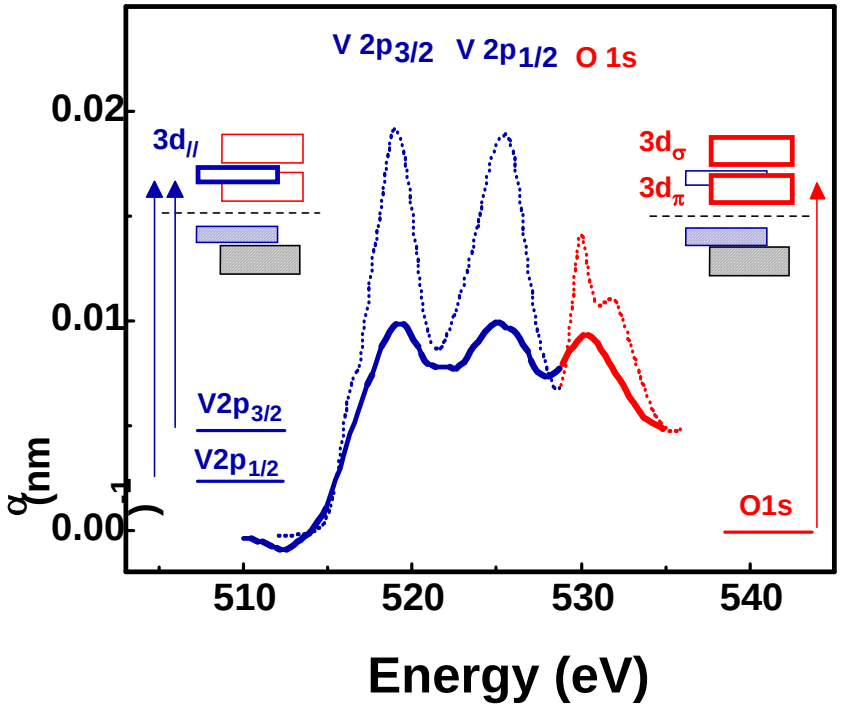
π orbitals



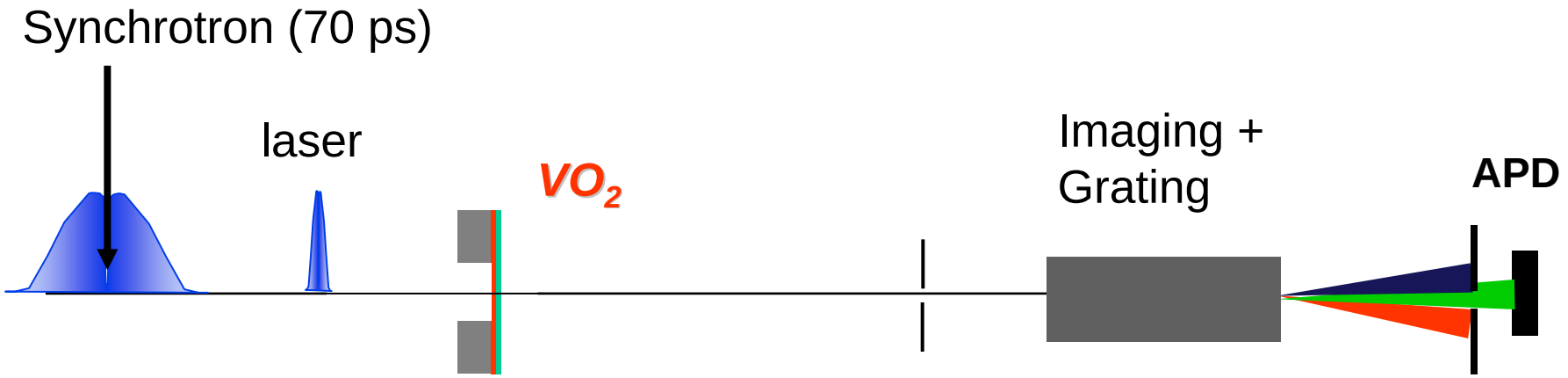
NEXAFS: Beamline 5.3.1

Synchrotron: 70 ps

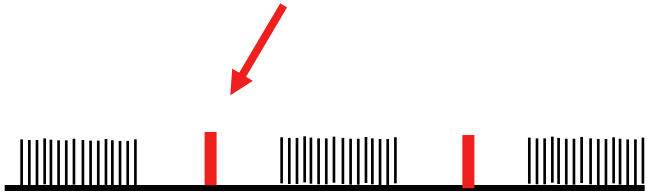




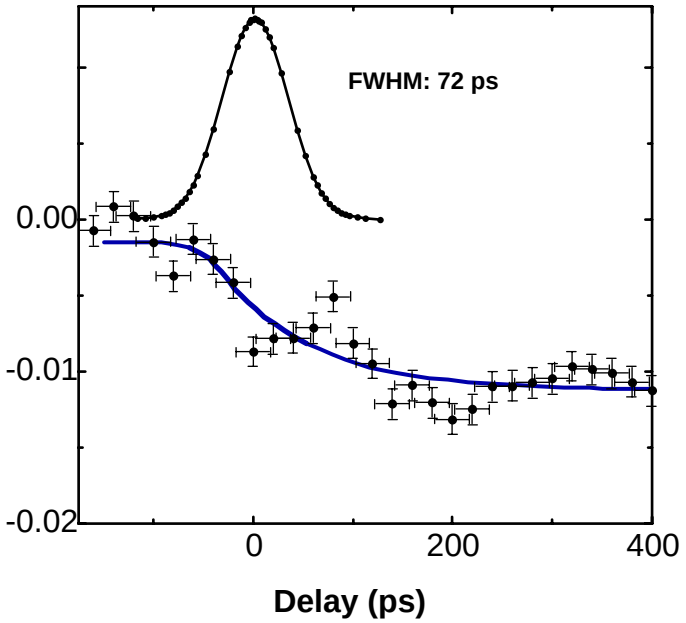
Time-resolved NEXAFS: ps scale



Gating @ 1 KHz

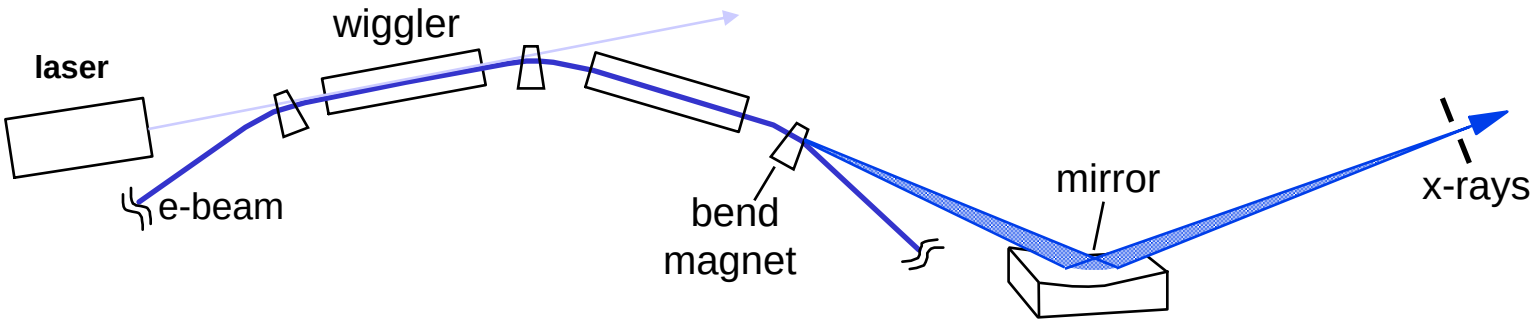


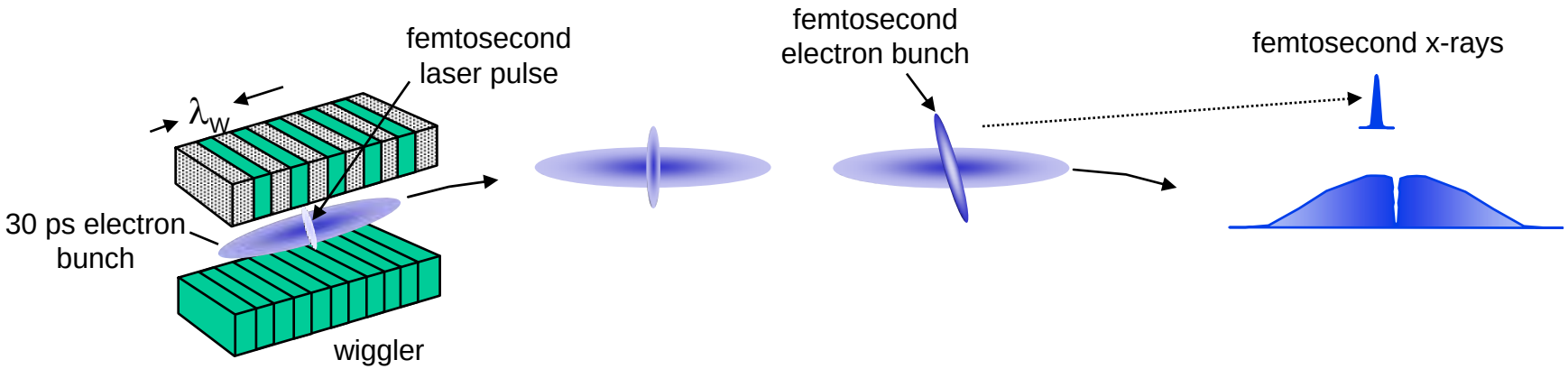
10^8 photons/(sec 1%BW)



Cavalleri et al., Physical Review B 69, 153106 (2004)

Tunable femtosecond X-rays at the ALS



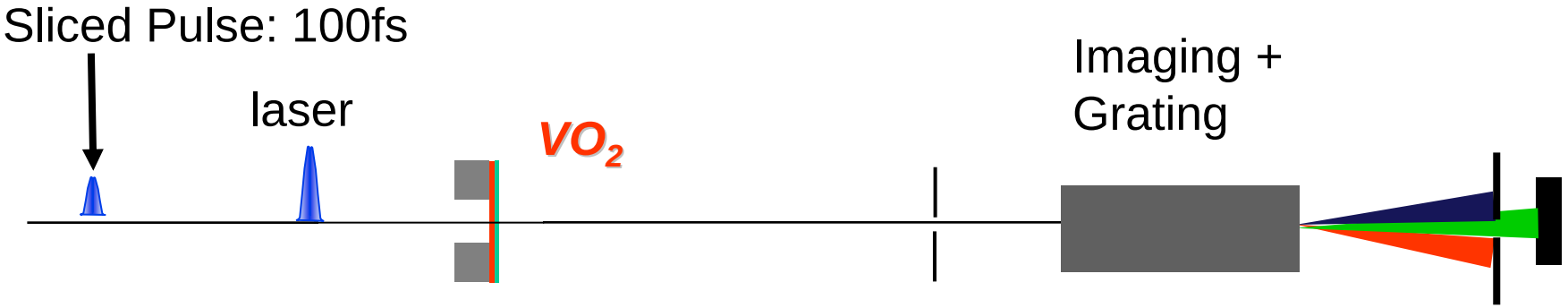


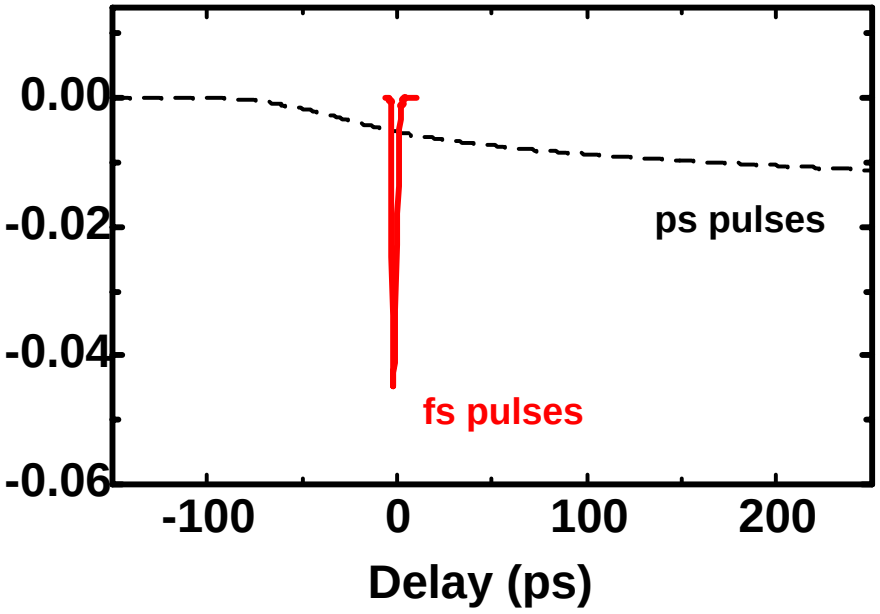
electron-photon
interaction in wiggler

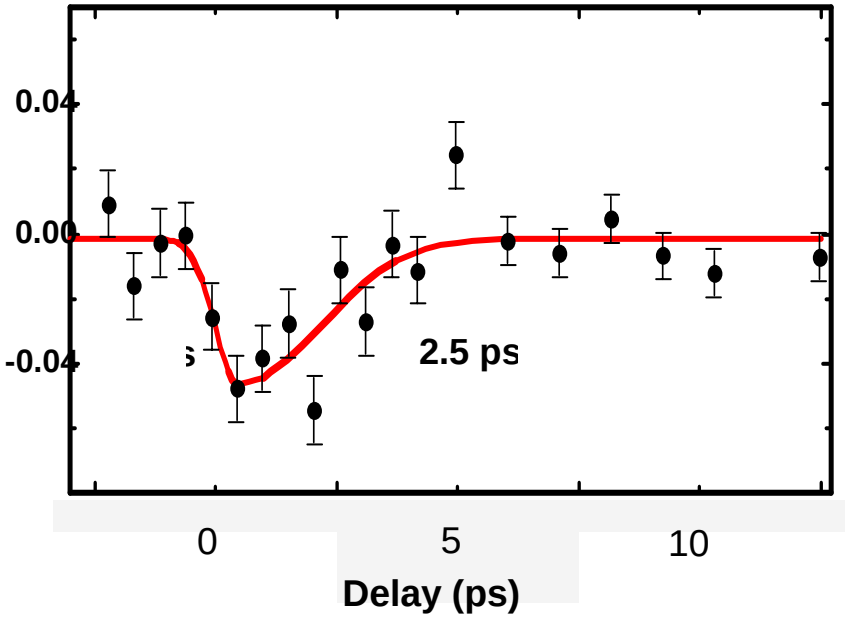
Zholents and Zolotorev, *Phys. Rev. Lett.*, 76, 916,(1996).

Schoenlein et al., *Science,* 287, (2000)

Transient Over Absorption

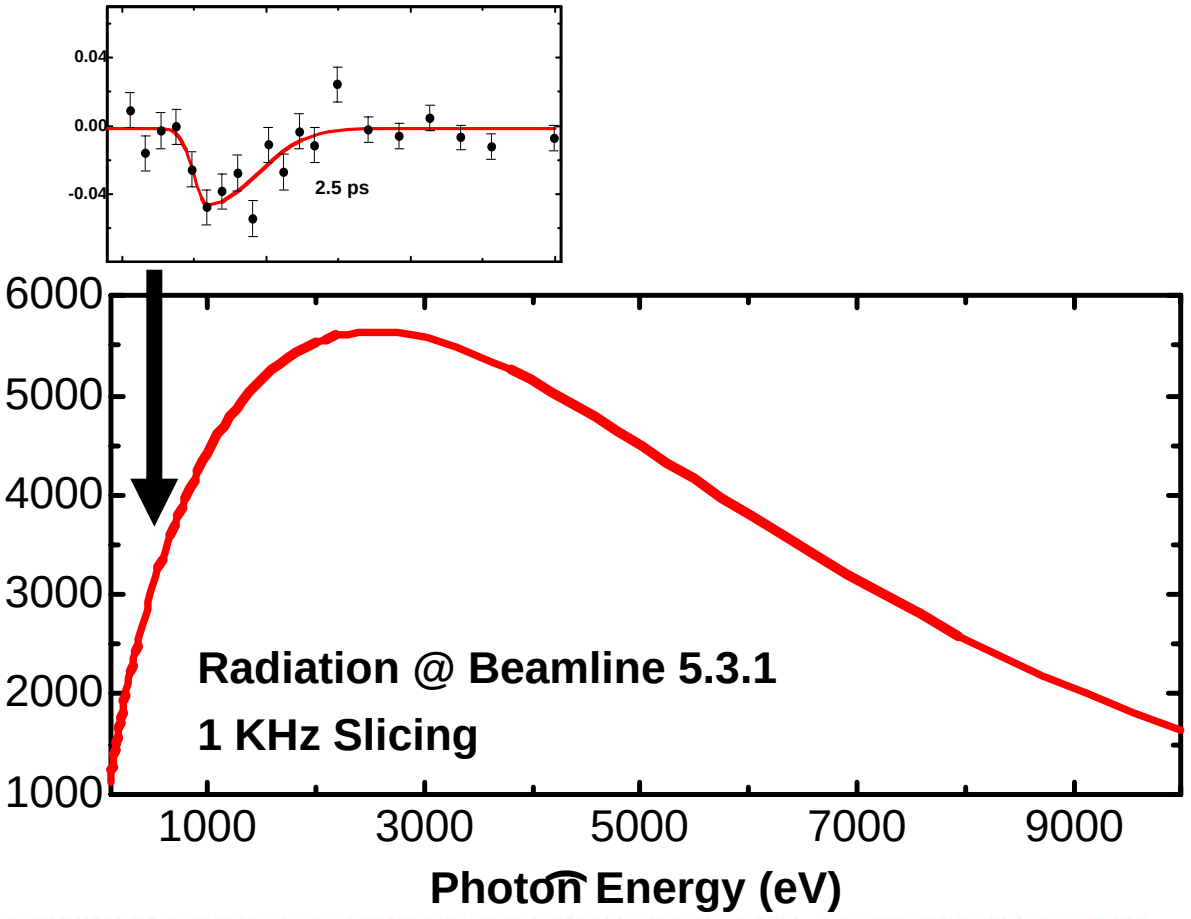




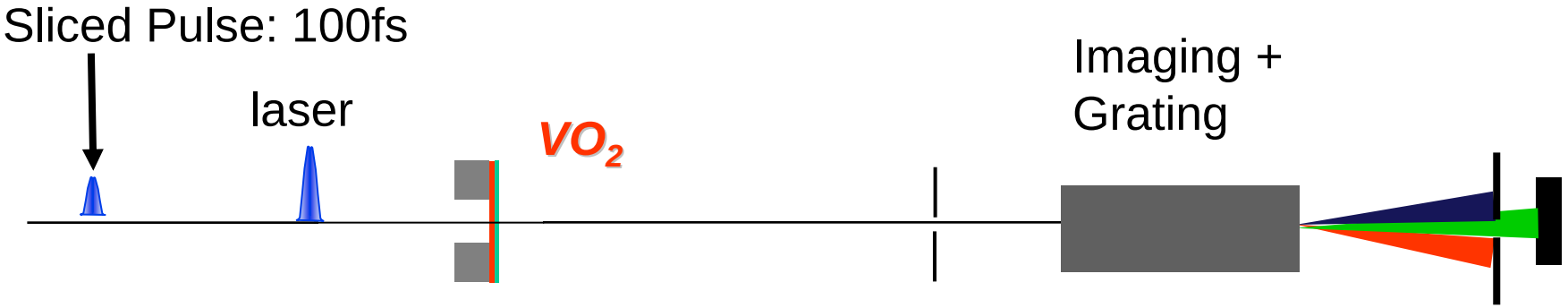


Technical Significance

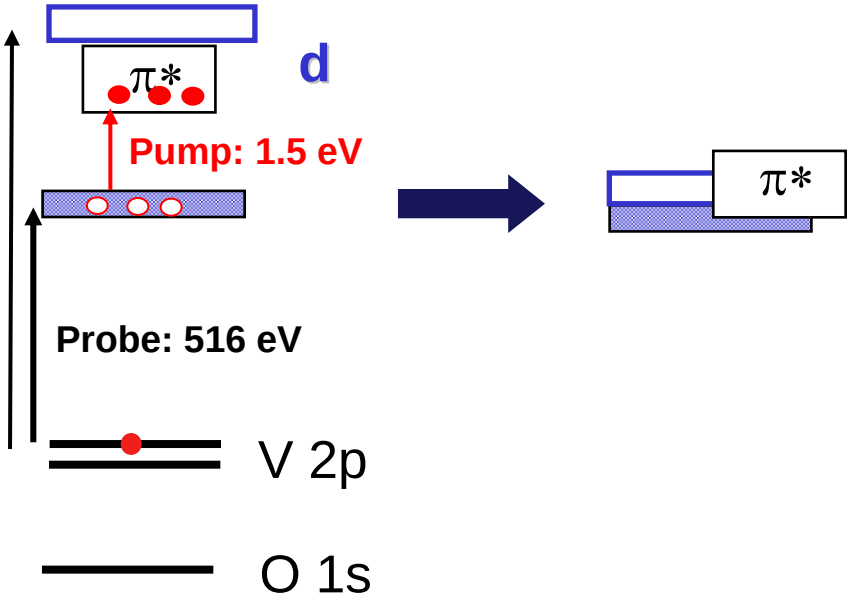
First femtosecond X-ray measurement with a fully tunable synchrotron beamline at 500 eV



Over Absorption: Hole photo-doping

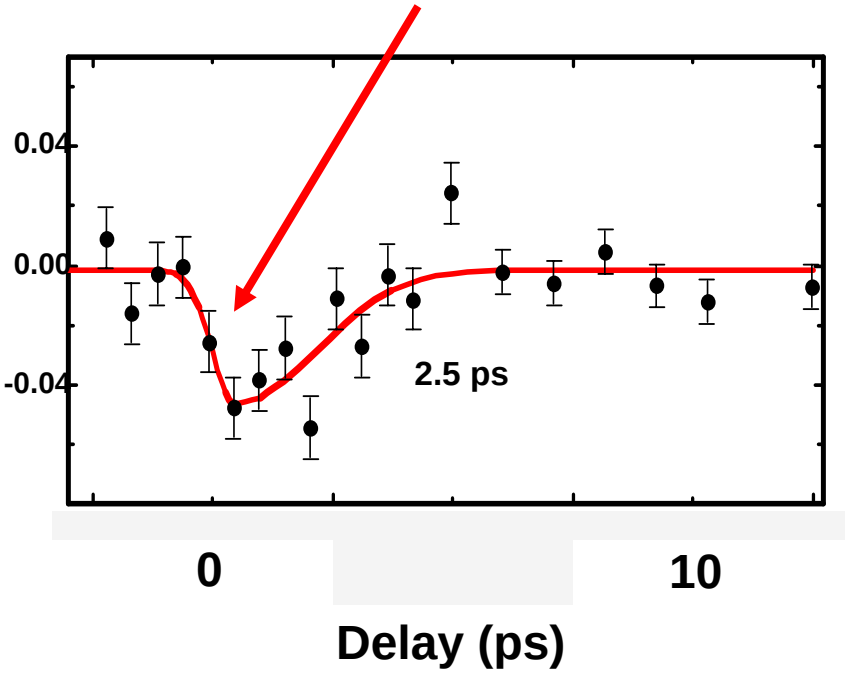


Hole Photo-doping

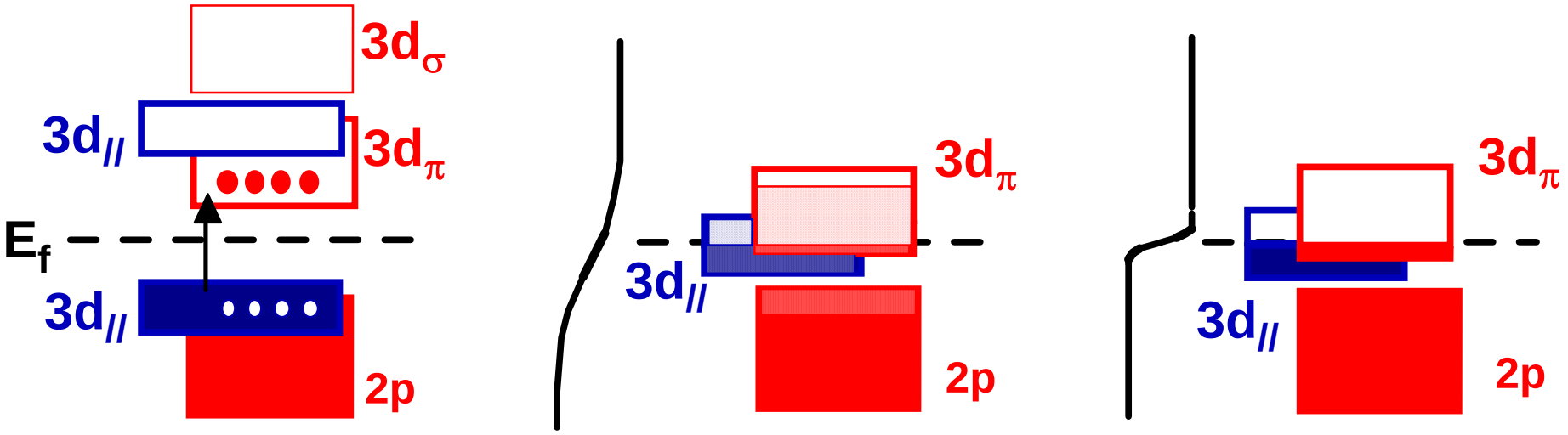


Metal

Hole photo-doping



Insulator-to-metal Transition



Insulating phase

Photo-doping

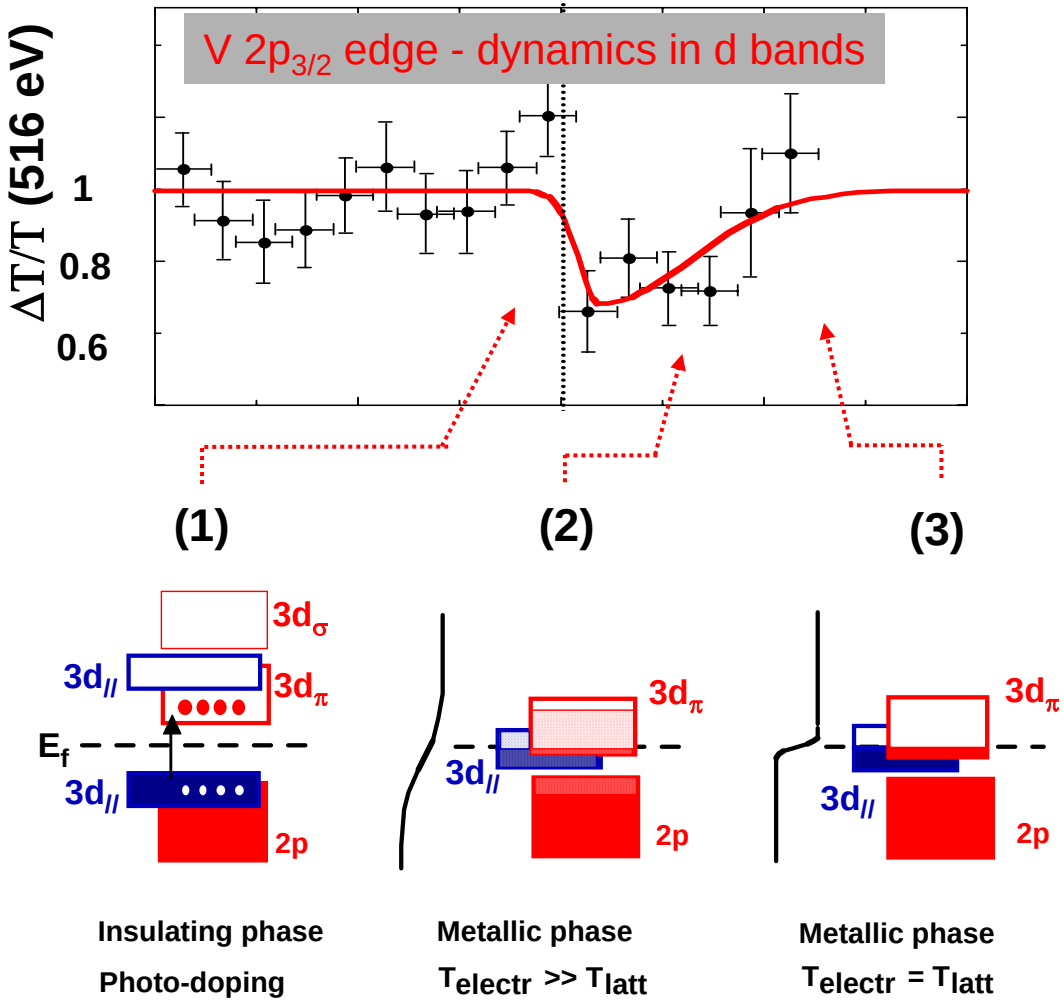
Metallic phase

$T_{electr} >> T_{latt}$

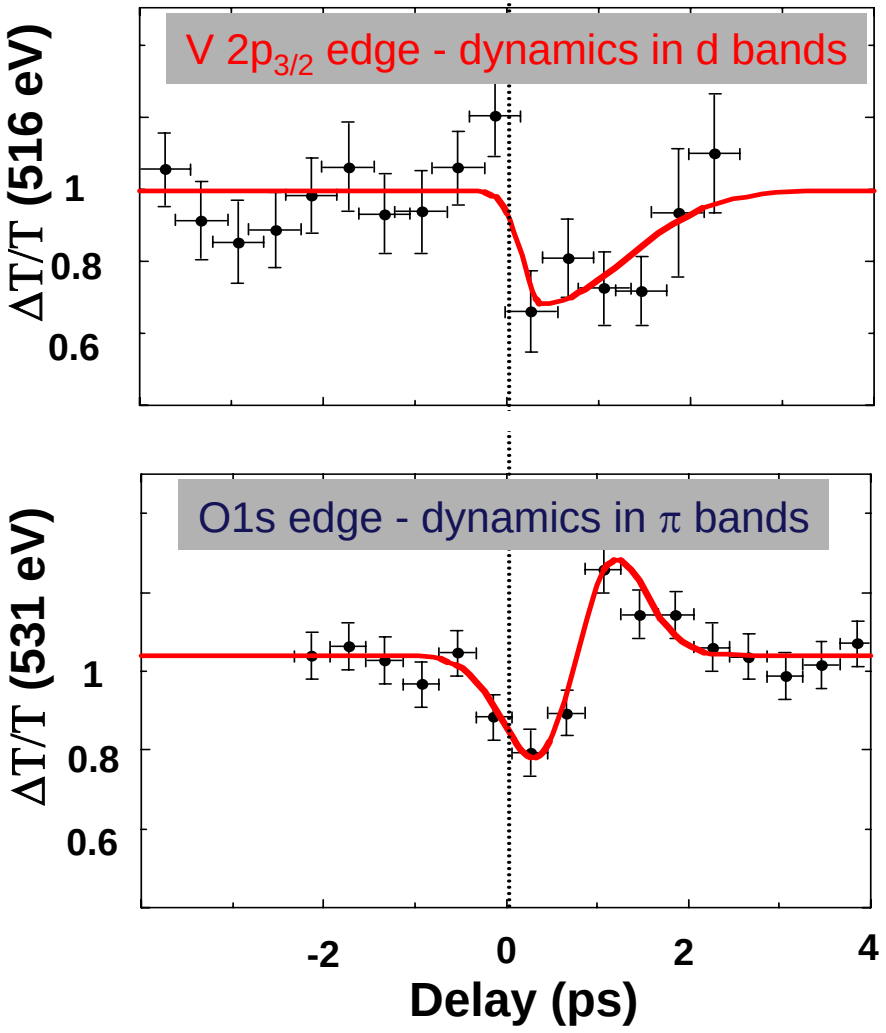
Metallic phase

$T_{electr} = T_{latt}$

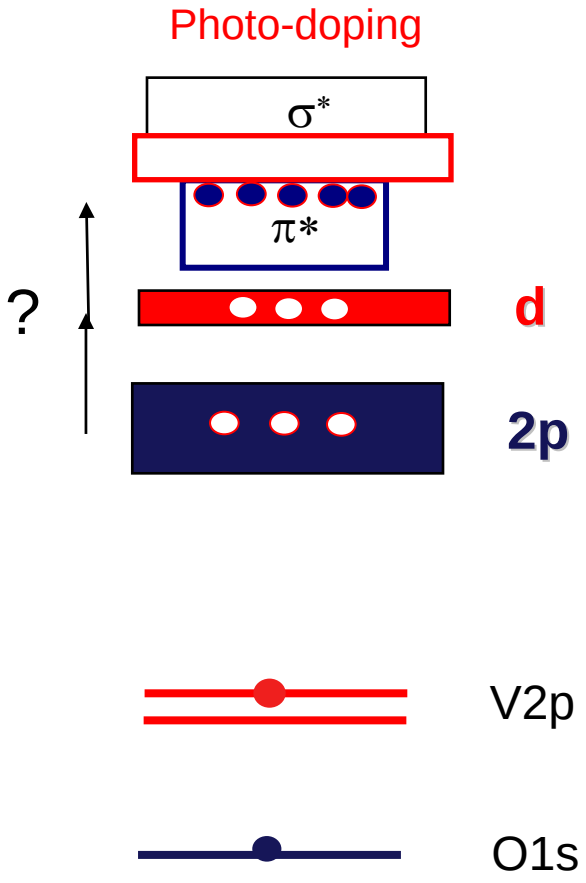
Hole Photo-doping

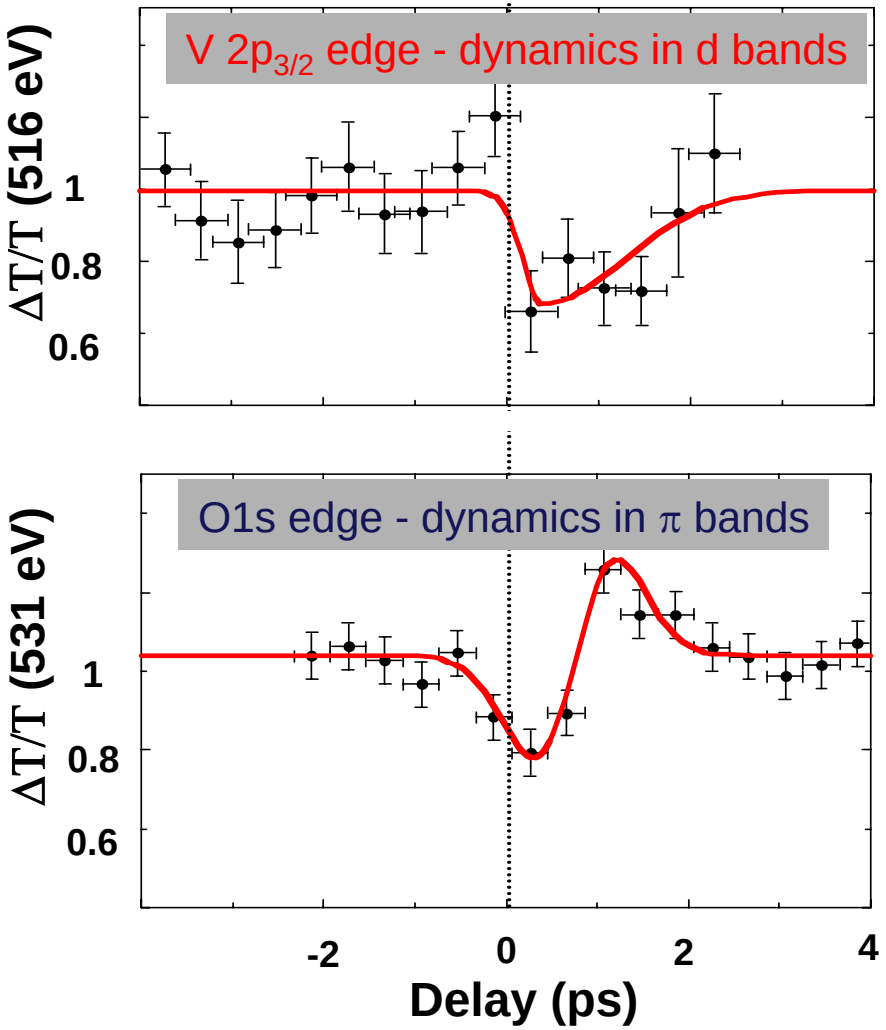


Oxygen Edge: p bands

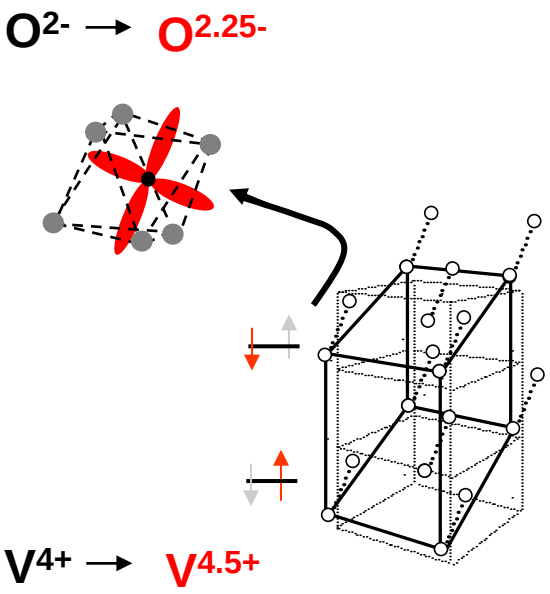


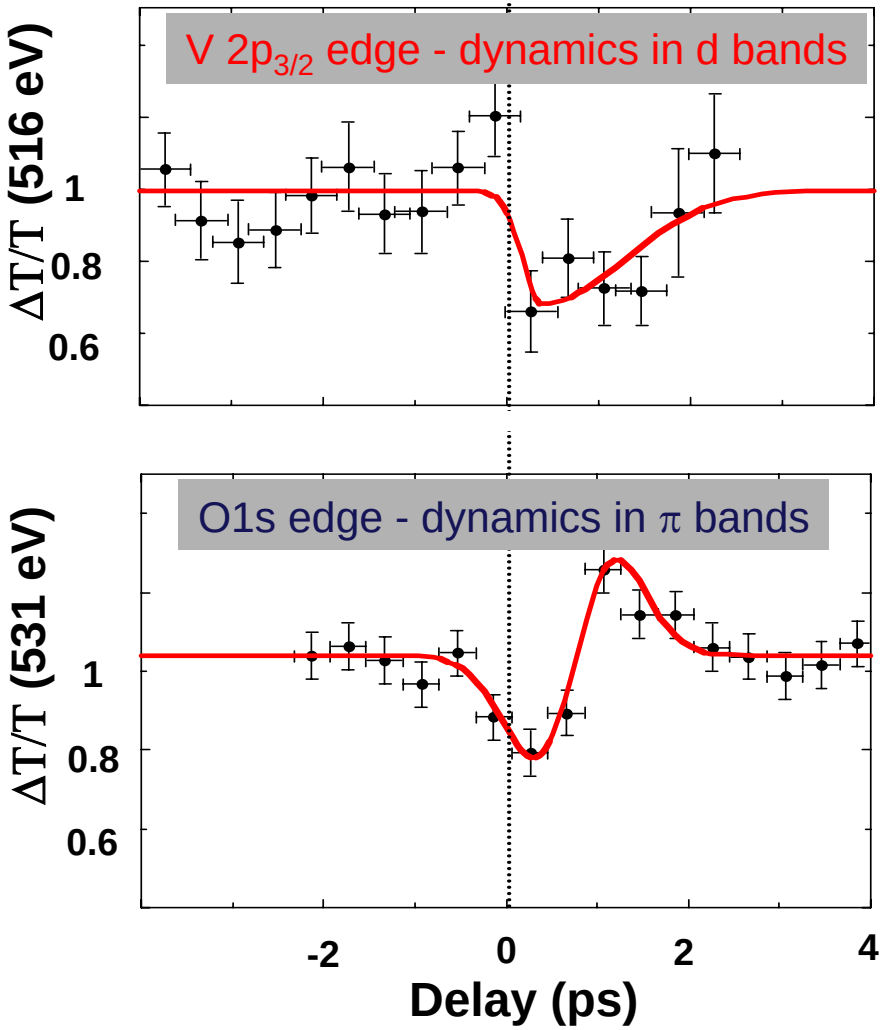
2-Photon Absorption ?



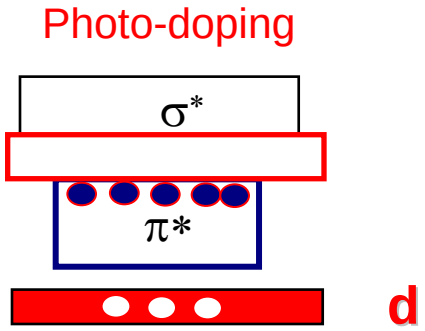


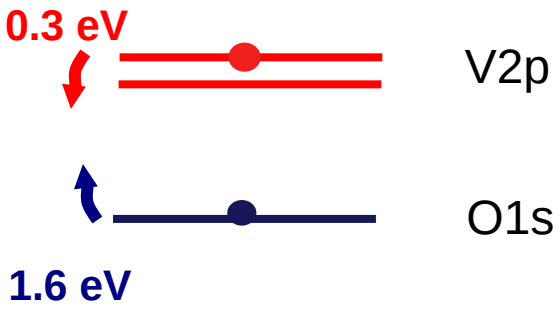
Valency Change ?

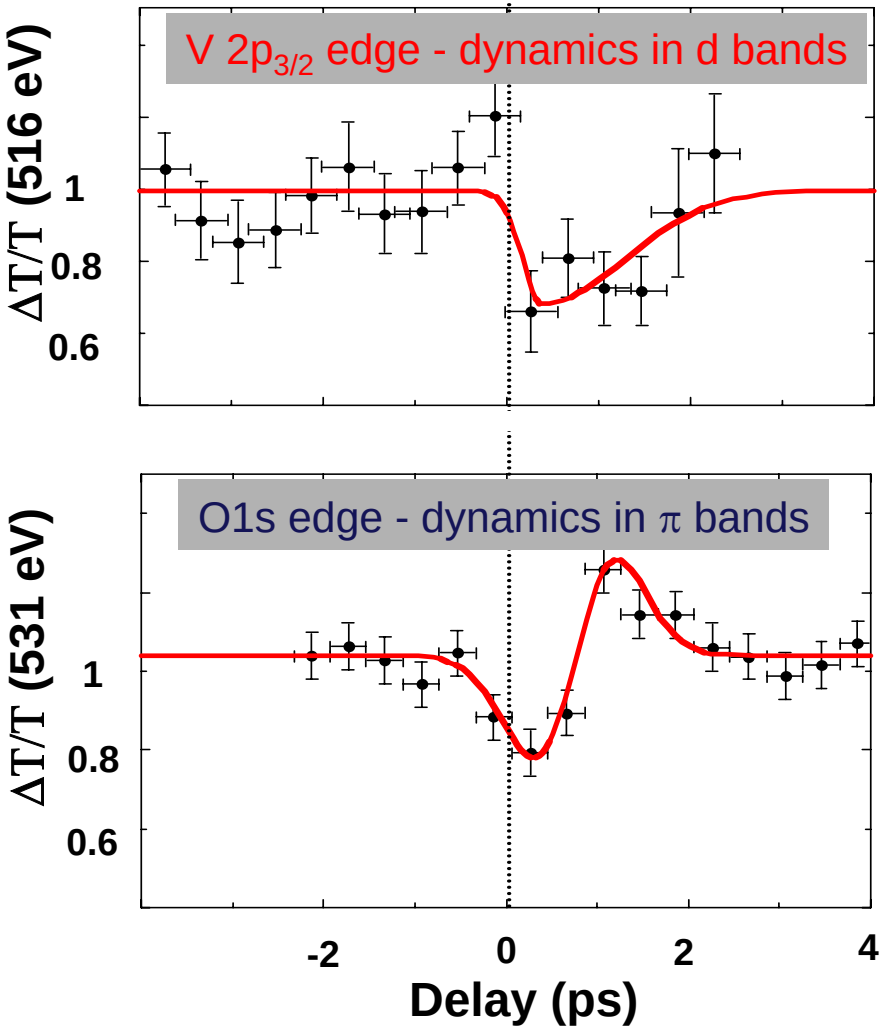




Dynamic Chemical Shift ?

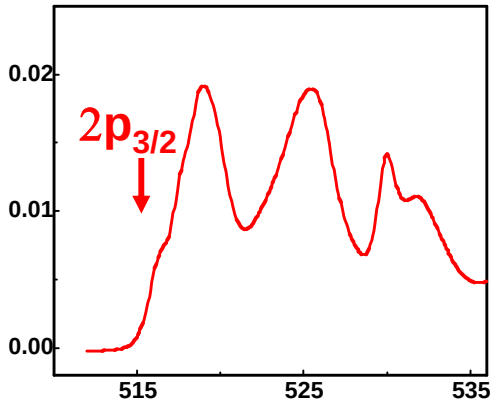


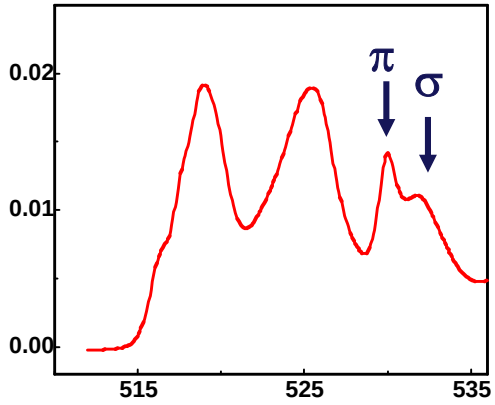




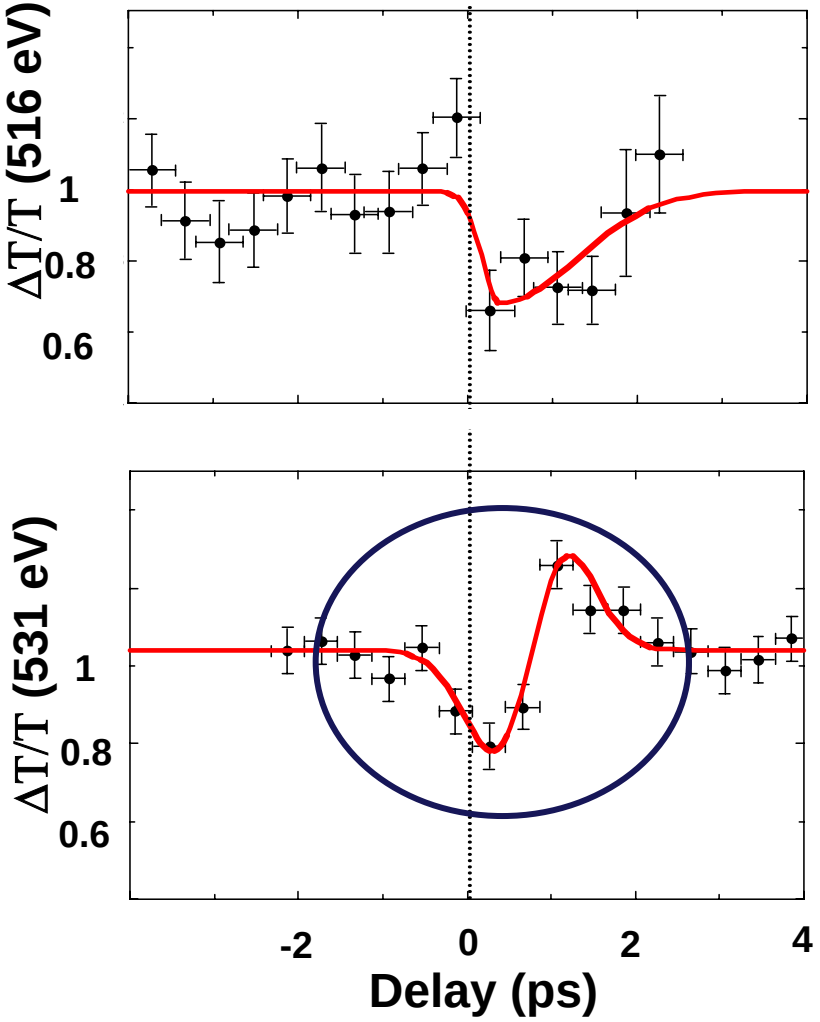
Future: high spectral resolution

Resolution (0.1 eV)





Fs NEXAFS Spectroscopy

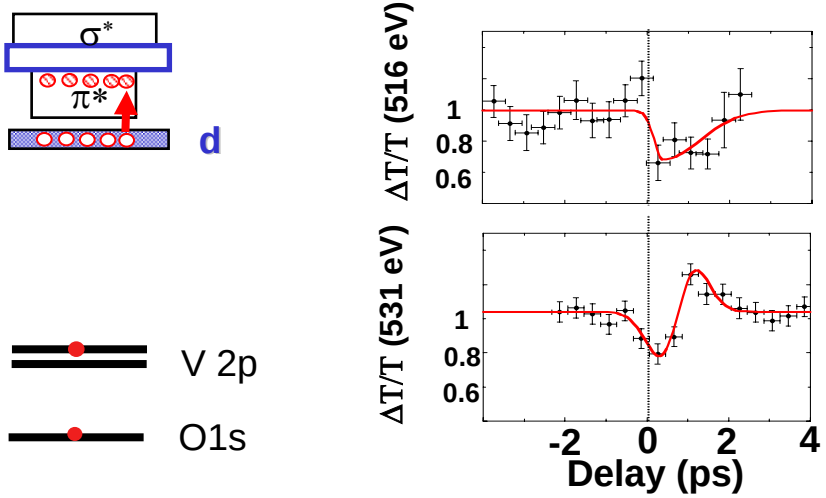


Summary

We study the photo-induced phase transition in VO_2

We observe a structural bottleneck after photo-doping: indicative of a band insulator

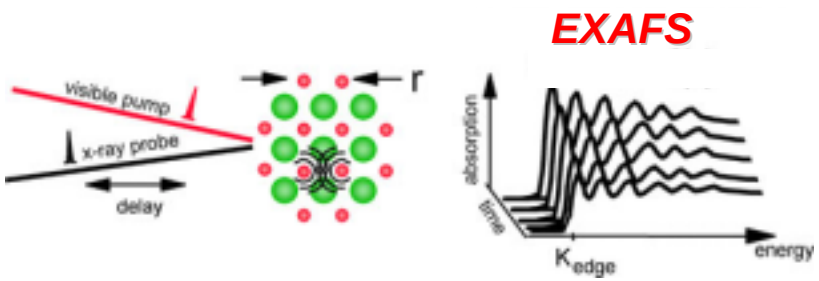
Fs NEXAFS measures the dynamics of the d and π bands

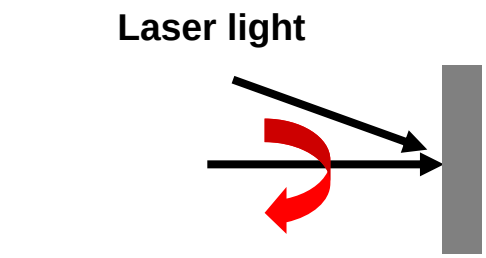


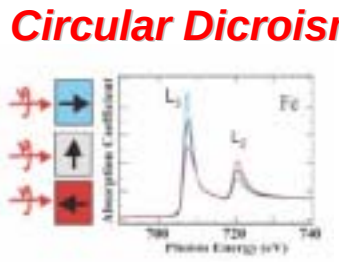
Summary

We demonstrate the first femtosecond NEXAFS at 500 eV

Tunable femtosecond x-rays open new opportunities



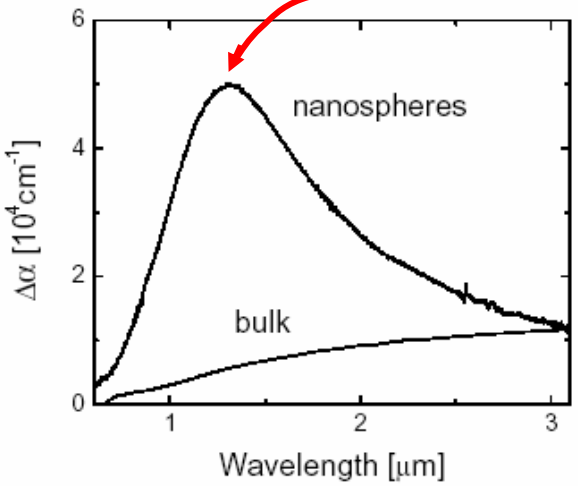




Nano-crystals: Surface Plasmons

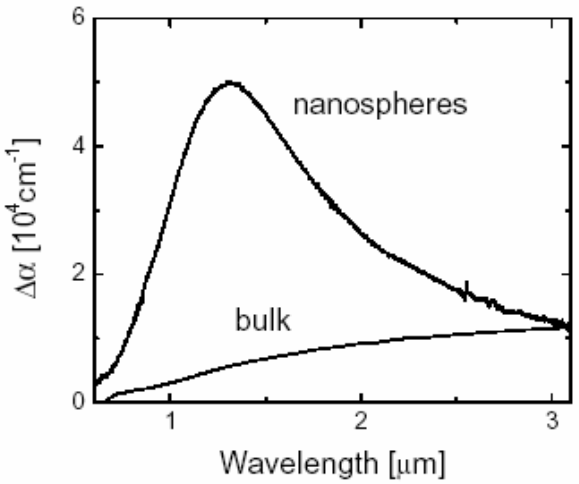
$\alpha_{met} - \alpha_{ins}$

Density of free carriers

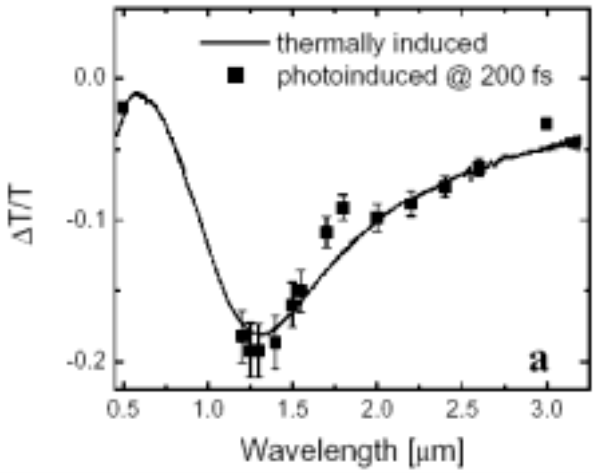


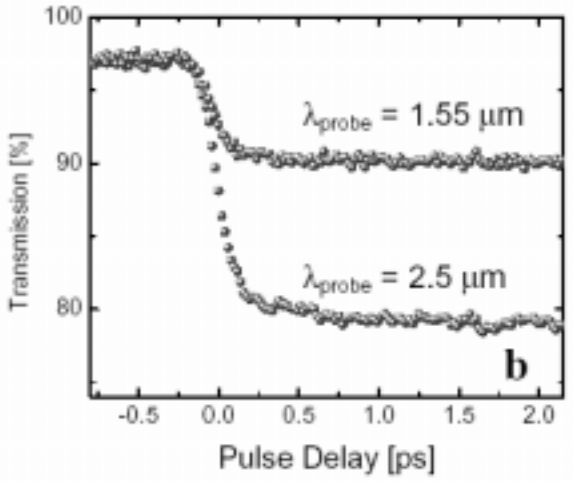
Nano-crystals: Surface Plasmons

Thermal response

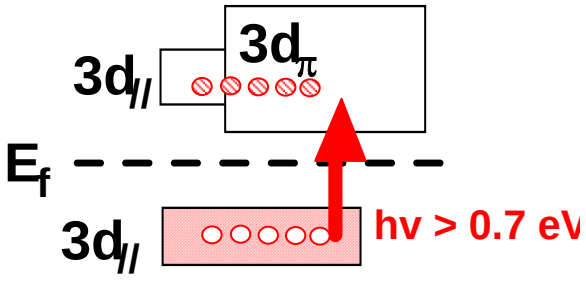


Thermal vs photo response

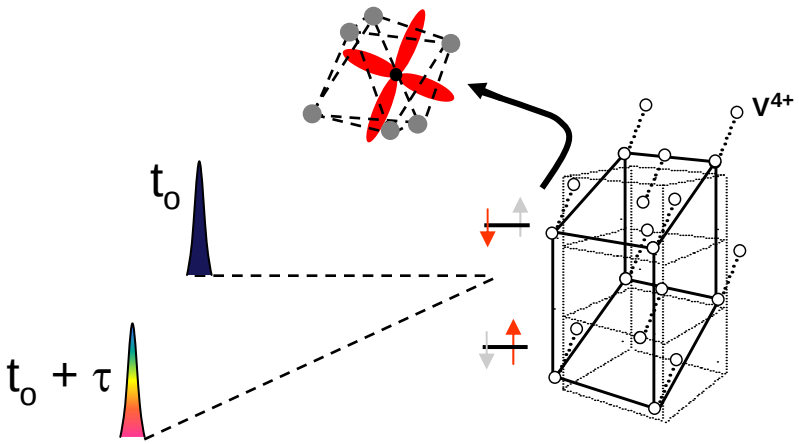


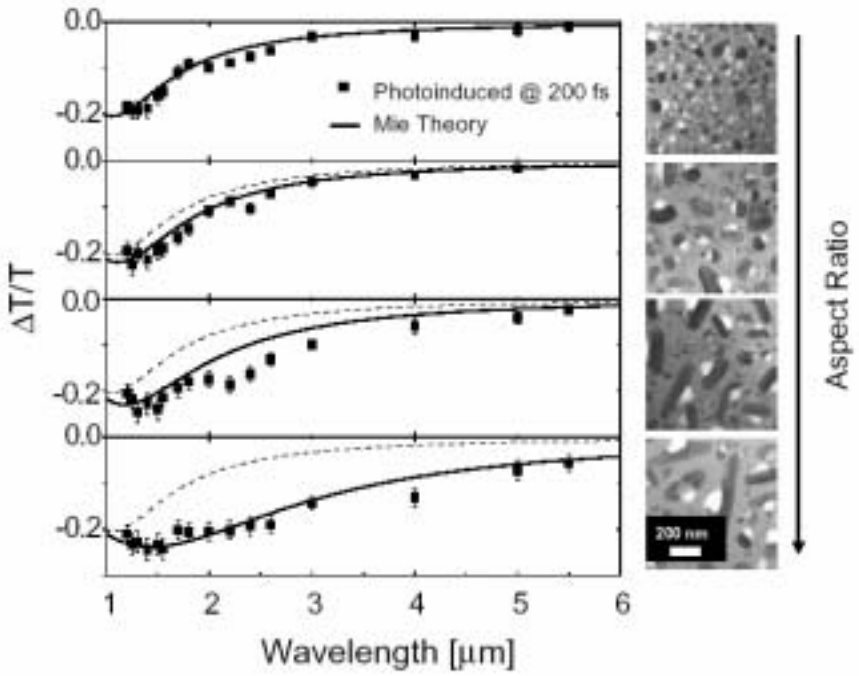


Nano-crystals: High-T phase



> 50% holes

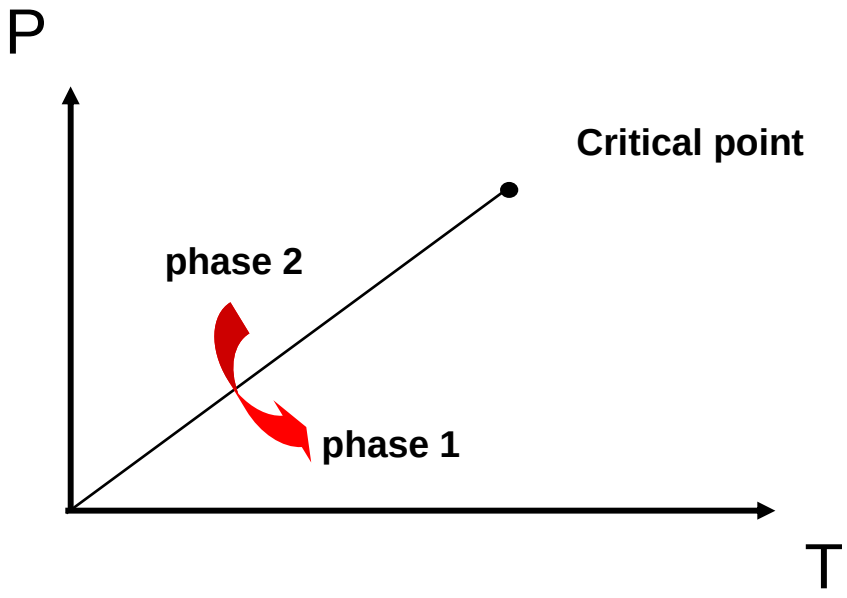


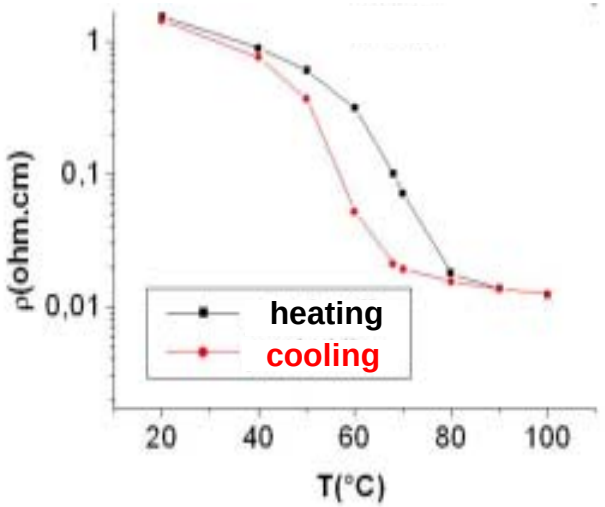


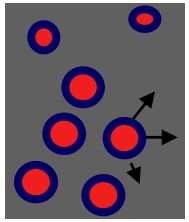
M.Rini et al. *submitted* (2004)

Time-integrated Experiments

Thermally-induced structural transitions are often First-Order and hysteretical.

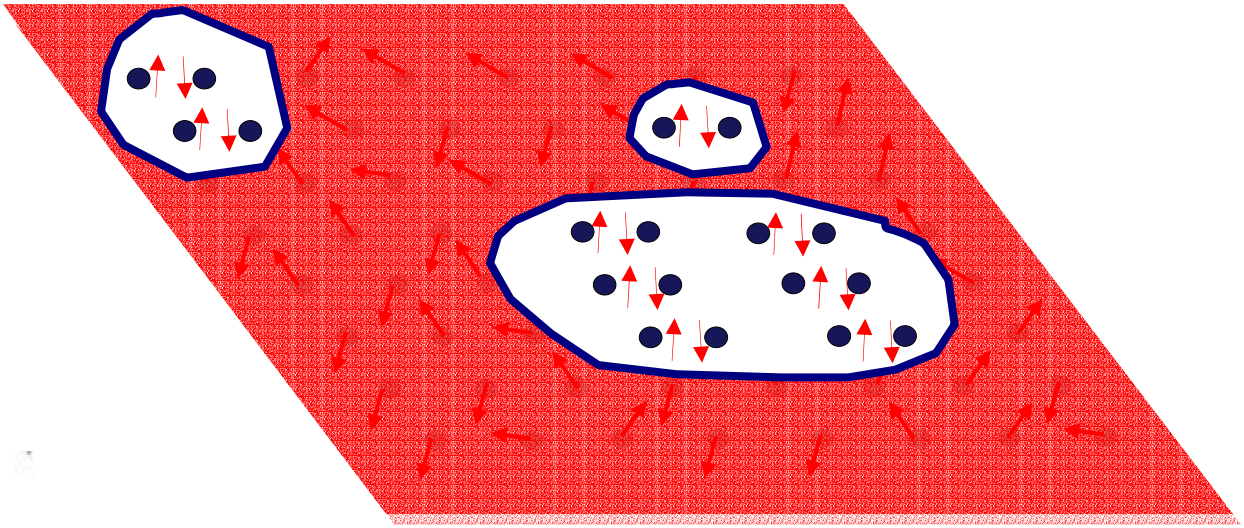


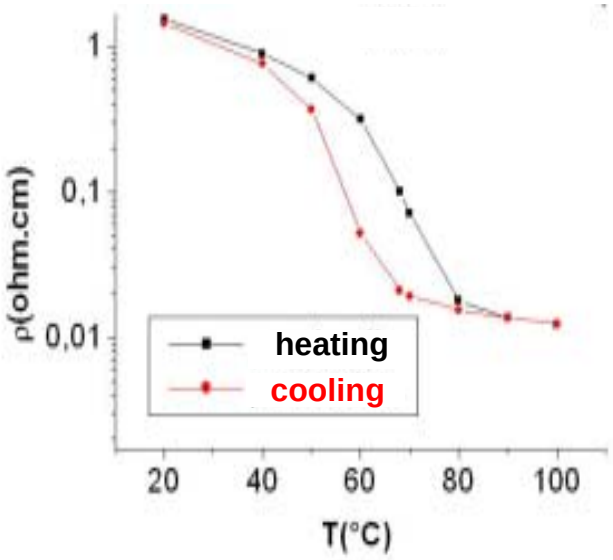




Mesoscopics mask microscopic behavior

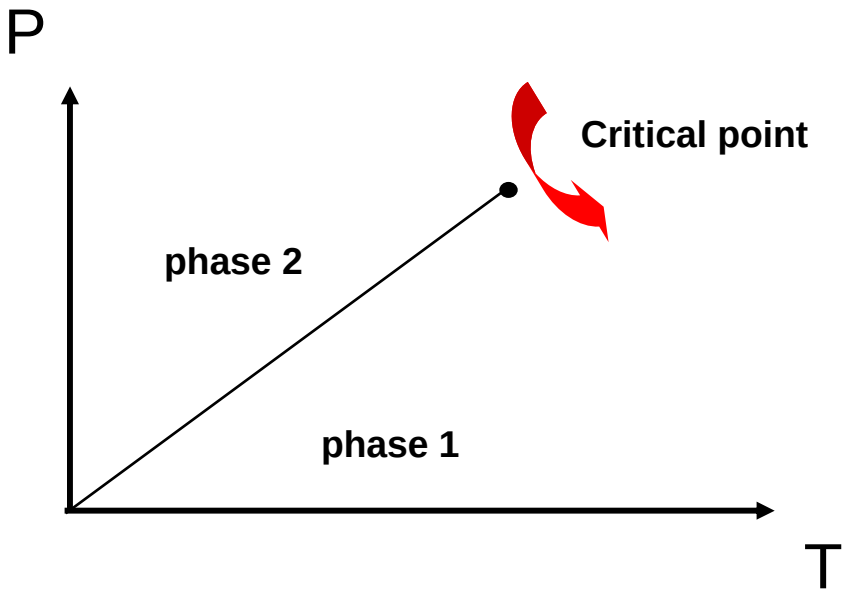
Phase Separation

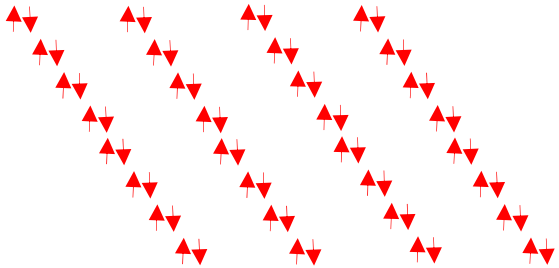


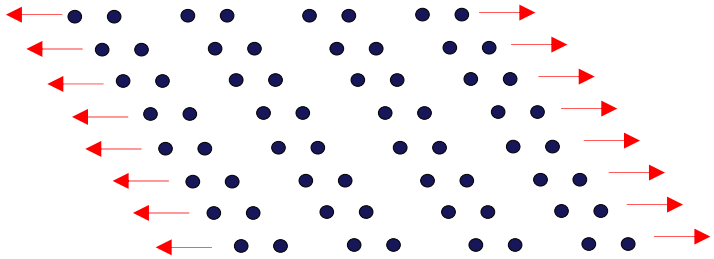


Critical behavior

Only at high pressures, i.e. above the critical point the transition is continous







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