

***Femtosecond X-ray Absorption Measurement of
the Insulator-to-metal Transition in VO_2***

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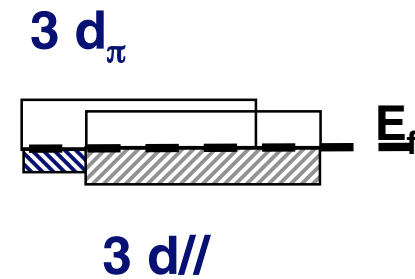
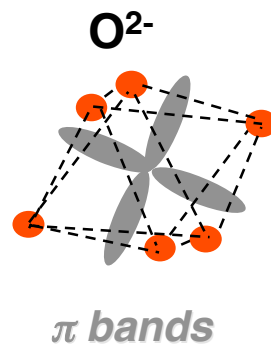
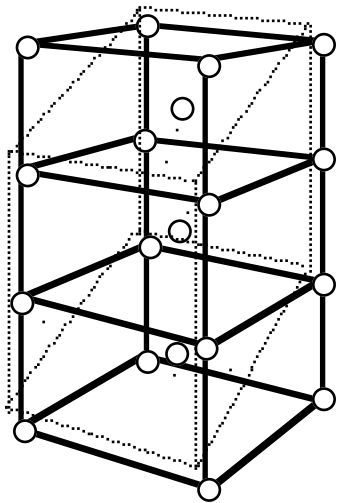
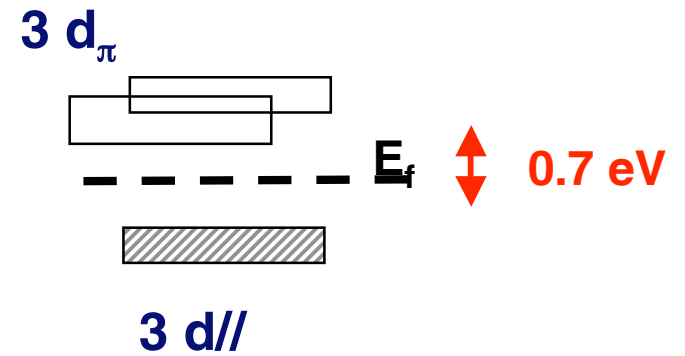
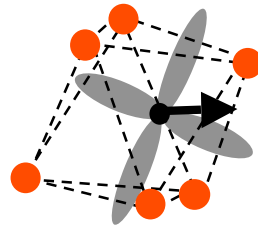
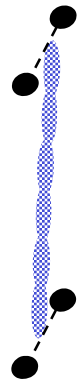
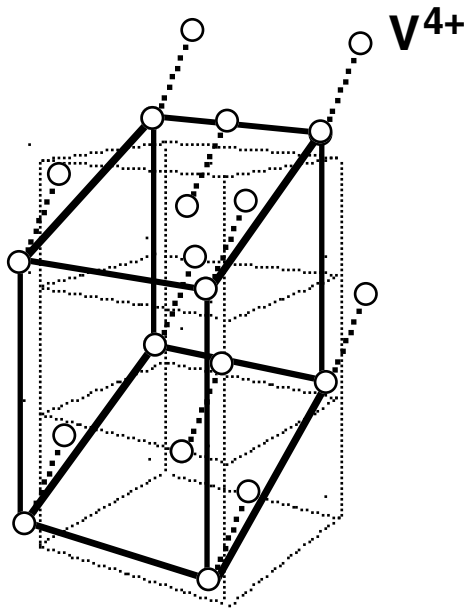
T.E. Glover,
P. Heimann,
H. Padmore

Advanced Light Source, LBNL

S. Fourmaux
J.K. Kieffer

University of Quebec

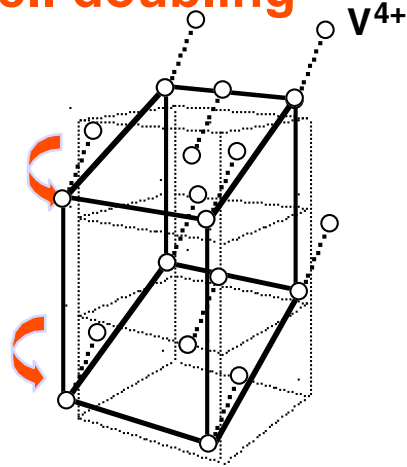
Metal-Insulator transition in VO_2



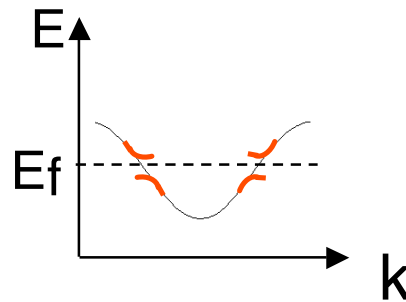
Insulator : Peierls or Mott Hubbard?



Cell doubling



? Peierls transition ?

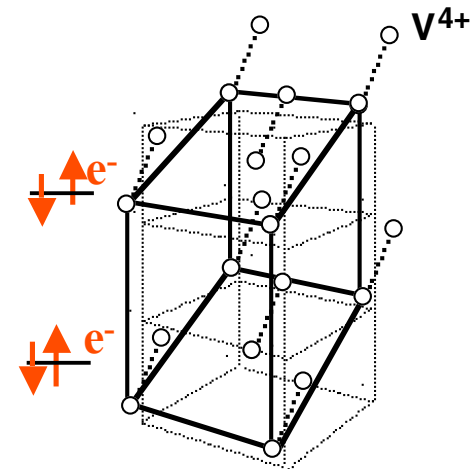


J.B. Goodenough *Phys. Rev.* 117, 1442 (1960)

Wentzcowitch et al. *Phys. Rev. Lett.* 72, 3389 (1994)

Localisation

? Mott-Hubbard Insulator ?



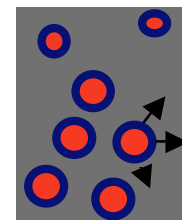
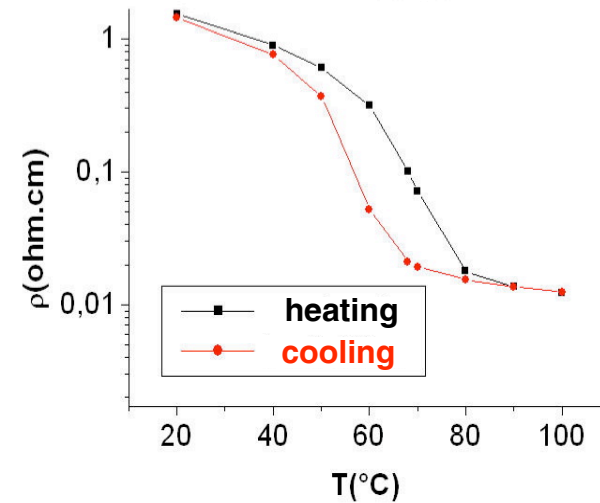
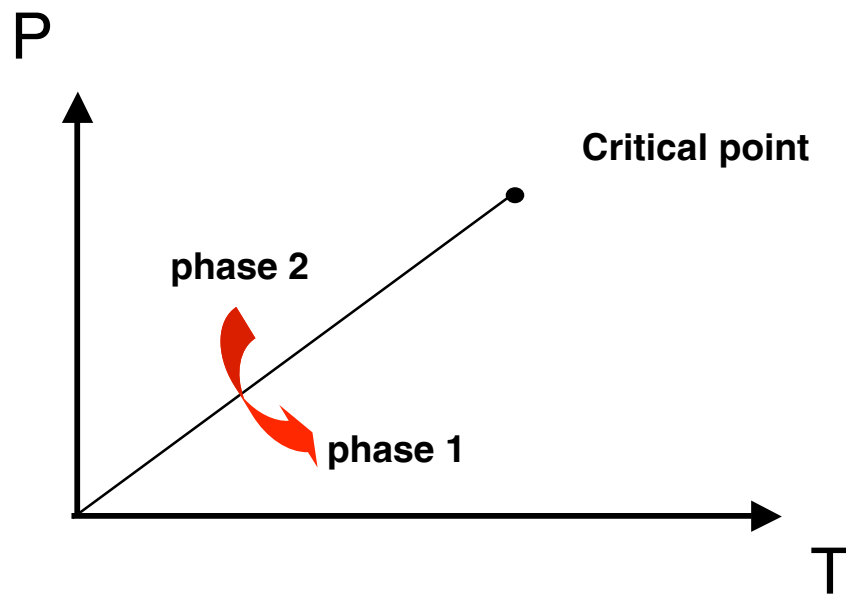
Zylbersztejn and N. Mott
Phys. Rev. B 11, 4383 (1975)

Pouget et al. *Phys. Rev. B* 10, 801 (1974);
Phys. Rev. Lett. 35, 873 (1975)

Time-integrated Experiments

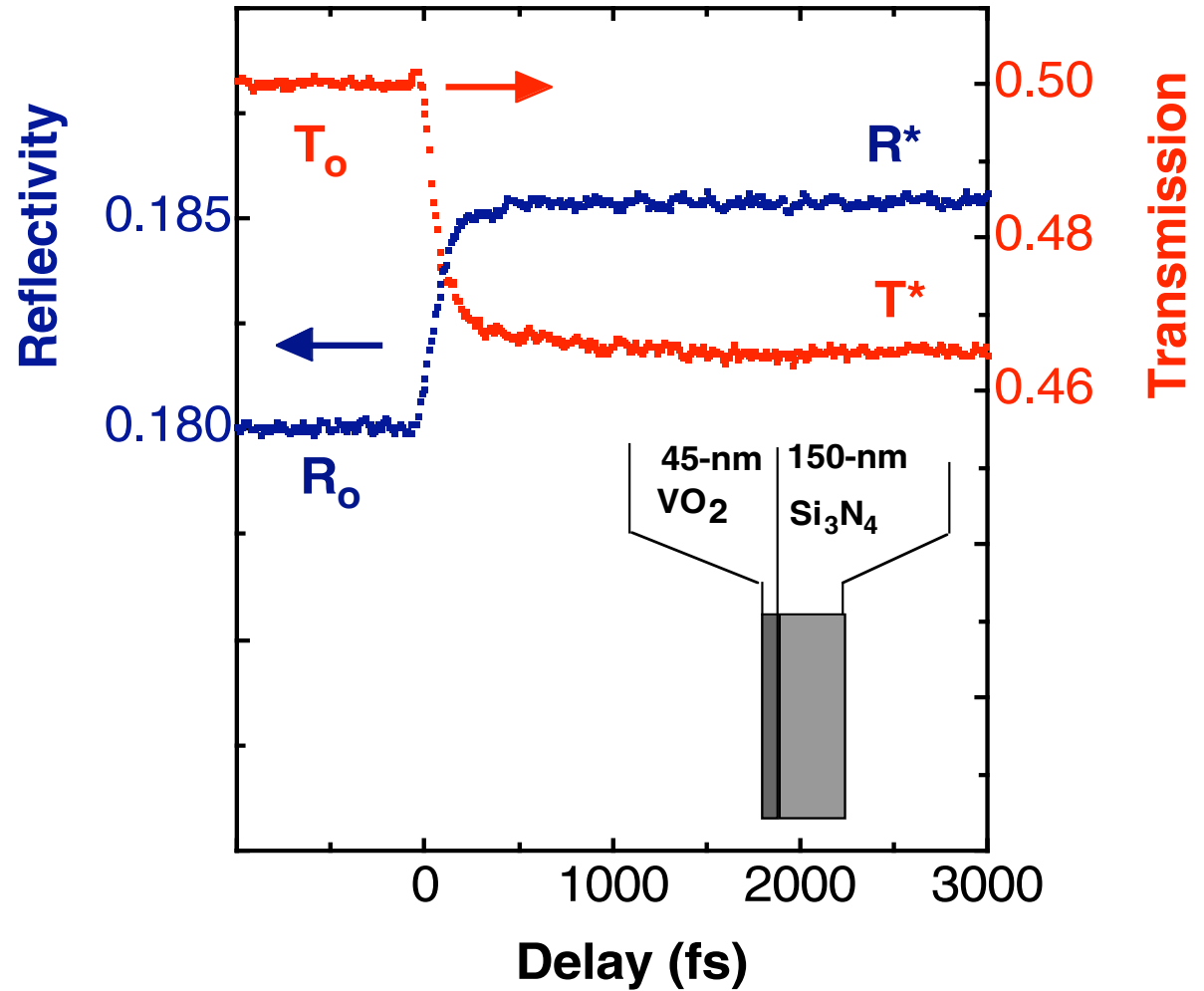
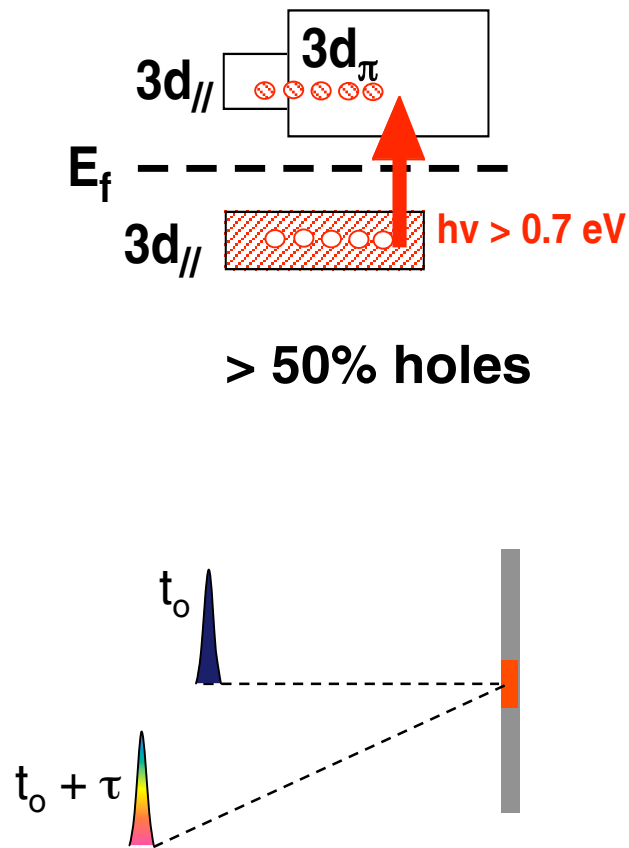


Thermally-induced structural transitions are often **First-Order** and **hysteretical**.

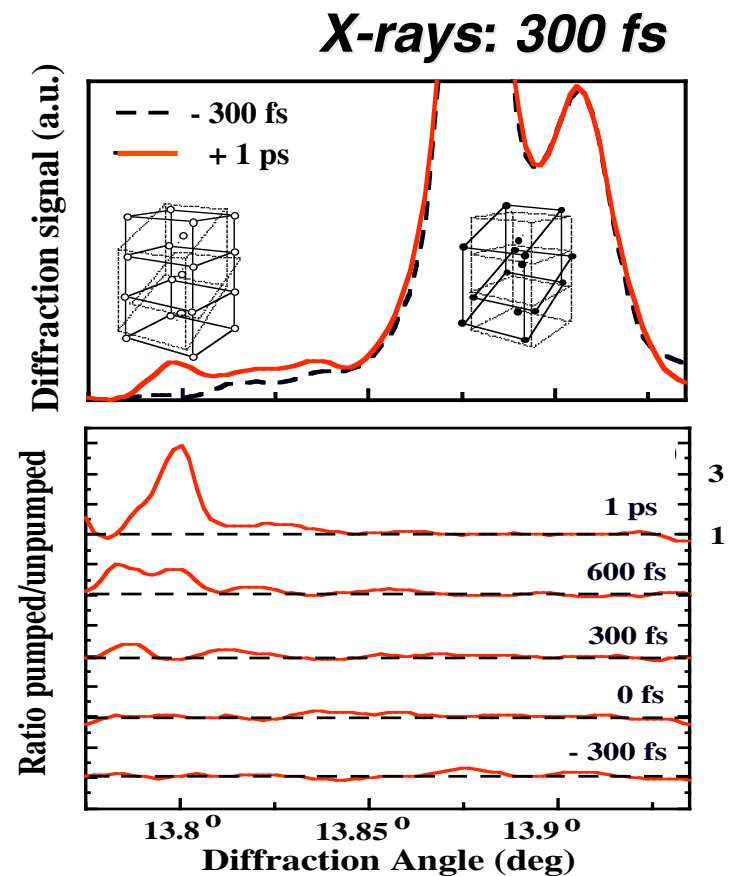
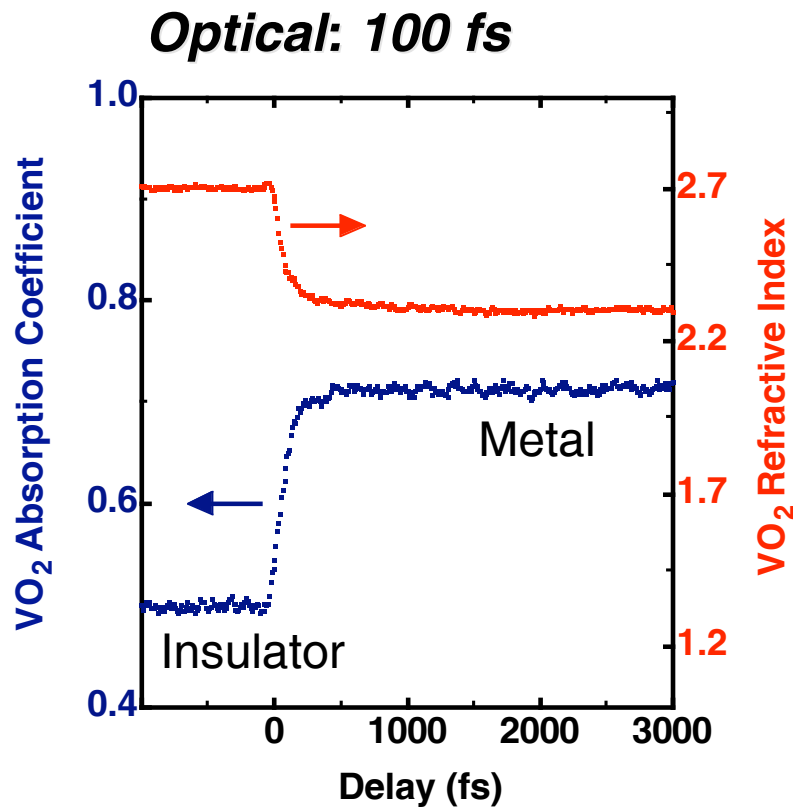
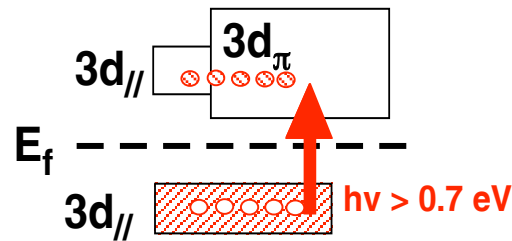


**Mesoscopics mask
microscopic behavior**

Photo-induced Insulator-to-Metal



Simultaneous transitions ?

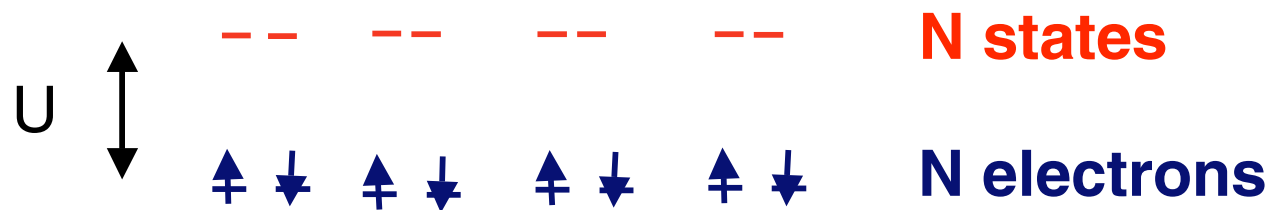
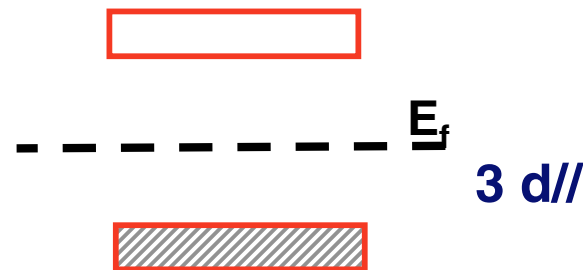


Cavalleri et al. *Phys. Rev. Lett.* 87, 237401 (2001)

Low T Phase: Mott Insulator?



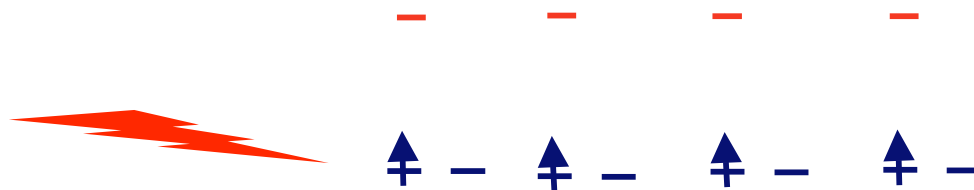
Mott Hubbard Insulator ?



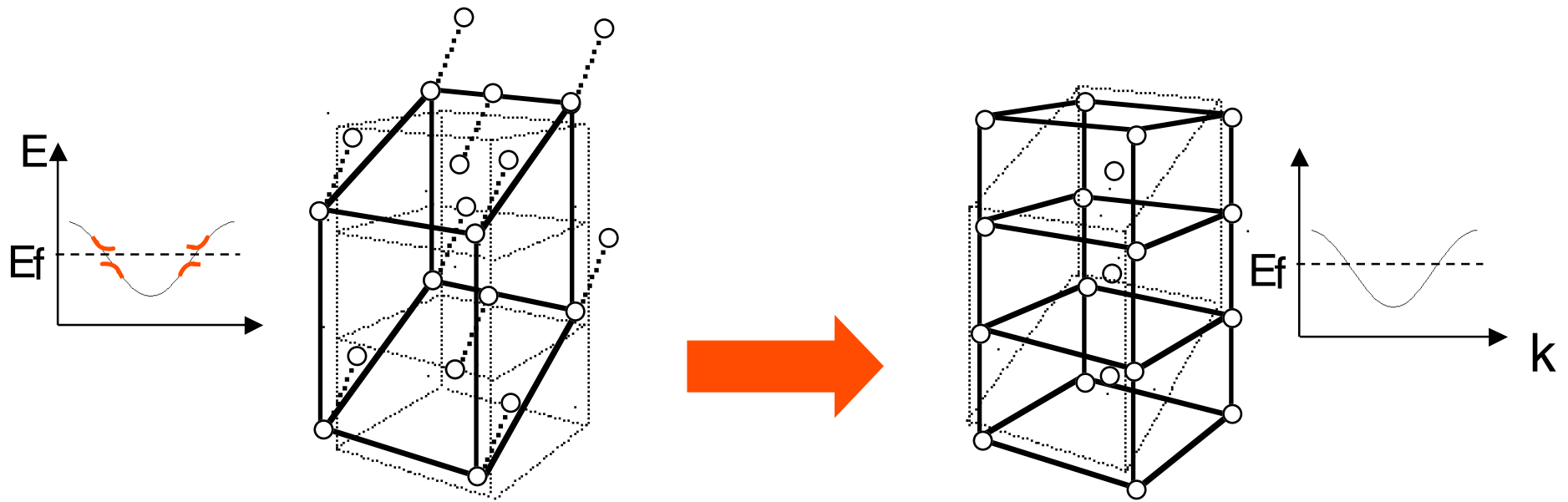
Mott Transition: Instantaneous



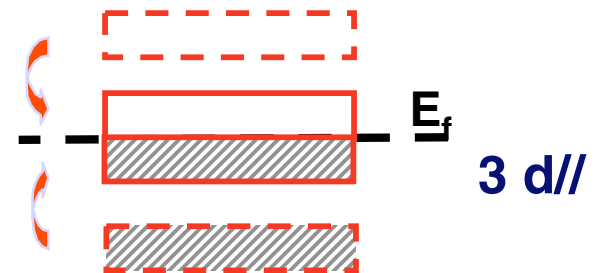
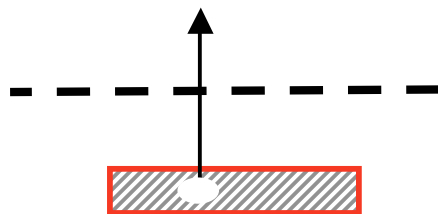
Hole Doping



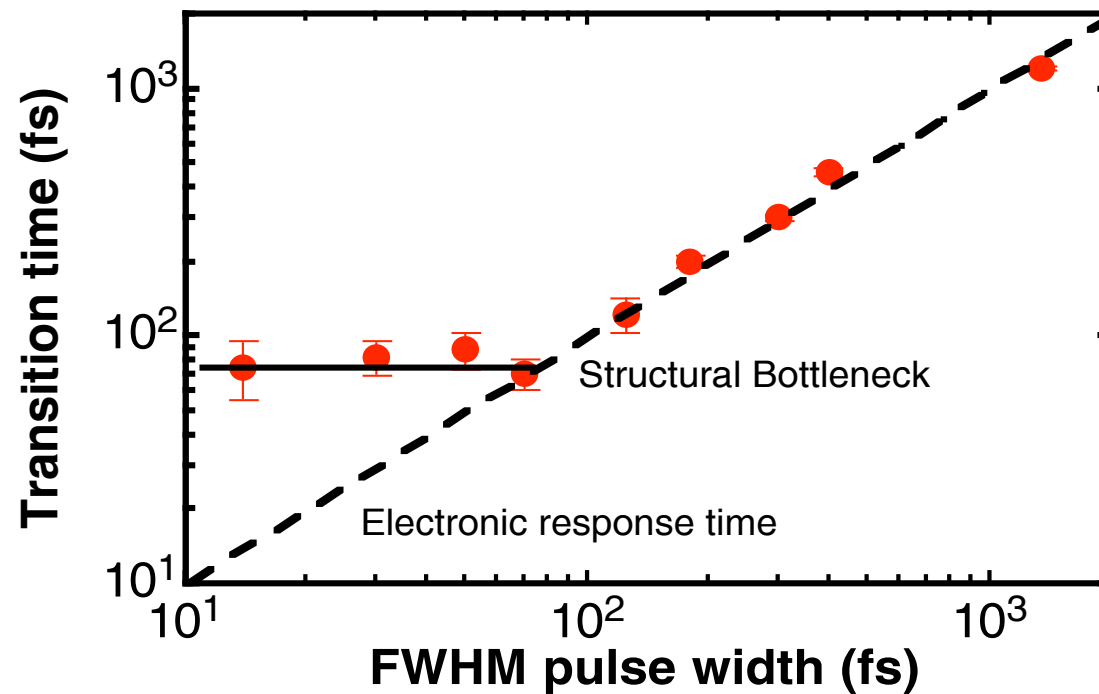
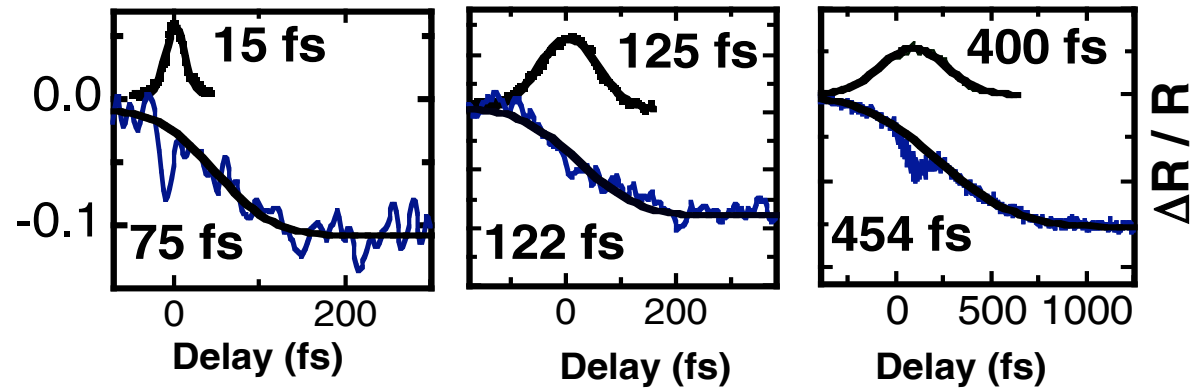
Reverse Peierls Transition: 100 fs



Optical Excitation



Phase Transition Time: 75 fs

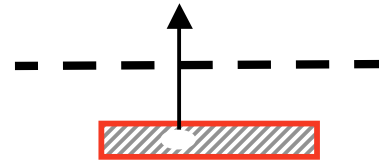


Hierarchy of Events

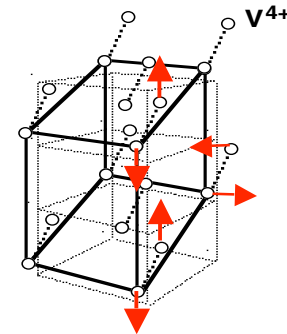


Structural Motion is necessary for the metallic phase

1) Hole injection



2) Structural Motion



3) Metal

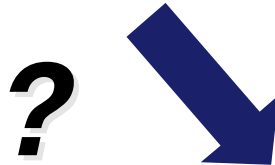
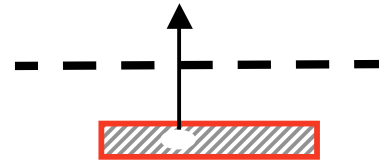


Hierarchy of Events

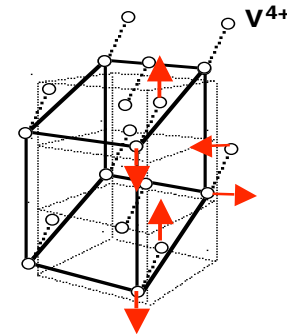


Structural Motion is necessary for the metallic phase

1) *Hole injection*



2) *Structural Motion*



3) *Metal*

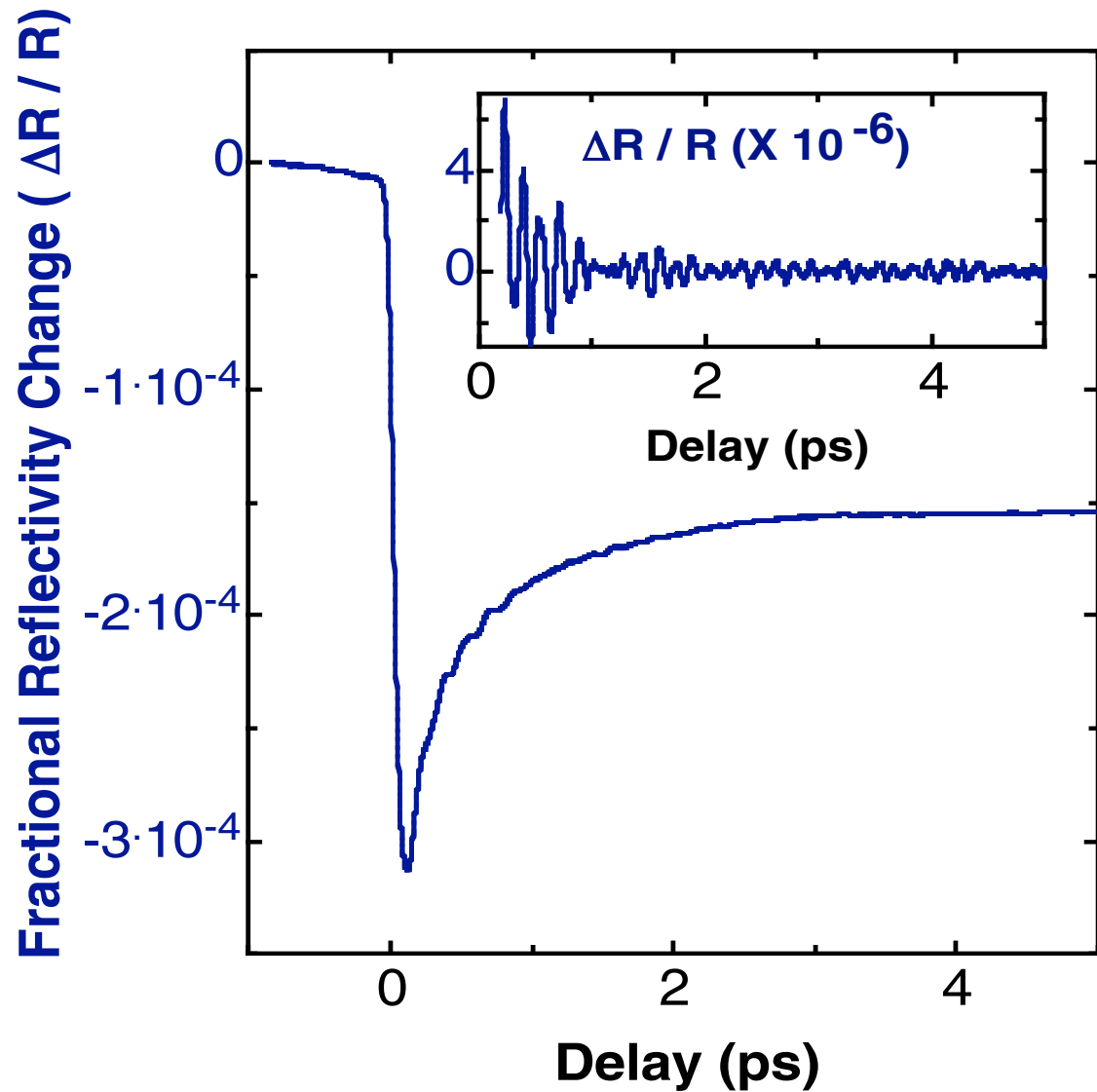
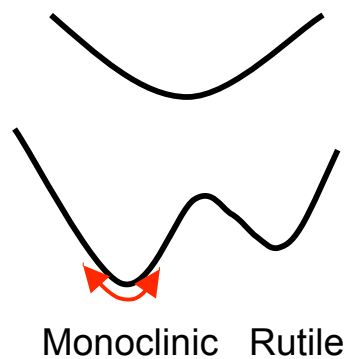


Impulsive excitation of Optical Phonons

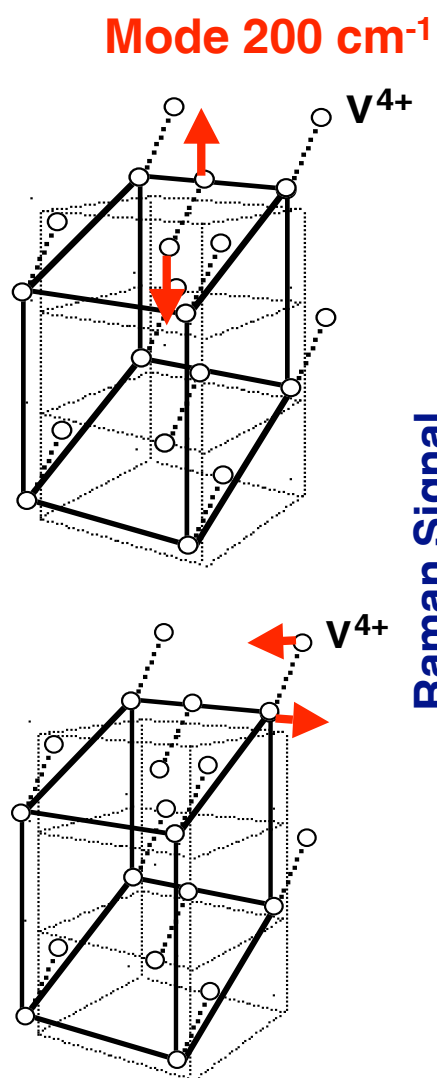


Low fluence

No Phase transition

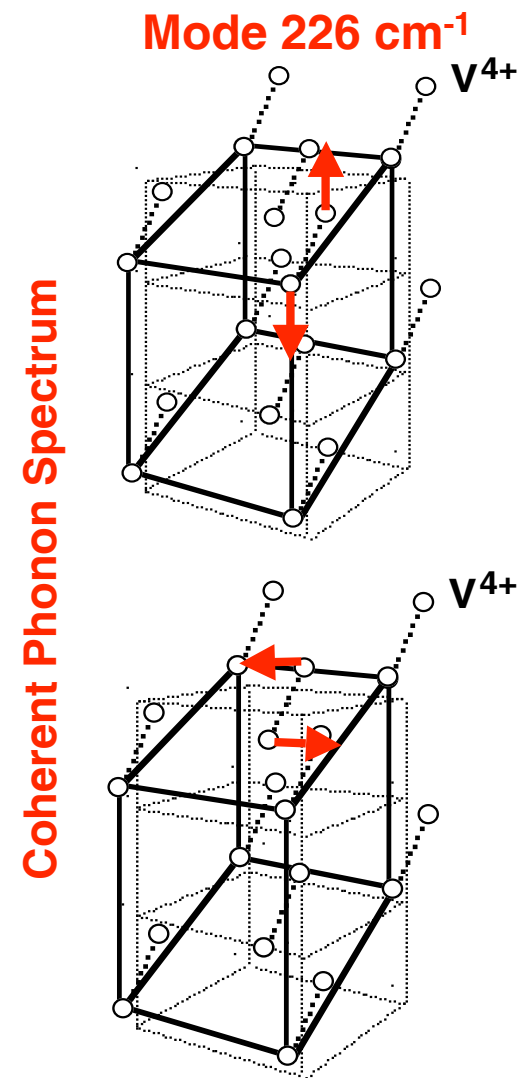
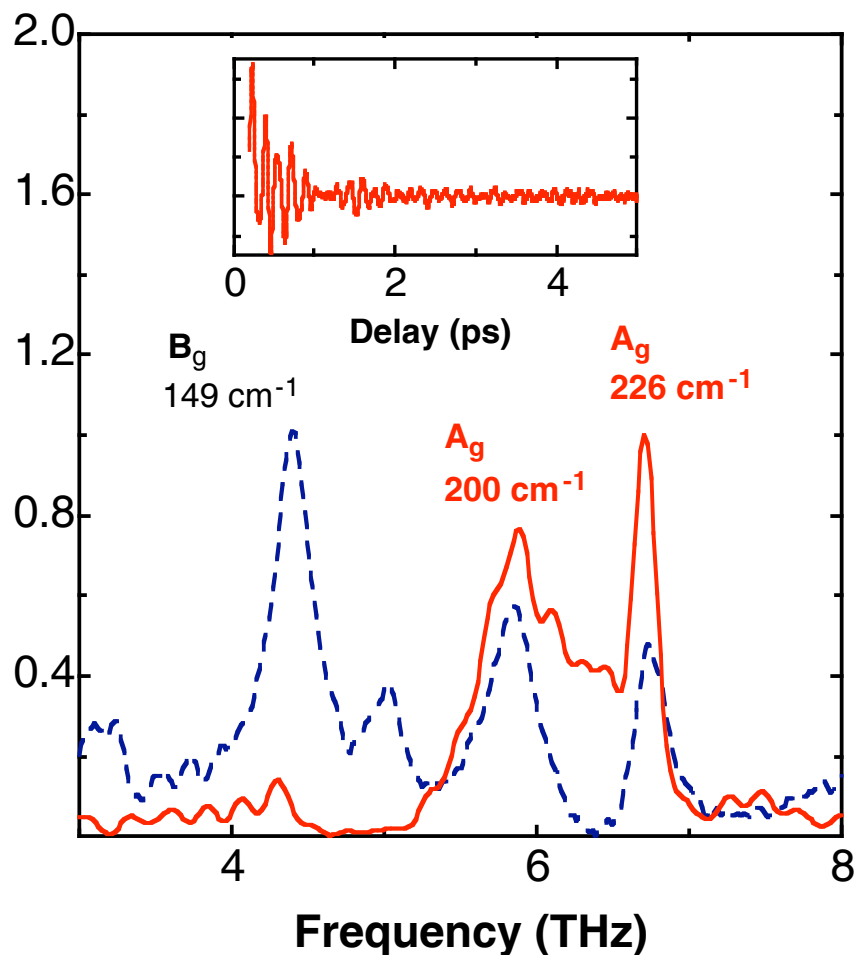


Excitation of symmetry-breaking modes



Raman Signal

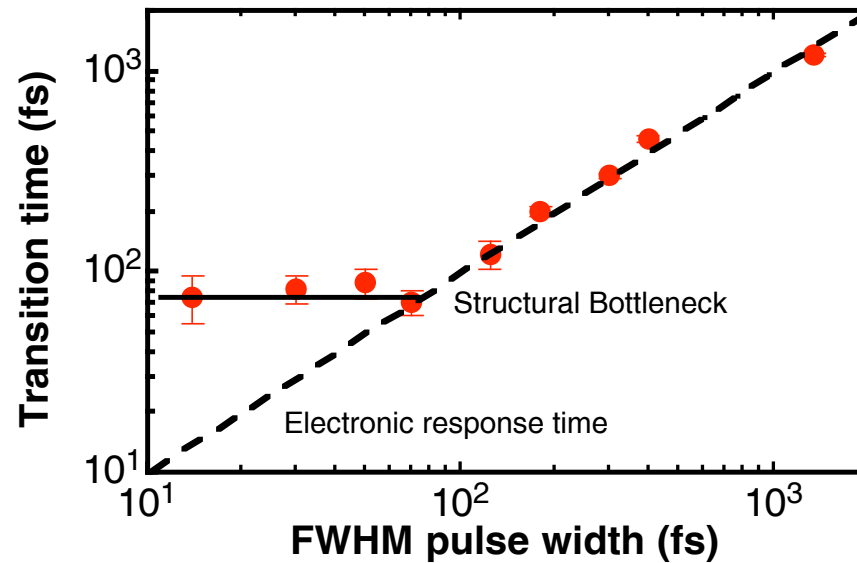
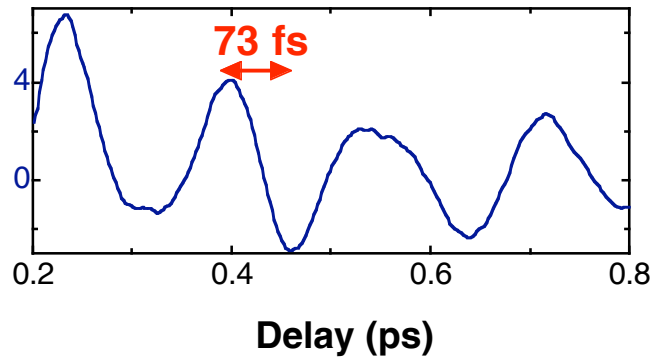
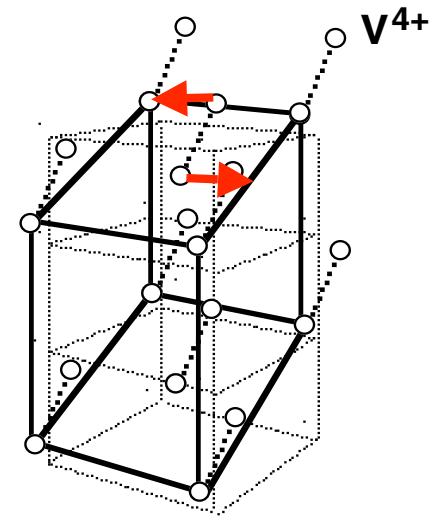
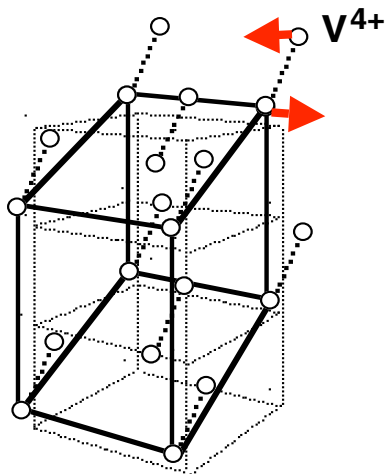
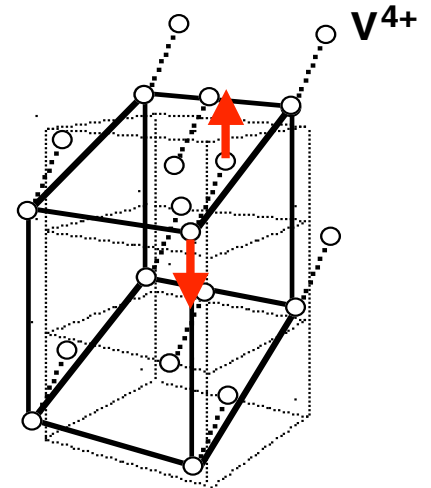
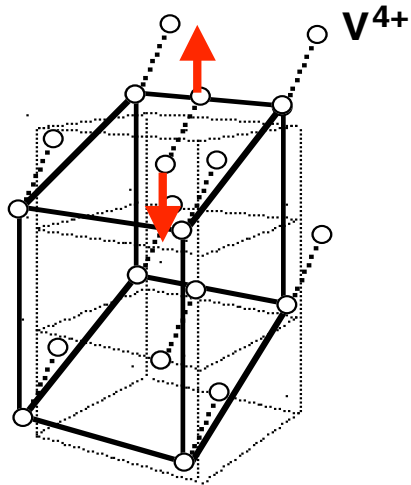
Impulsive Raman



Excitation of symmetry-breaking modes



Impulsive Raman



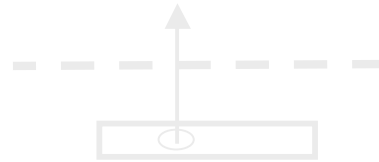
Cavalleri et al. cond-mat/0403214

How does the DOS rearrange ?



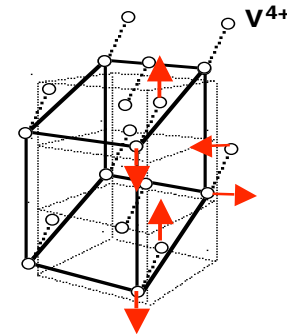
Structural Motion is necessary for the metallic phase

1) Hole injection



2) Structural Motion

?



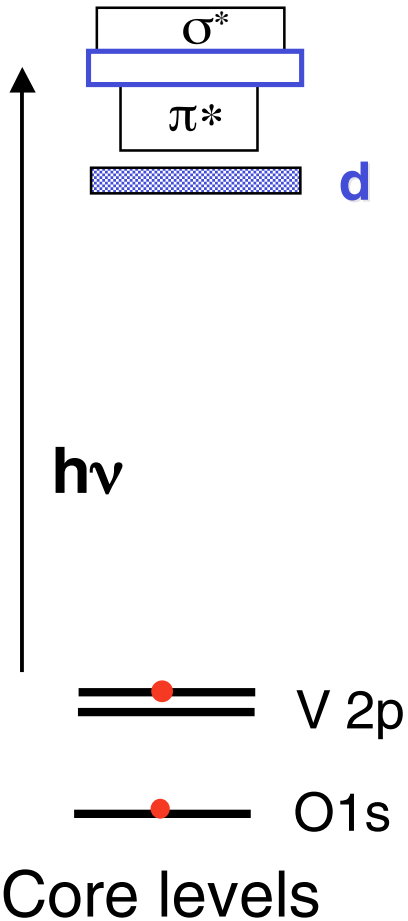
3) Metal



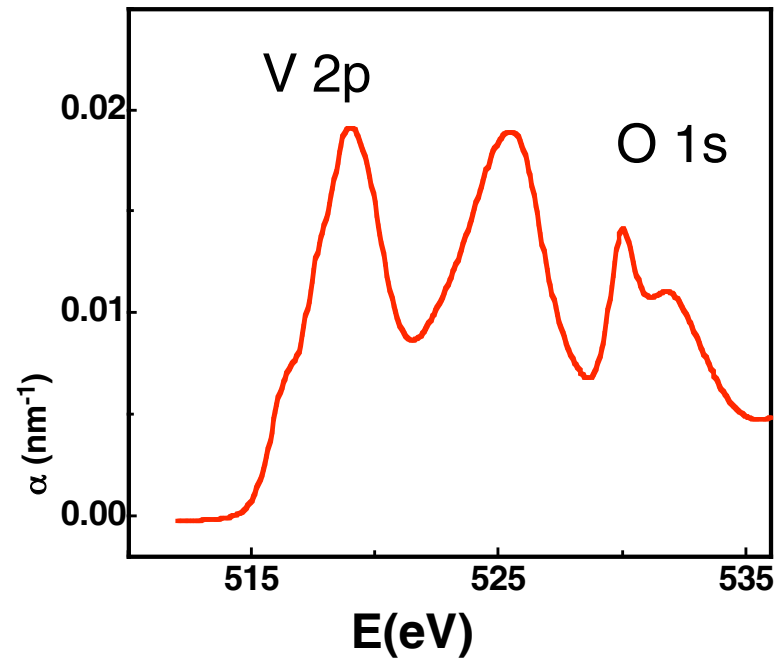
Electronic structure: NEXAFS



Insulator



Data: beamline 6.3.2 ALS



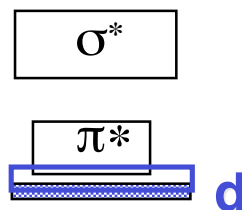
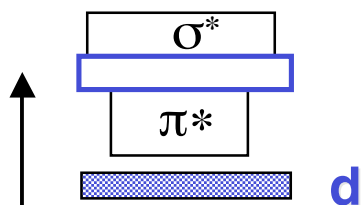
50 meV
resolution

Temperature driven transition

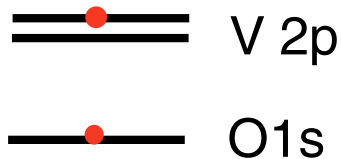


Insulator

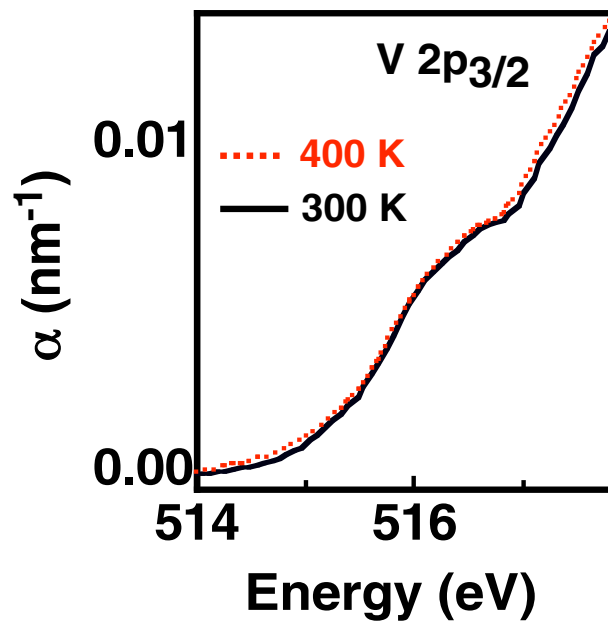
Metal



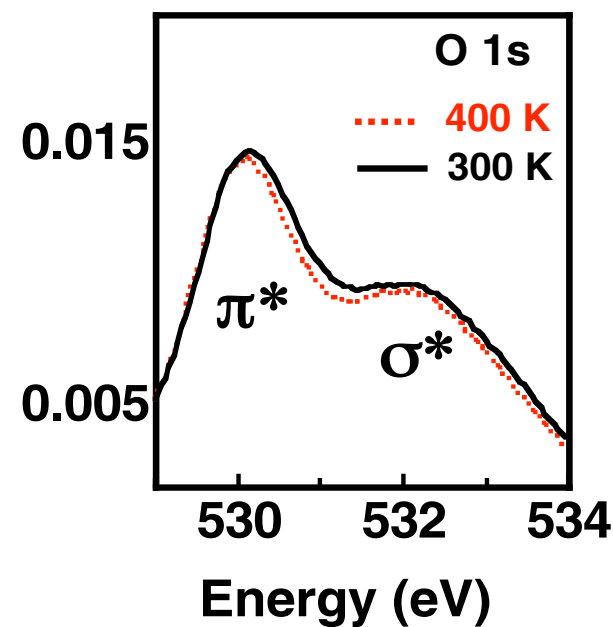
$h\nu$



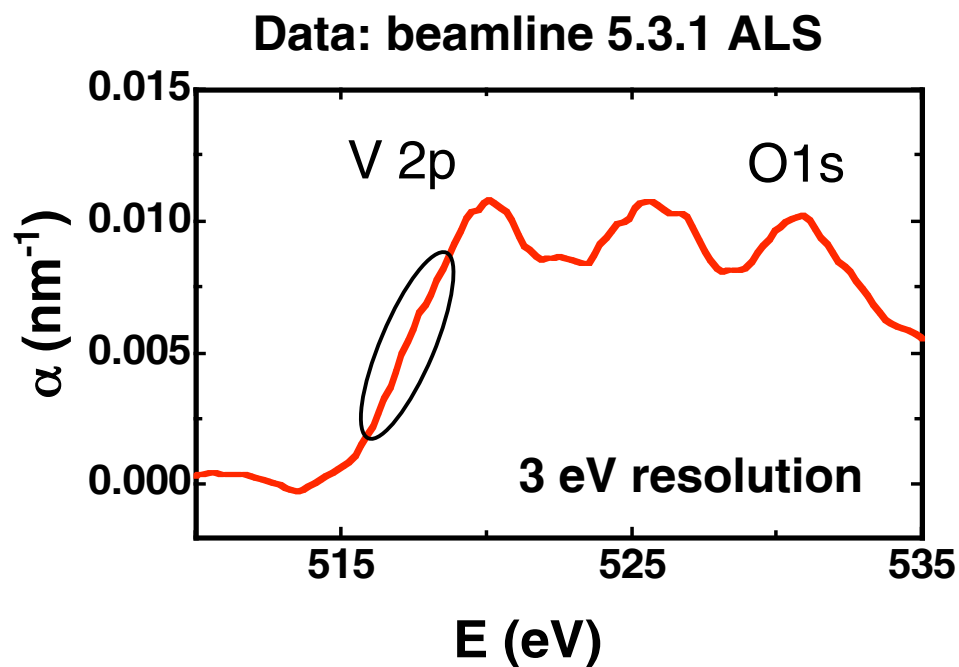
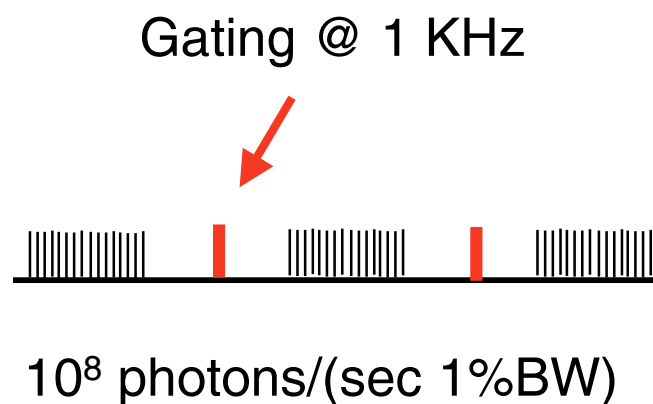
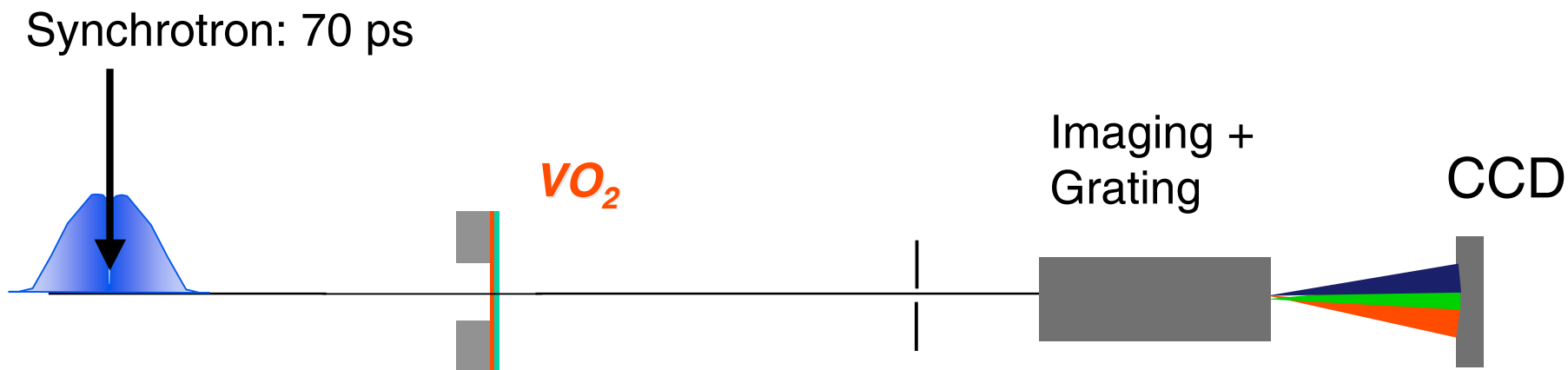
d orbitals



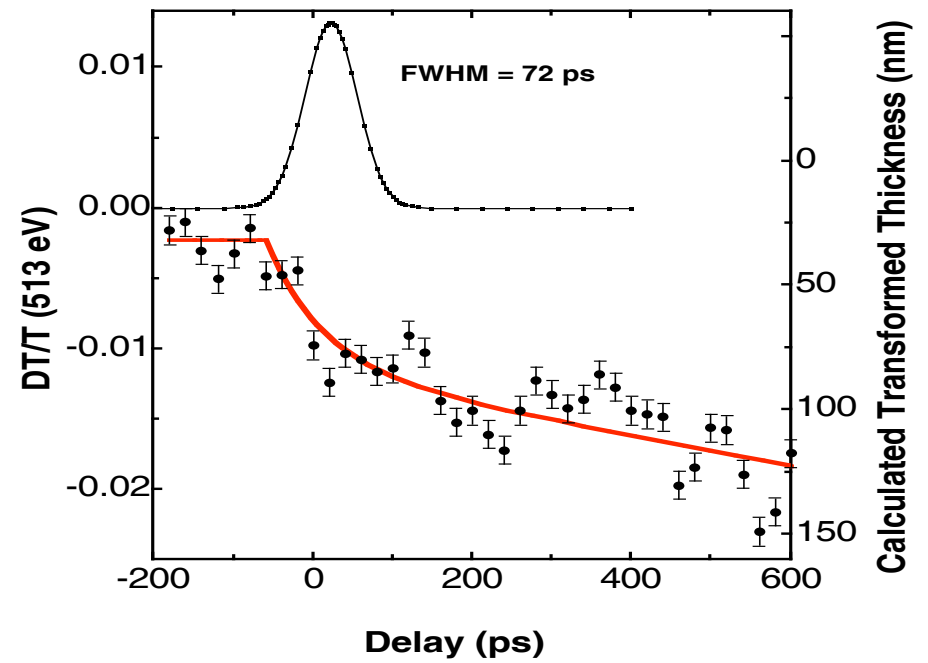
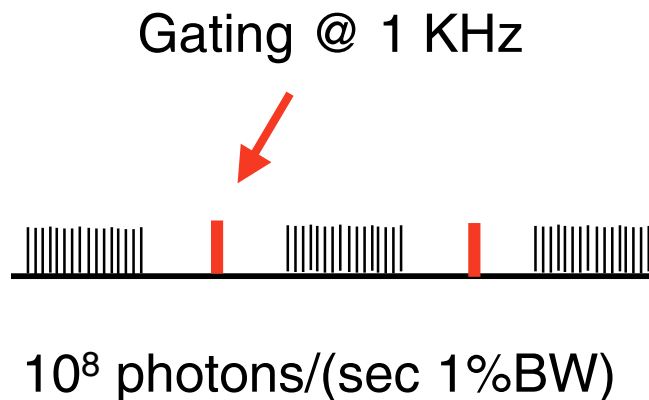
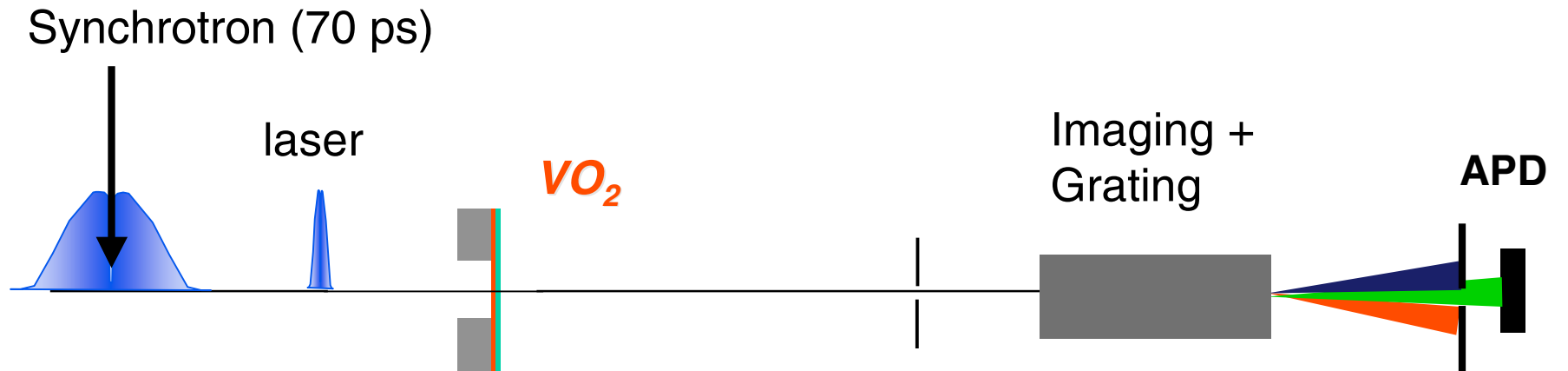
π orbitals



NEXAFS: Beamline 5.3.1

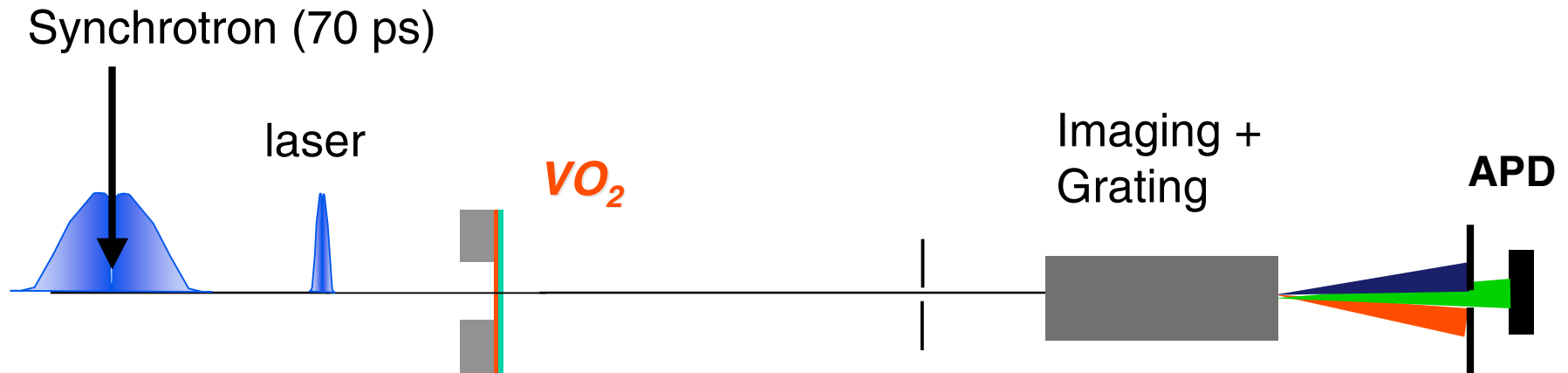


Time-resolved NEXAFS: ps scale

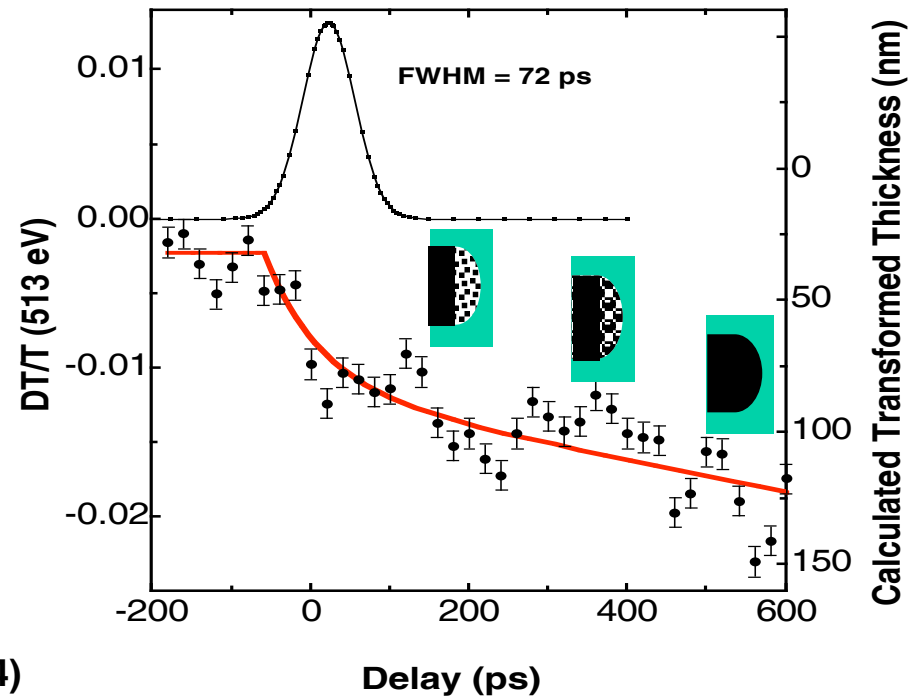


Cavalleri et al., Physical Review B 69, 1531XX (2004)

Time-resolved NEXAFS: ps scale

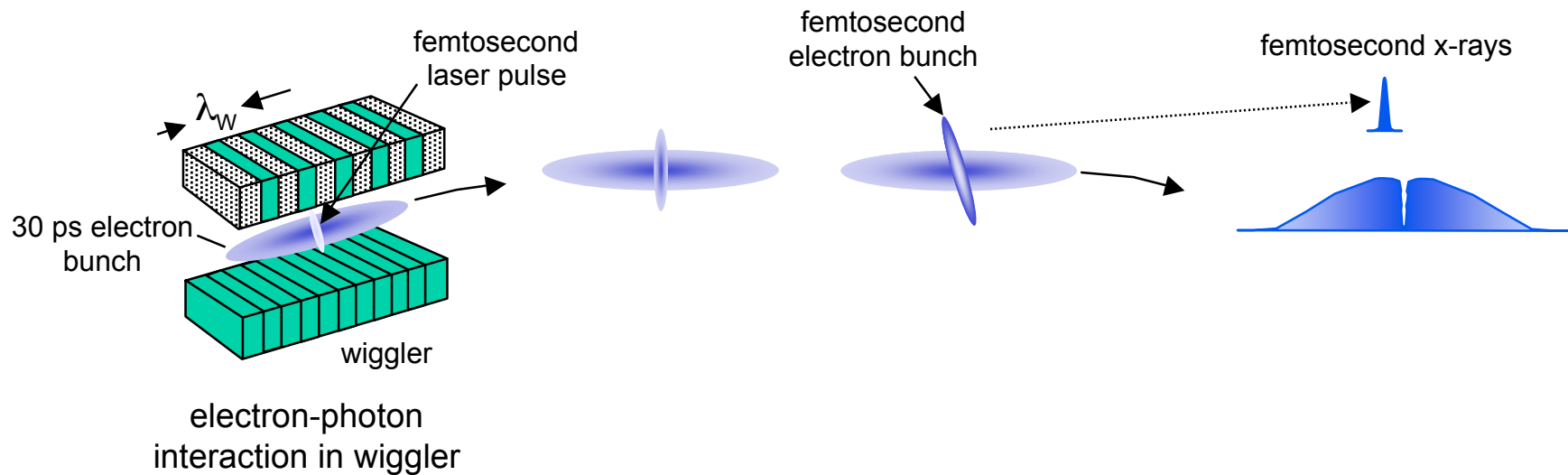
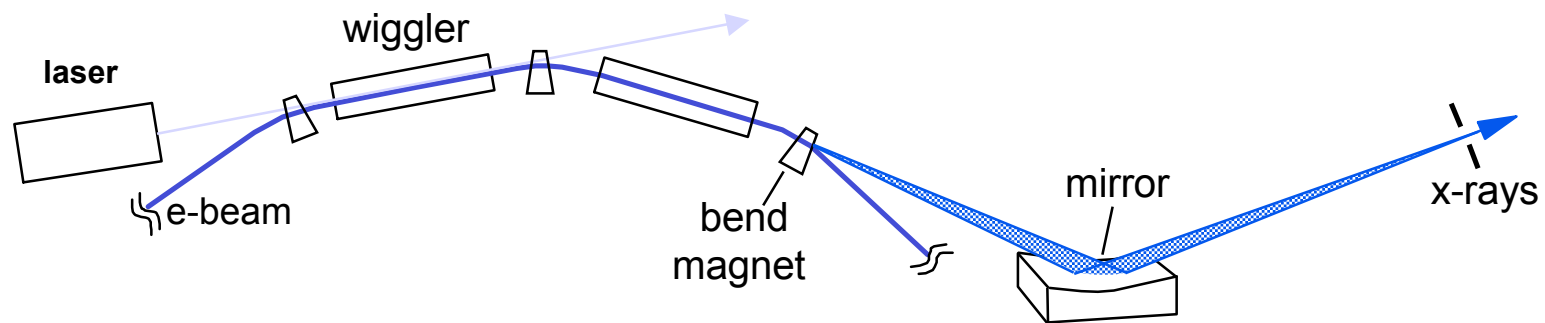


Ps signal dominated by mesoscopic growth



Cavalleri et al., Physical Review B 69, 1531XX (2004)

Tunable femtosecond X-rays at the ALS



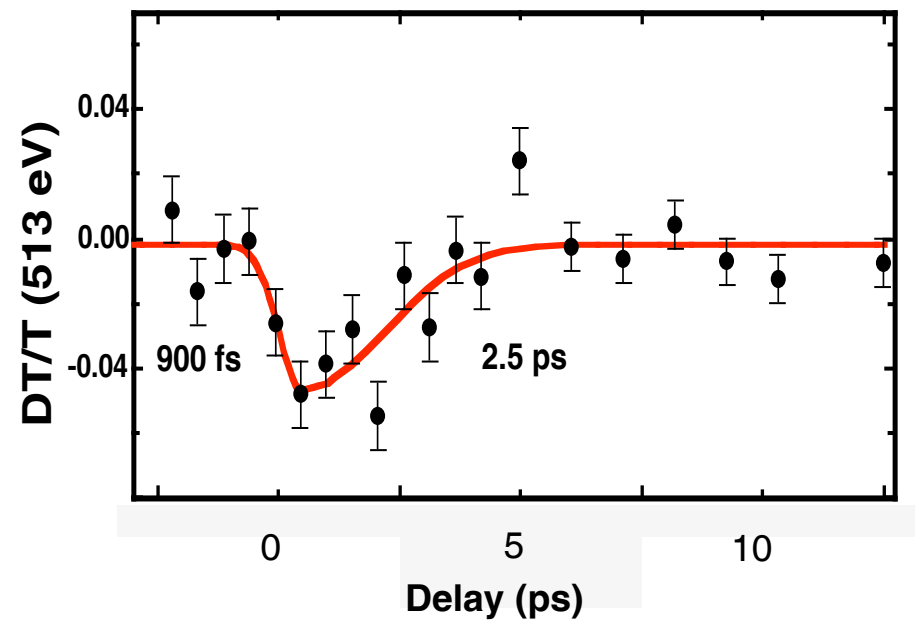
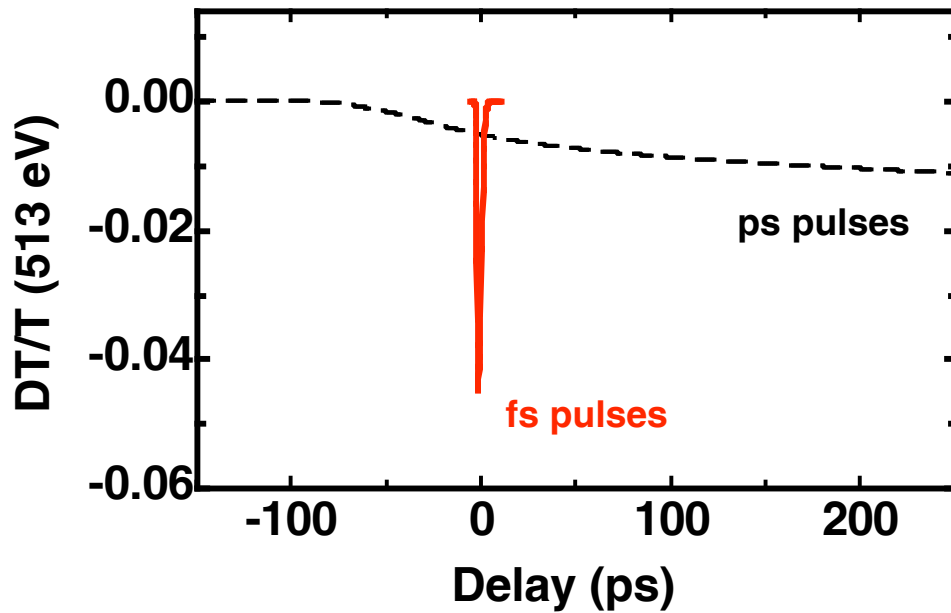
Zholents and Zolotarev, *Phys. Rev. Lett.*, 76, 916,(1996).

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

Transient Over Absorption



Sliced Pulse: 100fs



Over Absorption: Hole photo-doping



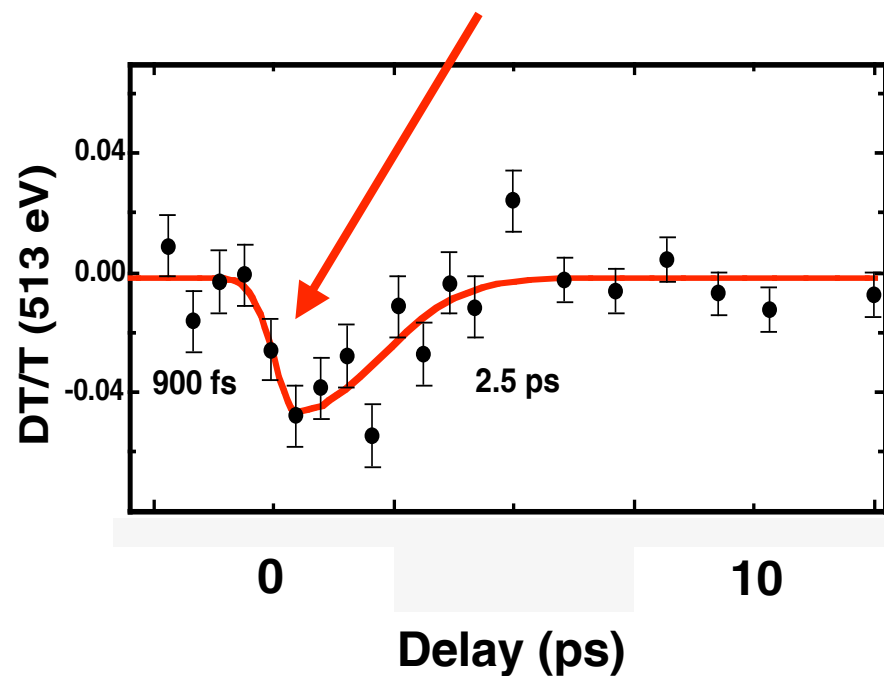
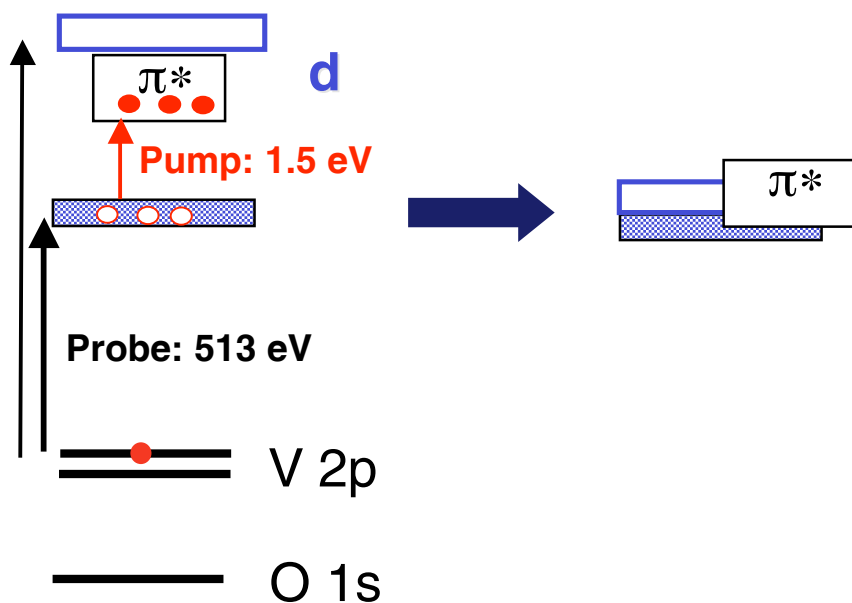
Sliced Pulse: 100fs



Hole Photo-doping

Metal

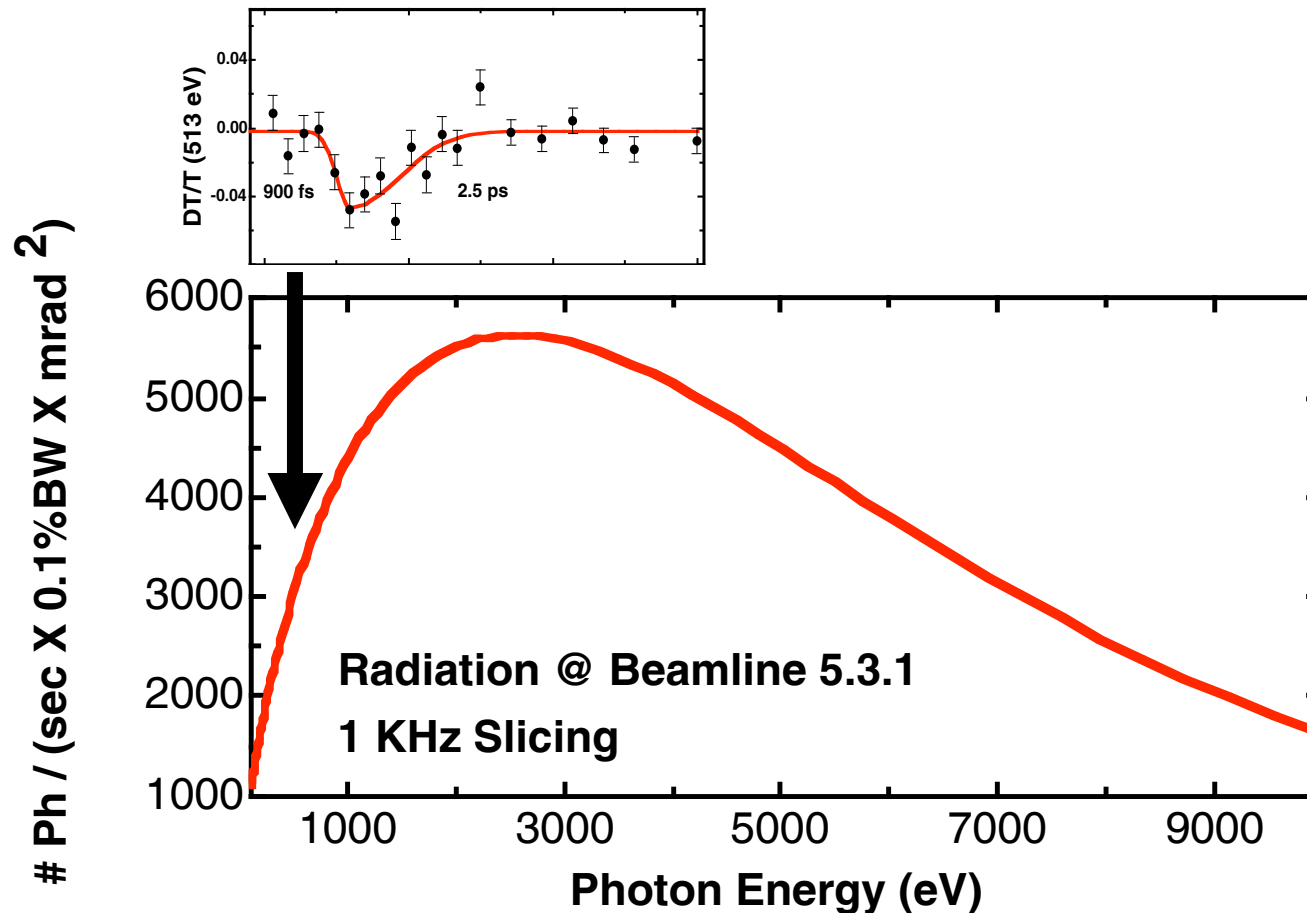
Hole photo-doping



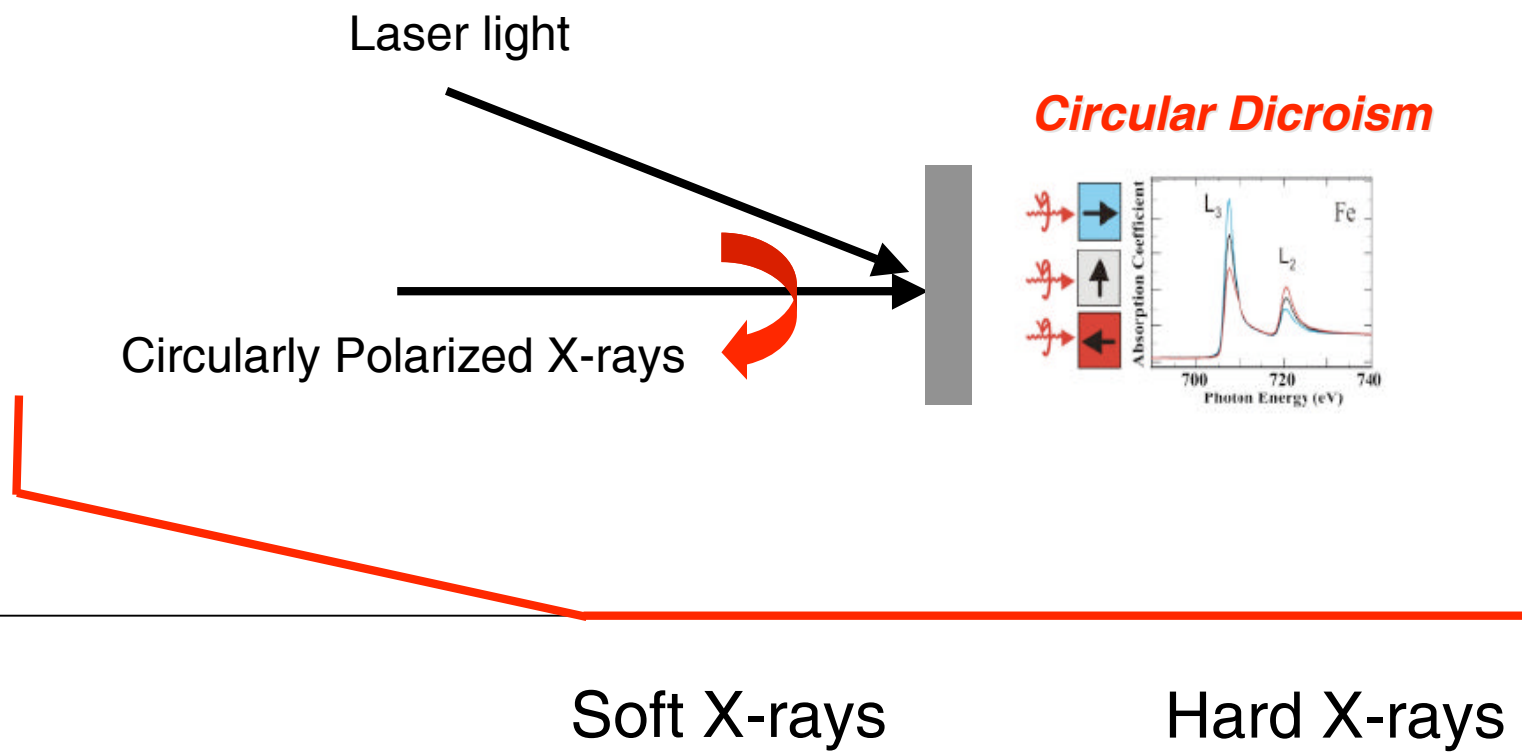
Technical Significance



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



Ultrafast X-rays: New Opportunities



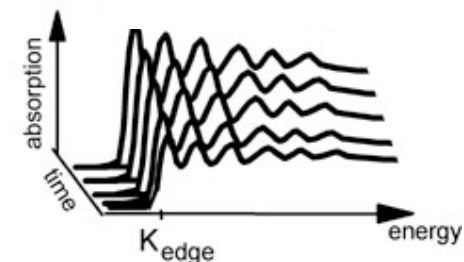
Ultrafast X-rays: New Opportunities



Laser light
Tunable x-rays



EXAFS



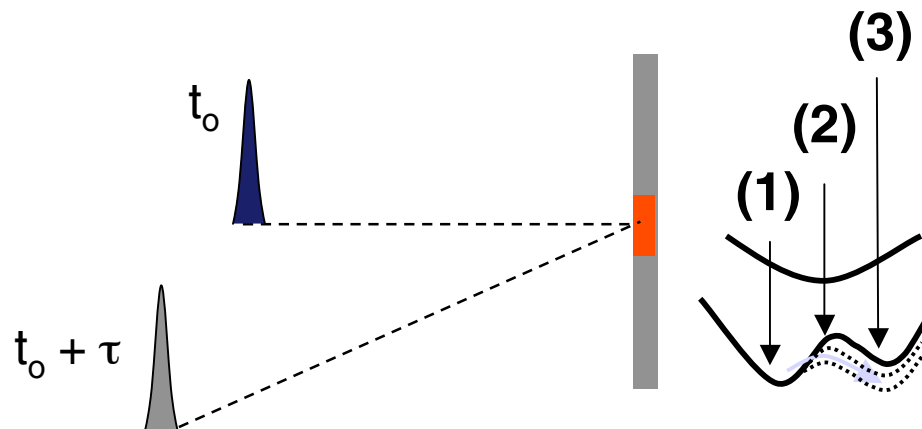
Soft X-rays

Hard X-rays

Summary



1) Driving phase transitions **impulsively** and probing them **dynamically** uncovers **fundamental microscopic physics**



2) **Sub-vibrational** measurements **assign cause and effect**

3) It is **essential** to have both **spectroscopy** and **diffraction** probes on the femtosecond timescale