Photo-induced Structural Dynamics in VO_2 probed with Fs X-rays

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Femtosecond X-ray Diffraction with Laser-Plasma Sources

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J.A. Squier

University of California, San Diego

J.C. Kieffer

University of Quebec

Femtosecond EXAFS with "Sliced" Syncrotron Radiation

R.W. Schoenlein,
H.H. Chong,
C.V. Shank

Materials Sciences Division, LBNL

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

Advanced Light Source, LBNL

A.M. Lindenberg

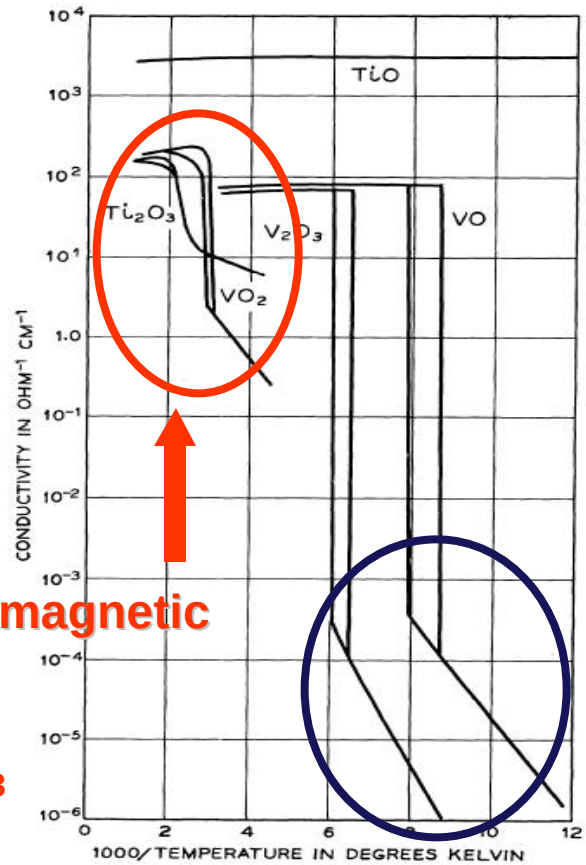
UC Berkeley

Metal-Insulator Transition in Oxides

VOLUME 3, NUMBER 1 PHYSICAL REVIEW LETTERS JULY 1, 1959

OXIDES WHICH SHOW A METAL-TO-INSULATOR TRANSITION AT THE NEEL TEMPERATURE

F. J. Morin
Bell Telephone Laboratories, Murray Hill, New Jersey
(Received June 5, 1959)



Non-magnetic

VO_2

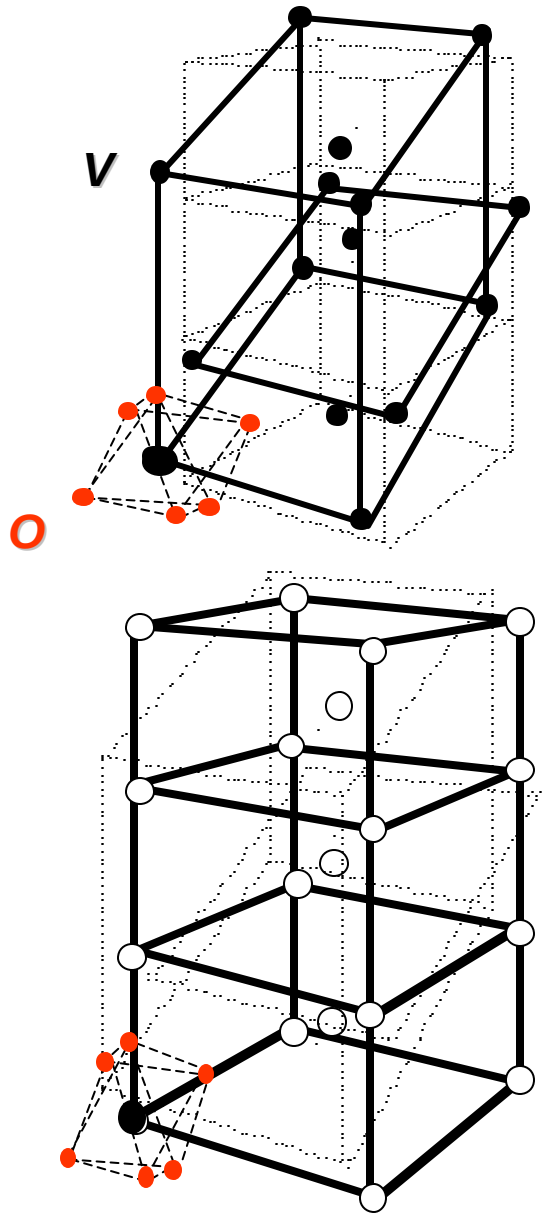
Ti_2O_3

Anti-ferromagnets

V_2O_3

VO

Structural + Electronic transition in VO_2



T < 340 K

Monoclinic ***Insulator***

T > 340 K

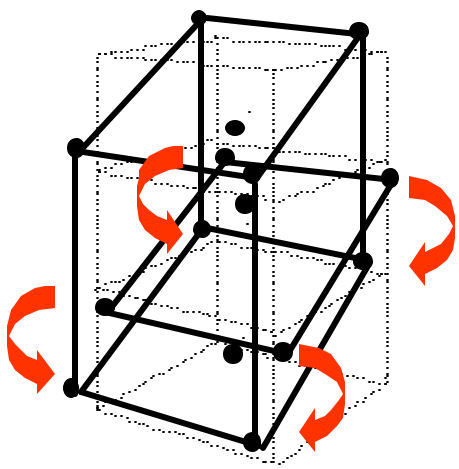
Rutile ***Metallic***

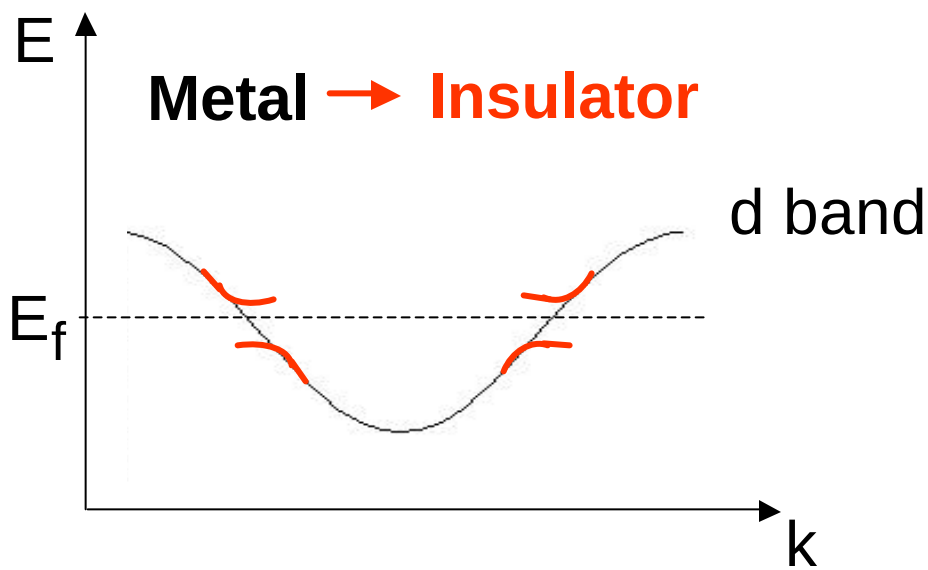
VO_2 : History

Band insulator?

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

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





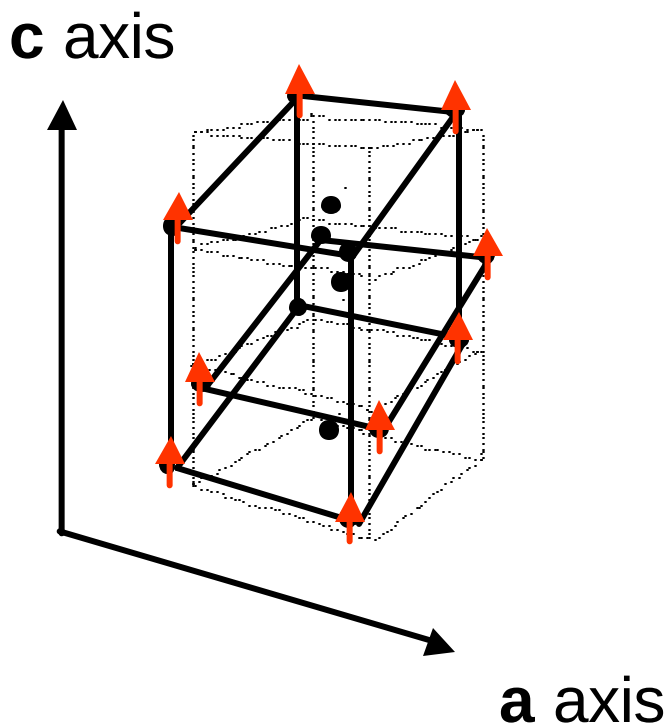
VO_2 : History

Mott-Hubbard insulator?

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

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

S. Shin et al. *Phys. Rev. Lett.* 41, 4993 (1990) **(UPS)**



On site Coulomb Repulsion

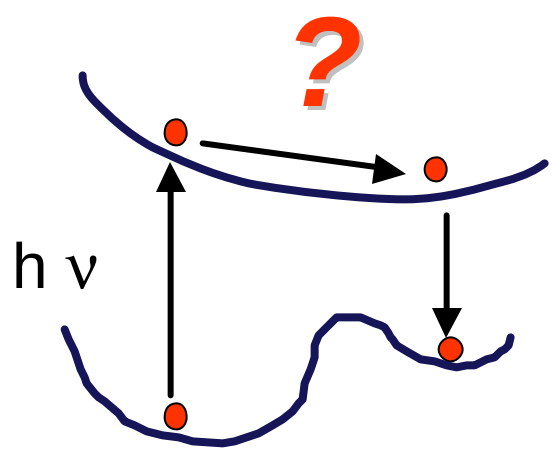
Hubbard U

Correlated Bandgap

VO_2: Photo-induced phase transition

Temperature driven: strongly first order

Photo-induced



VO_2 : Optical non-linearities

Mott-Hubbard insulators

letters to nature *June 22, 2000*

Gigantic optical nonlinearity in one-dimensional Mott–Hubbard insulators

H. Kishida*, H. Matsuzaki*, H. Okamoto*†, T. Manabe‡, M. Yamashita§||, Y. Taguchi¶ & Y. Tokura¶#

VO_2

THG

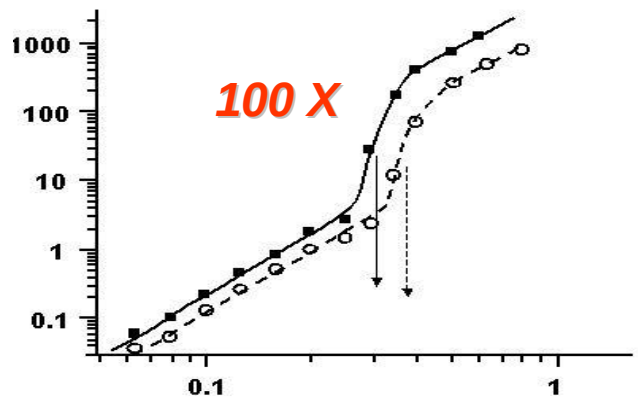
30 X

10

1

Temp

THG



Pulse Energy

Courtesy of Jeff Squier, UC San Diego

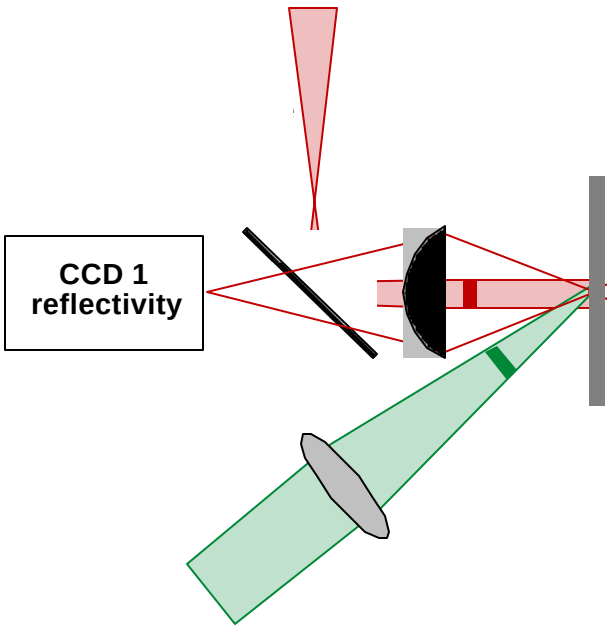
Questions: photo-induced phase transition

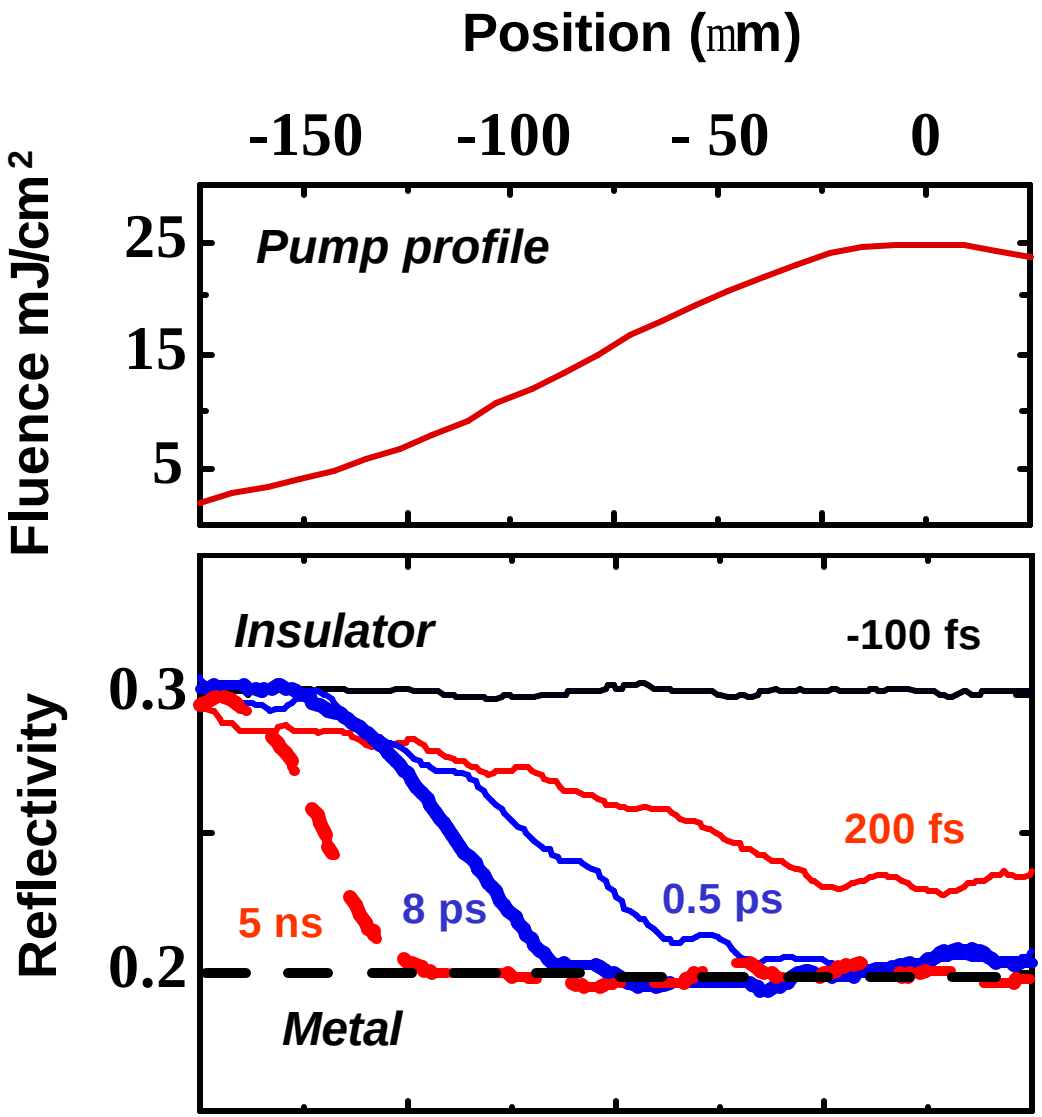
How fast are electrical and structural transitions?

What distortions initiate the structural transition?

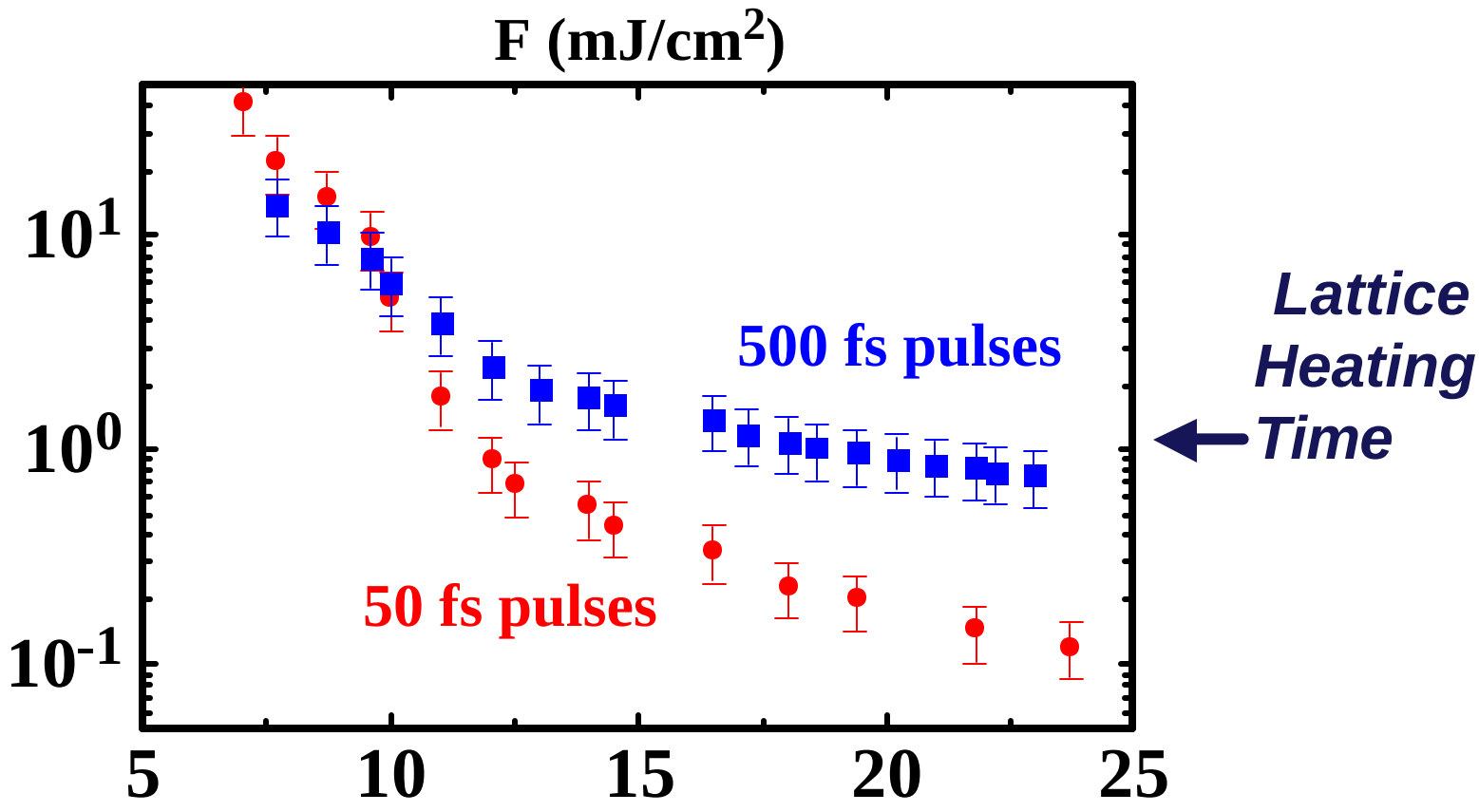
How are the two transitions related ?

Insulator to Metal: How fast?

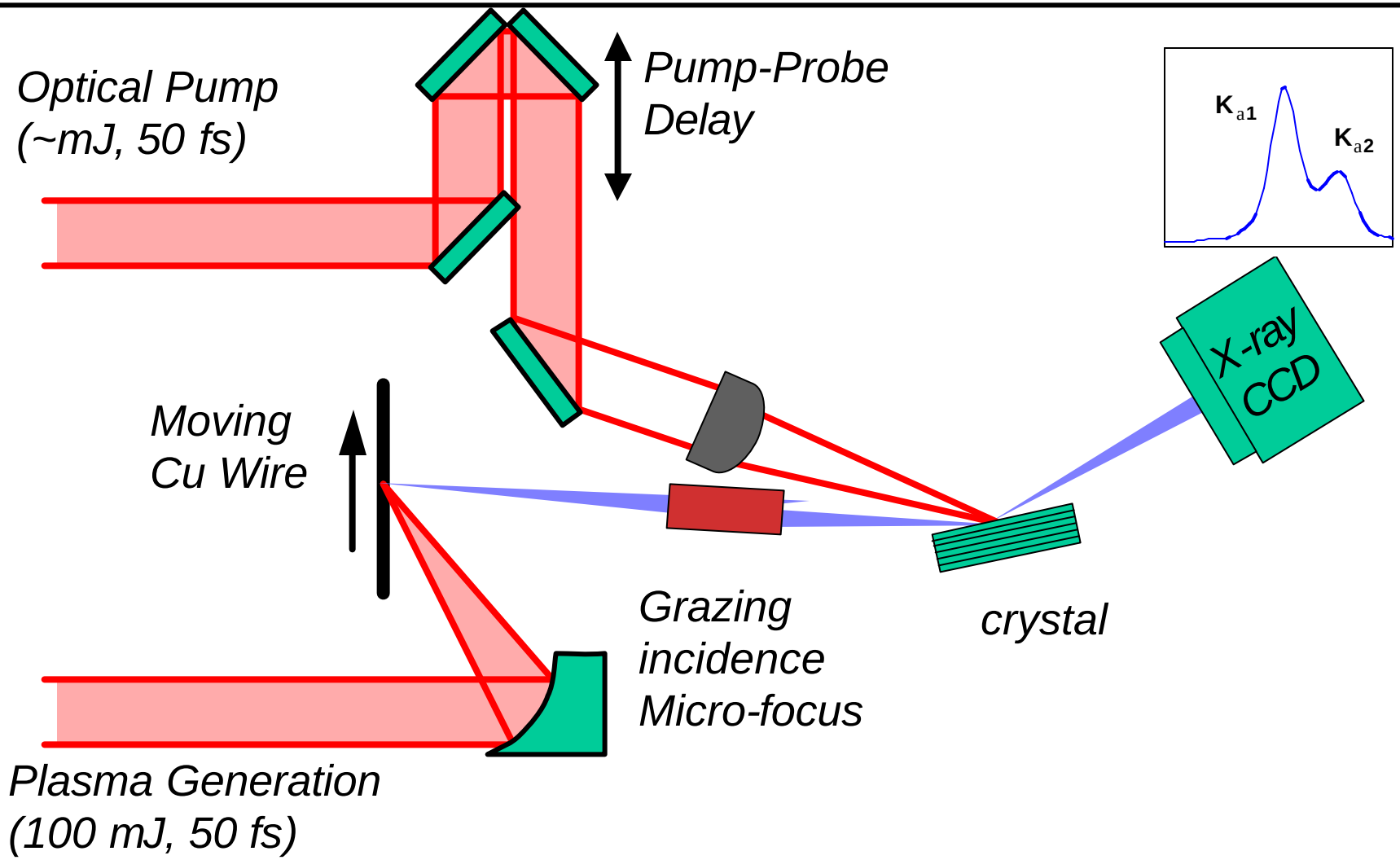




Insulator to Metal: Temperature driven?



Pump-probe setup: VO_2

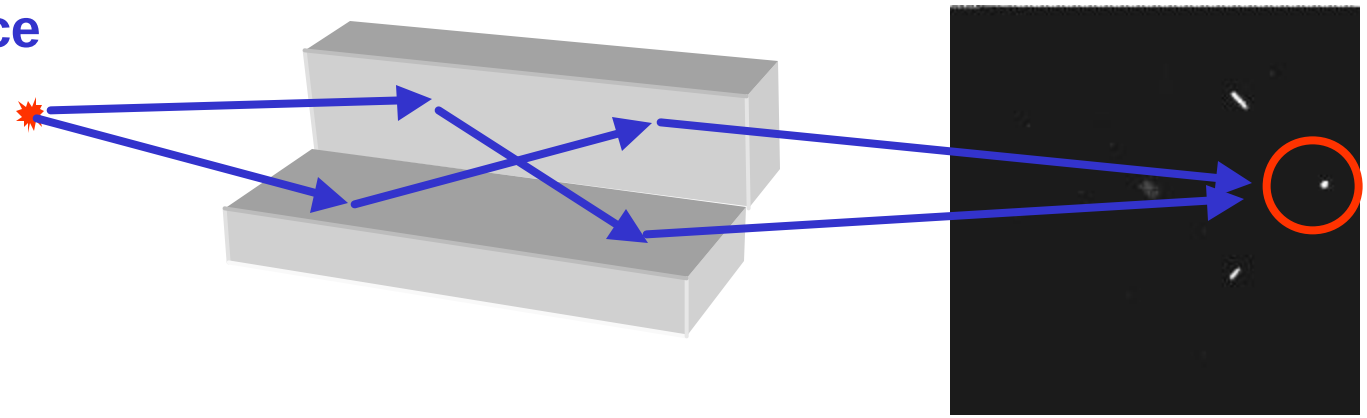


X-ray Focusing

Elliptical surfaces

with graded Bragg coating

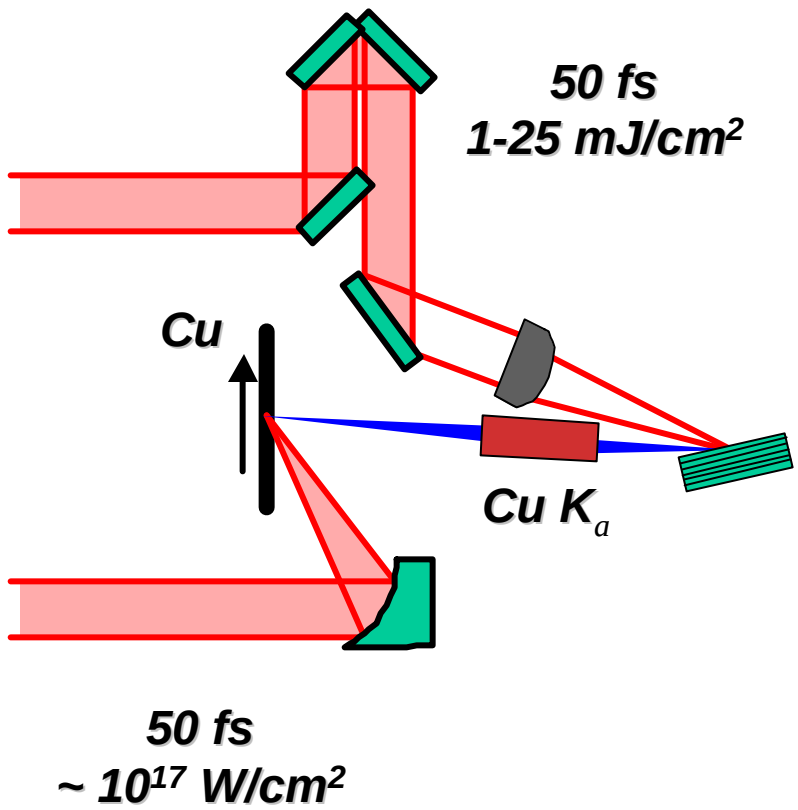
X-ray source

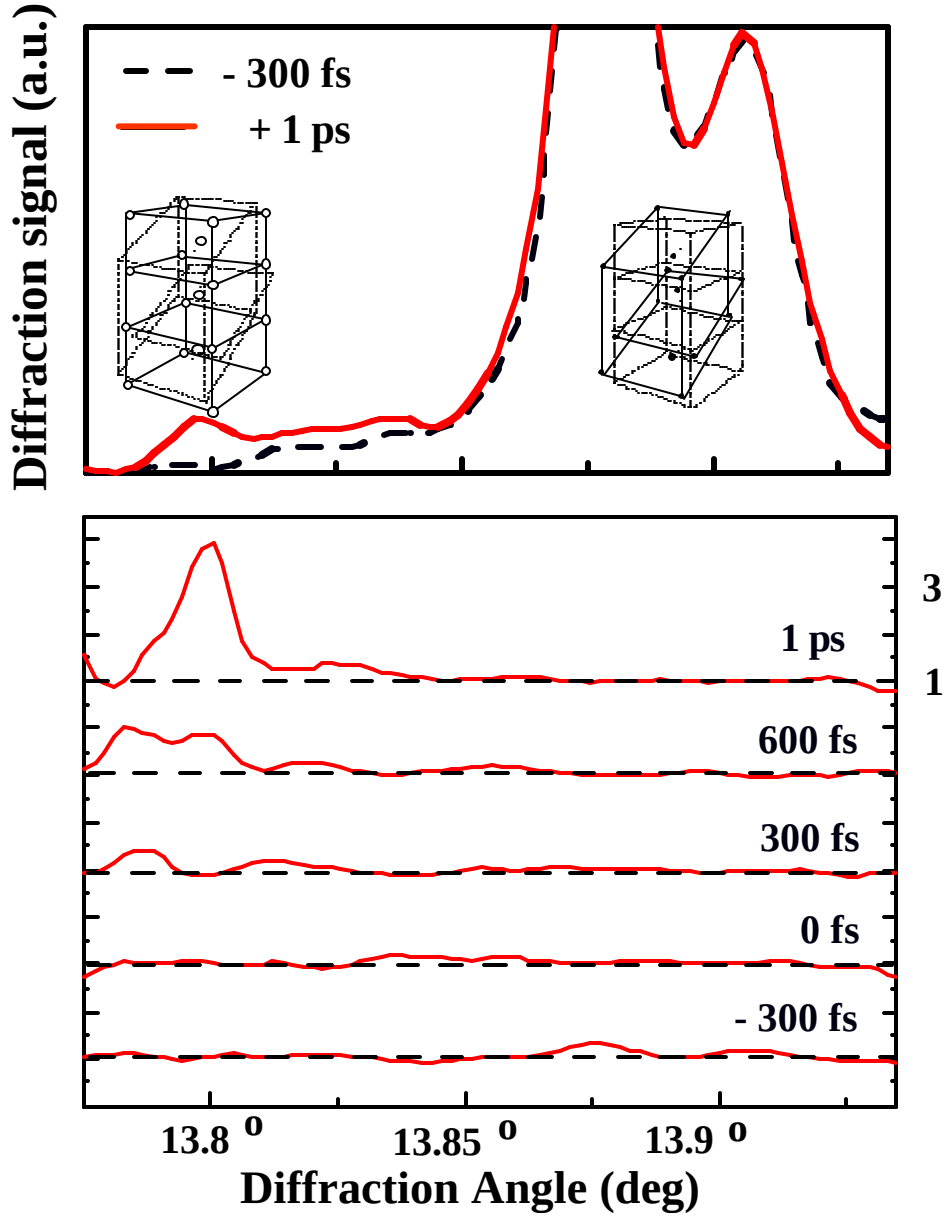


Spot diameter = 54 μm

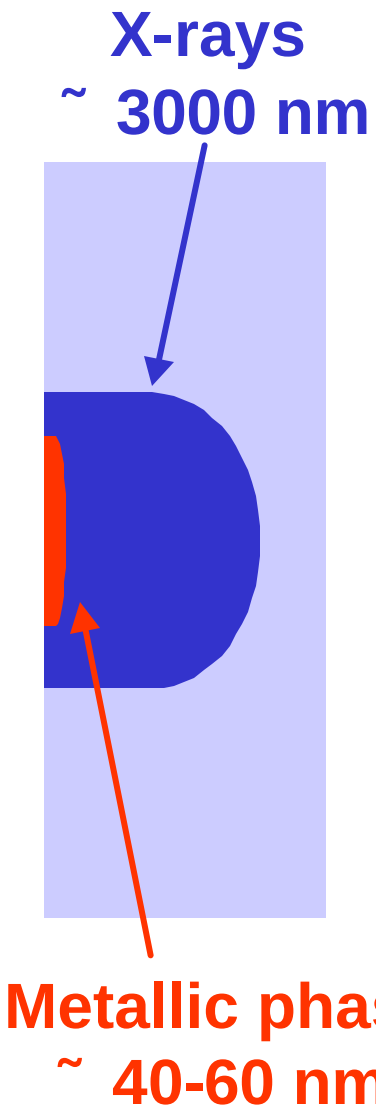
2000 photons / shot

Monoclinic to Rutile (110): How fast ?

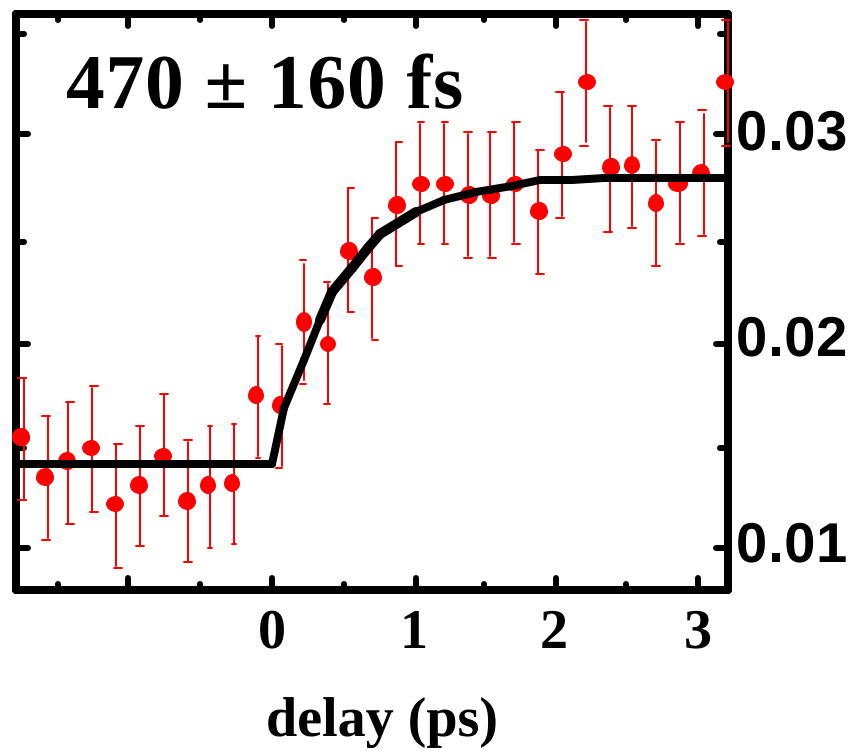




Monoclinic to Rutile



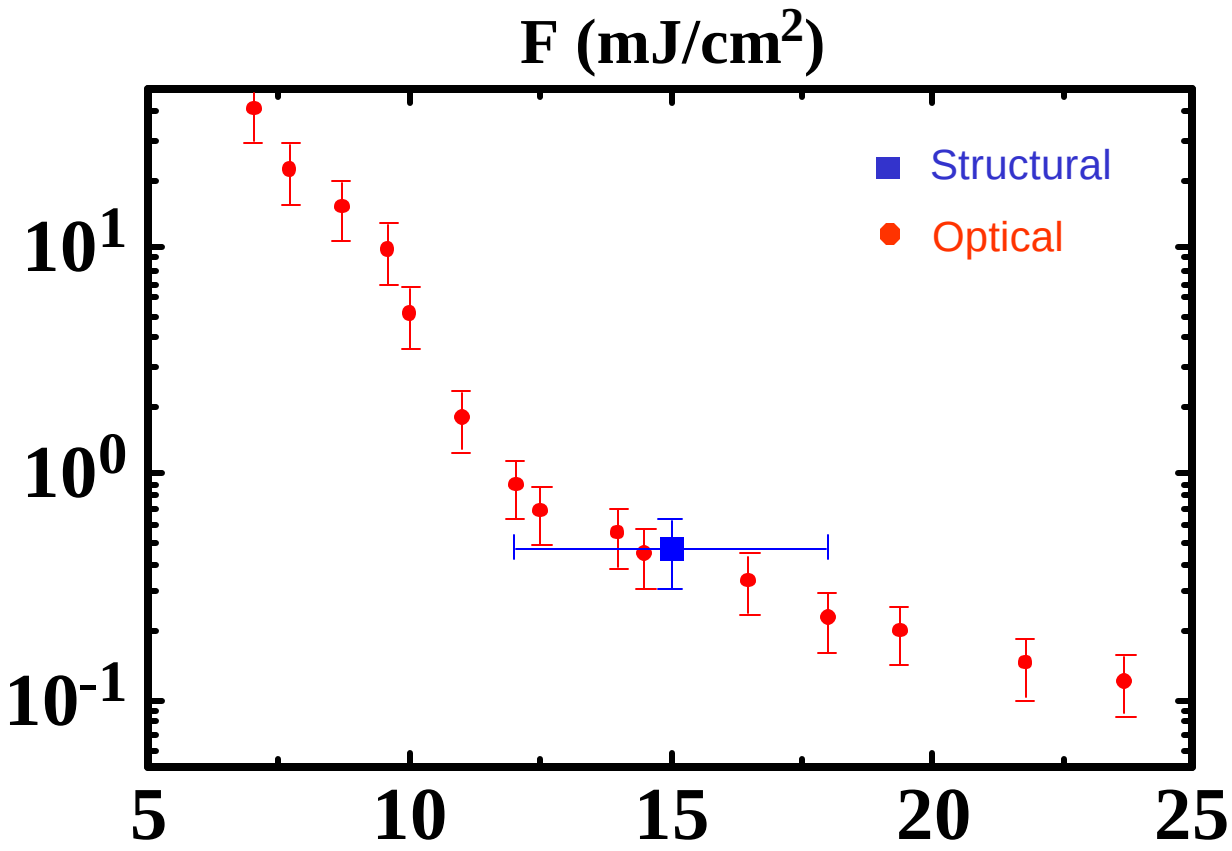
✓ **Femtosecond Solid-Solid Transition !**



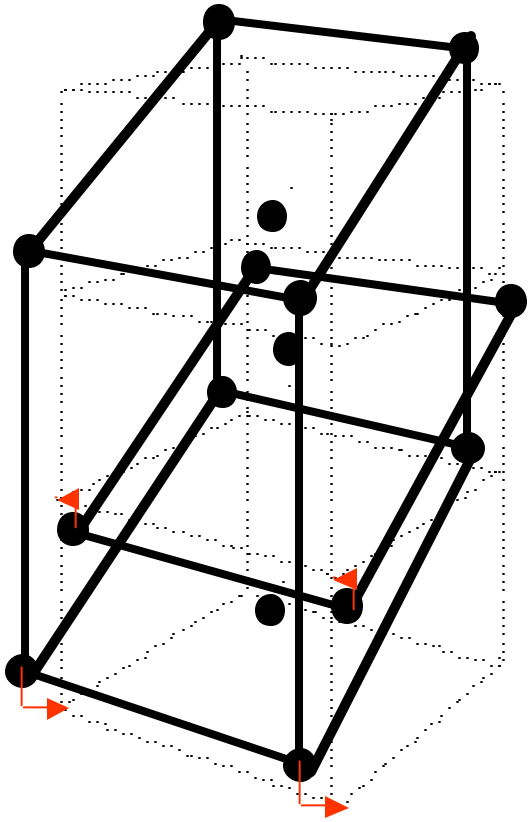
A. Cavalleri et al. *Phys. Rev. Lett* 87, 237401 (2001)

Optical vs Structural Rate

Simultaneous transitions



Optical Phonons drive the Transition



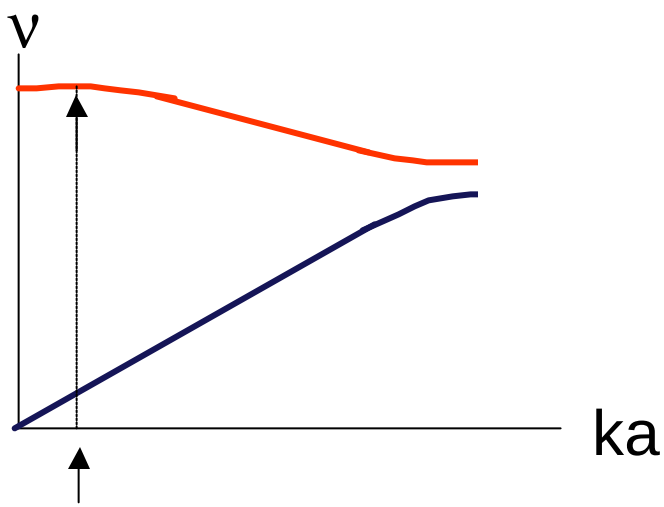
Long-range order change

k ~ 2 10^{5} cm^{-1}

t ~ 1 ps

Optical phonons

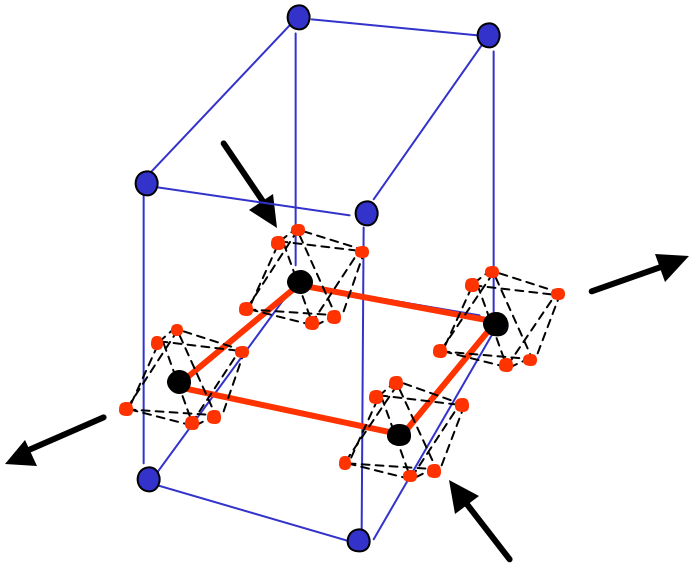
~ THz

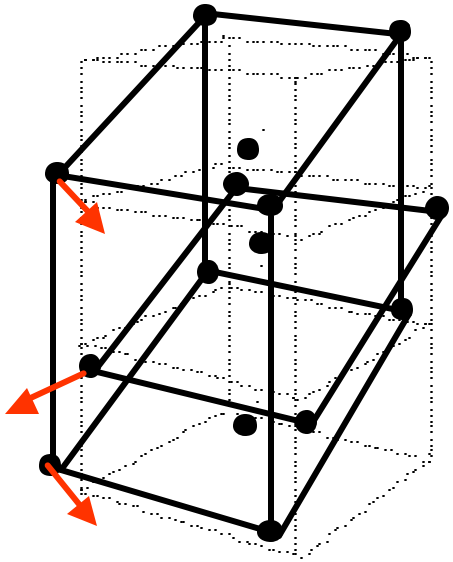


~~***ka ~ 6 10^{-3}***~~

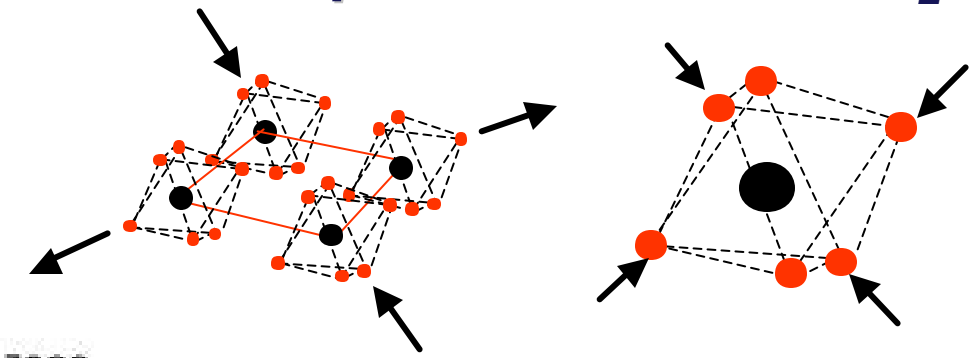
~~***~ 100 GHz***~~

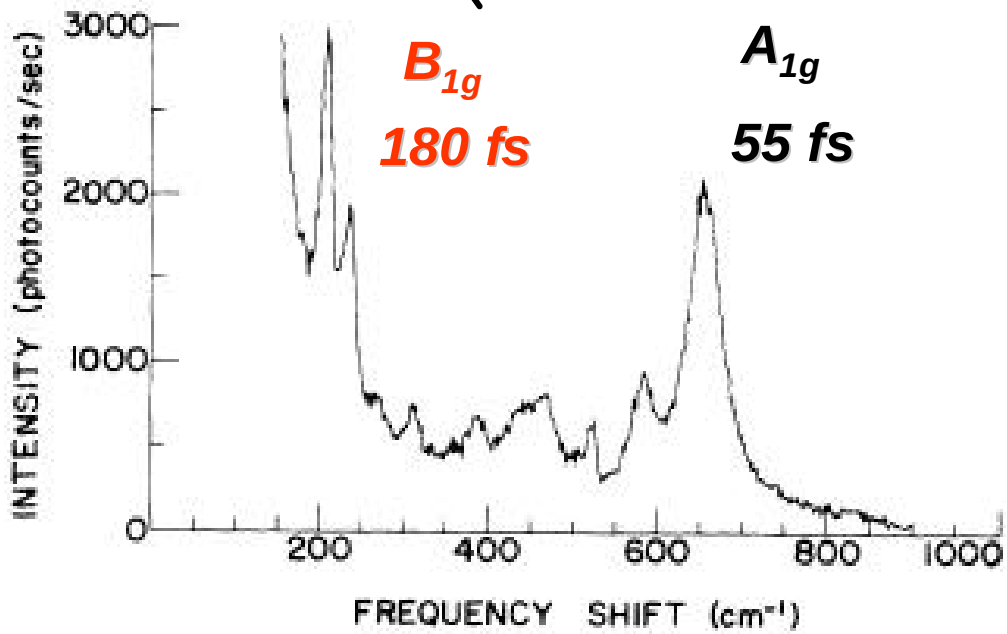
Optical Phonons drive the Transition





Raman Spectrum Low -T VO_2





Ramakant Srivastava, L.L. Chase, Phys. Rev. Lett. 727, 27 (1971)

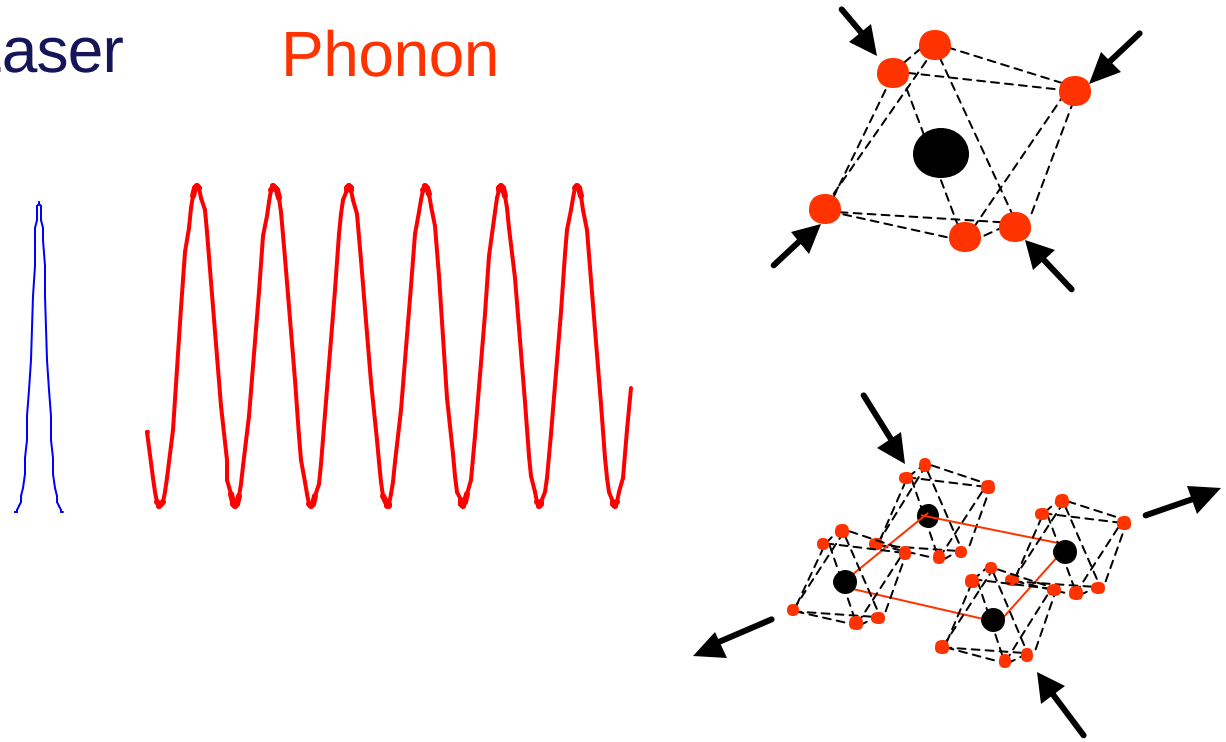
Raman Excitation?

$t_{pulse} < t_{phonon}$

$t_{pulse} = 50$ fs

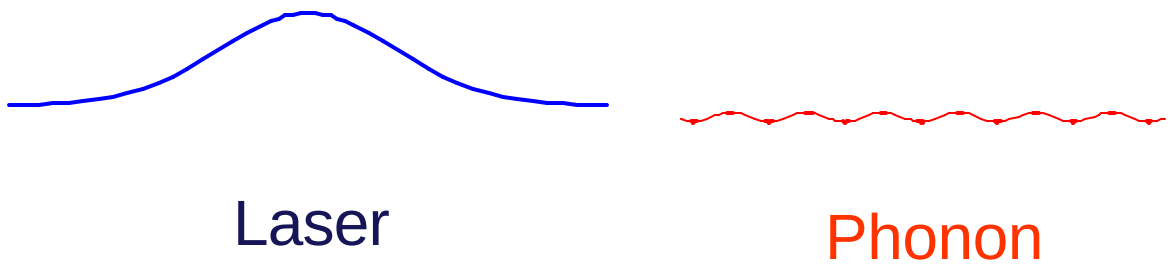
$t_{ph1} = 180$ fs

$t_{ph2} = 55$ fs



$t_{pulse} > t_{phonon}$

$t_{pulse} = 500$ fs

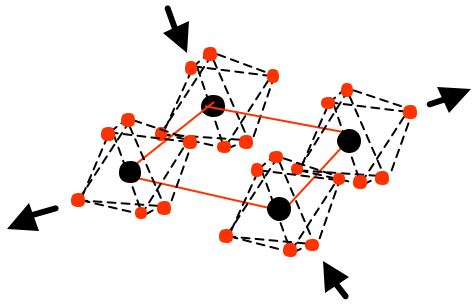


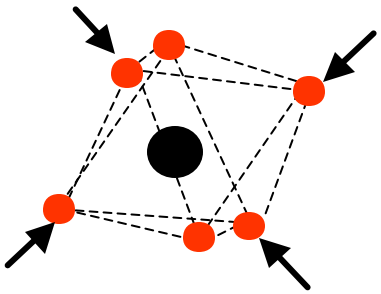
Sensitivity to short-range dynamics

Needed: measurement of unit cell dynamics

Measurement of short-range structure

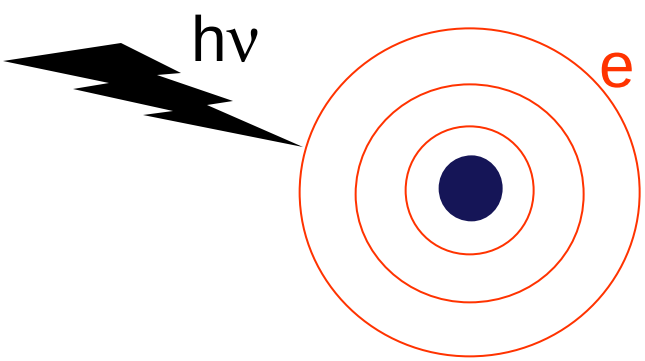
Sensitivity to Light Elements (Oxygen)

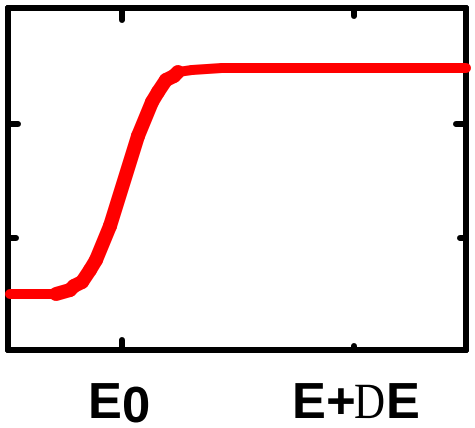




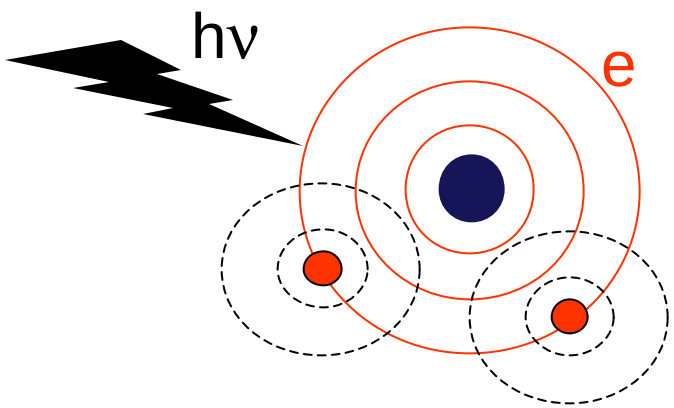
Quantum Interference: EXAFS

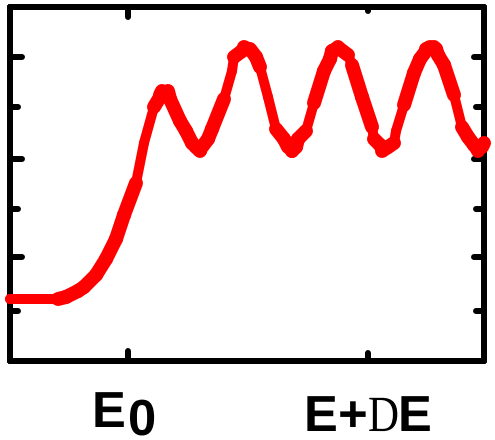
Isolated Atom





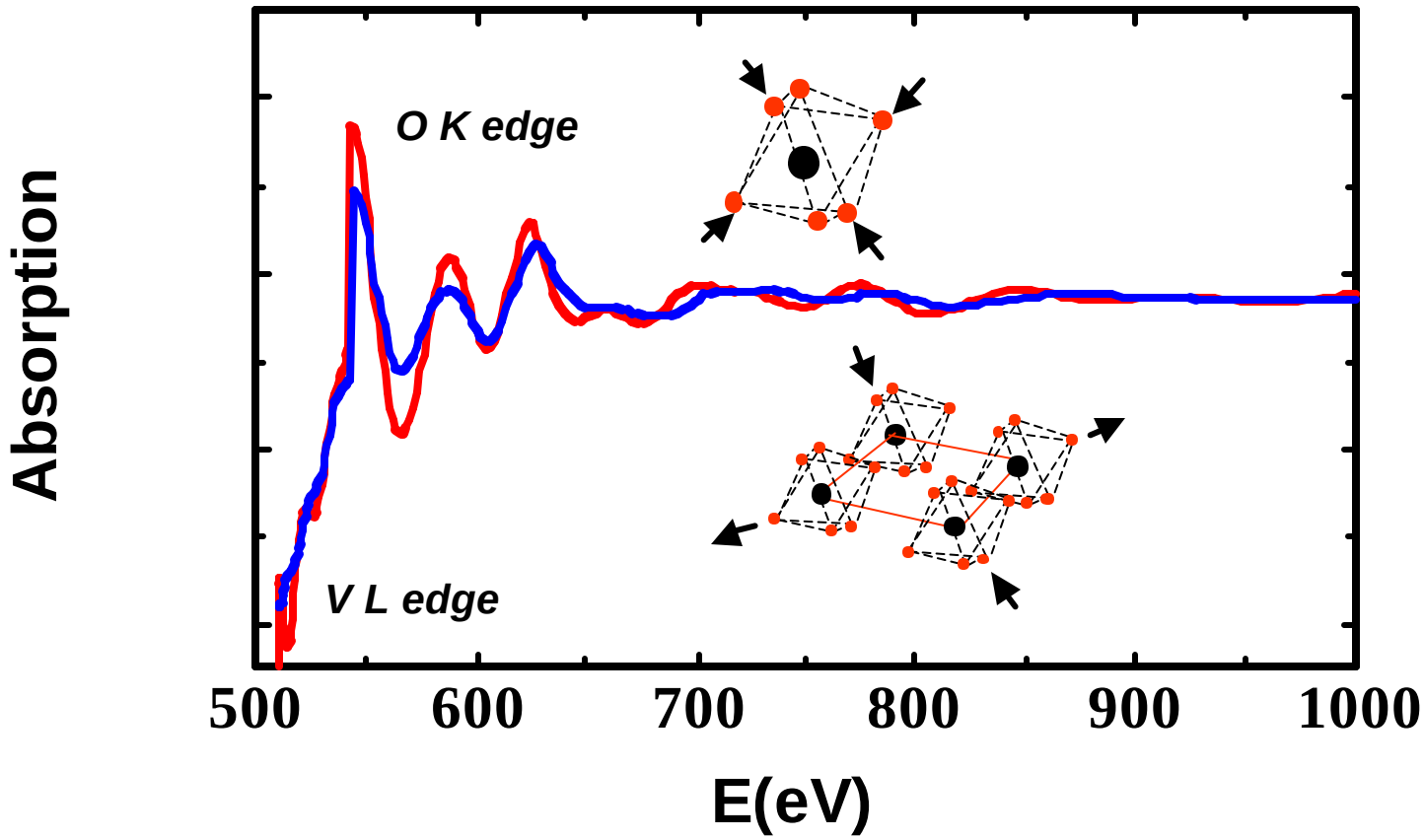
Atom + Neighbors



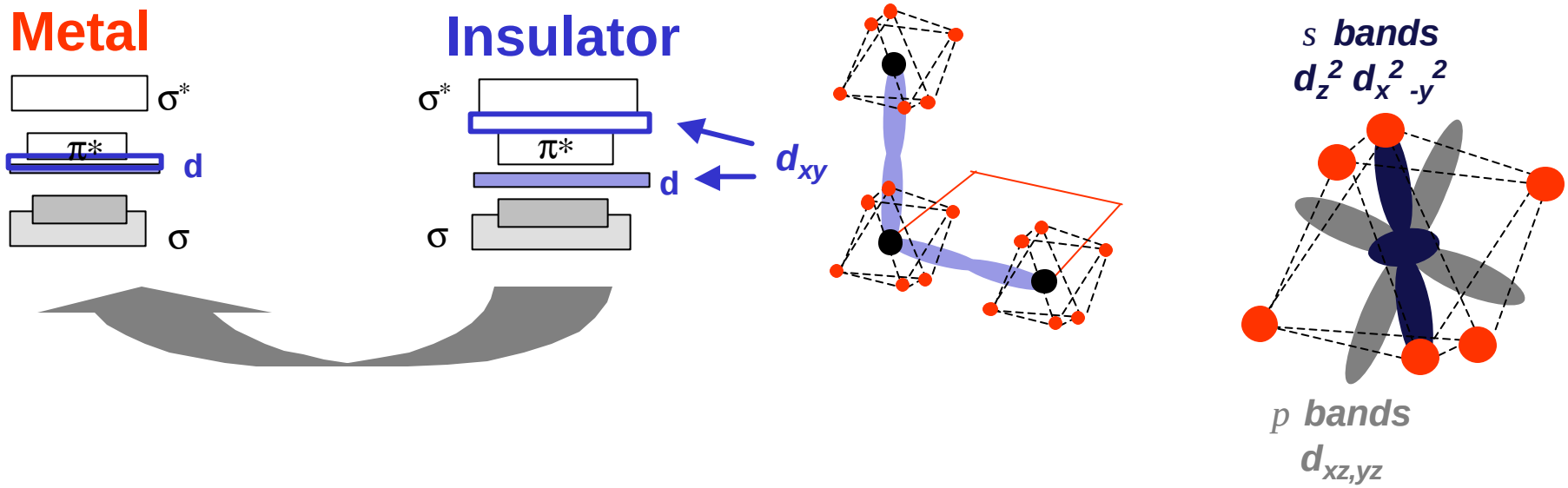


VO_2: Calculated EXAFS

Low-symmetry (Low T) → ***High symmetry (High T)***



Metal-Insulator Transition: NEXAFS



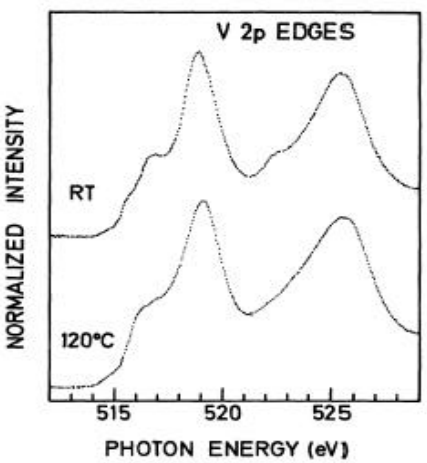


FIG. 3. V 2*p* absorption spectra of VO_2 taken at room temperature and $T = 120\,^\circ$C.

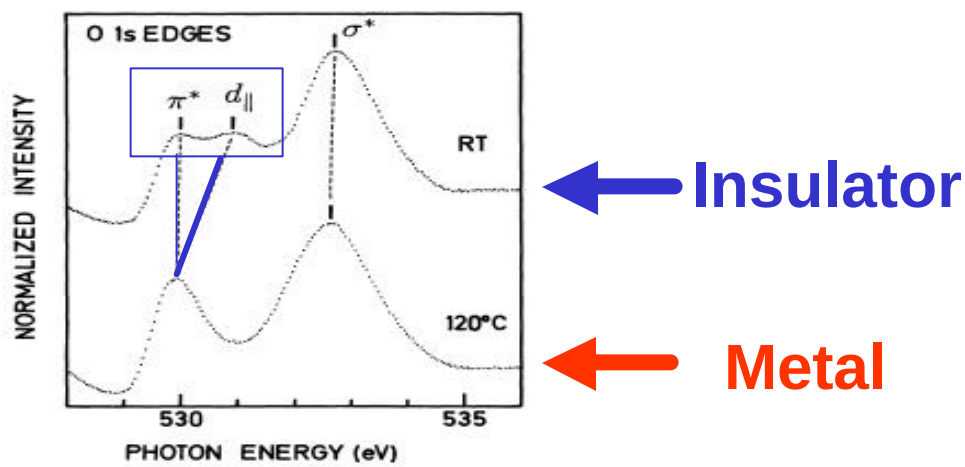
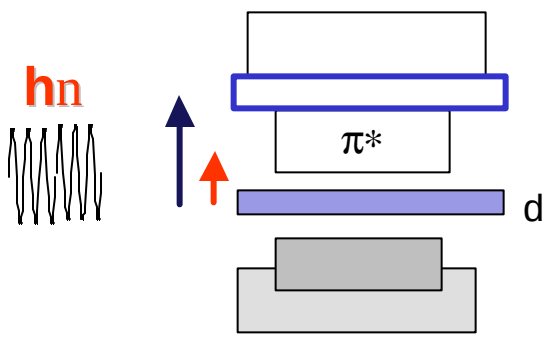


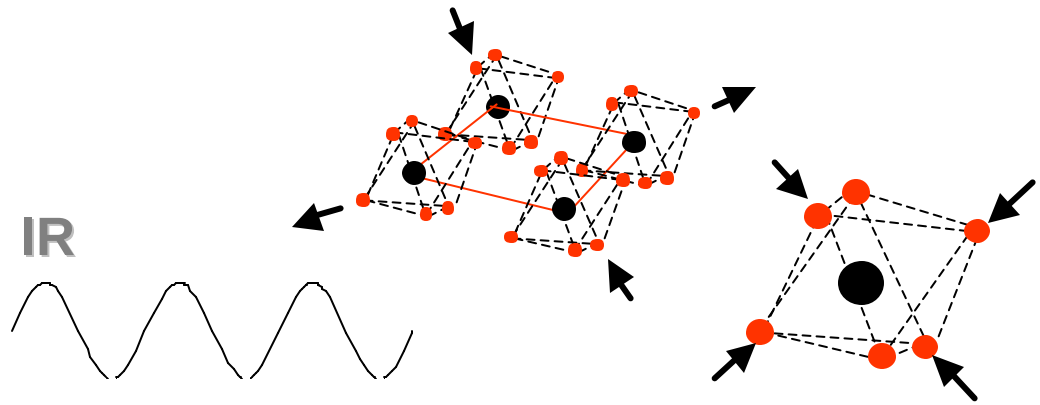
FIG. 1. O 1*s* absorption spectra of VO_2 taken at room temperature and $T = 120\,^\circ$C.

Abbate et al. *Phys. Rev. B* 43 (1991)

What are we after?

Manipulate excitation





Probe electronic and structural transitions

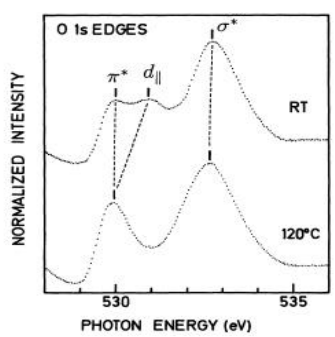
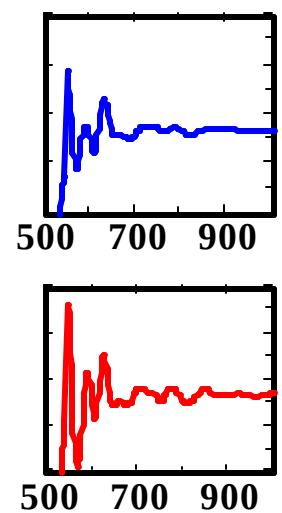
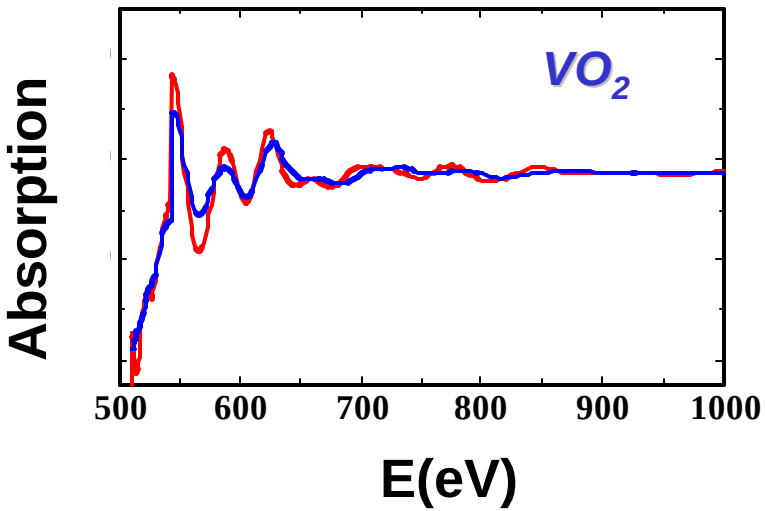


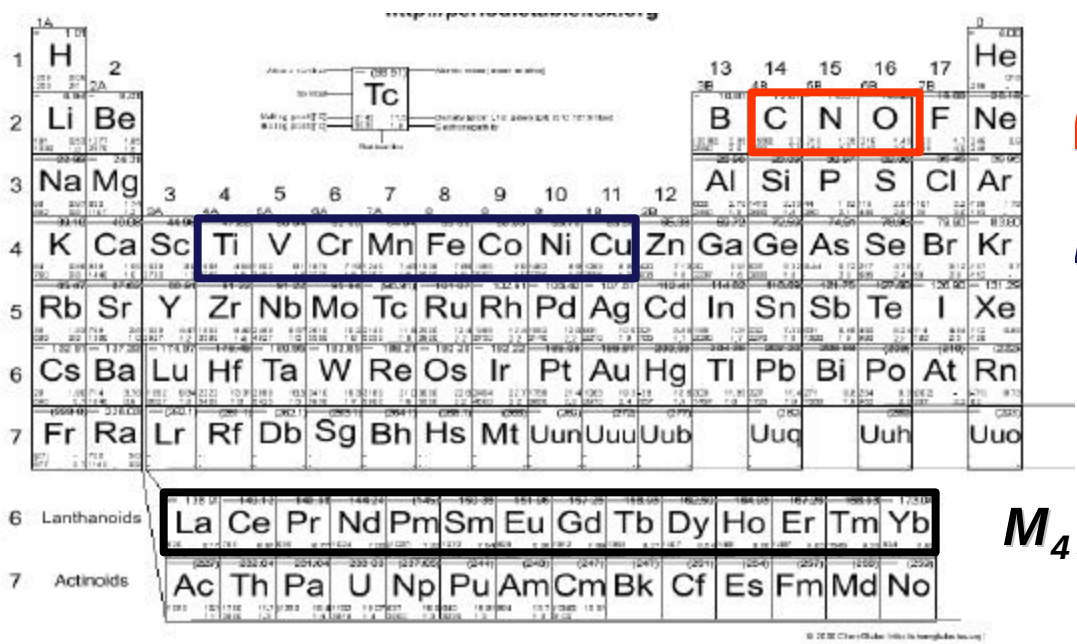
FIG. 1. O 1s absorption spectra of VO_2 taken at room temperature and $T = 120$ °C.



Transition Metal Oxide Spectroscopy: Source

- ✓ **Broadband**
- ✓ **Soft x-rays (electrons)**



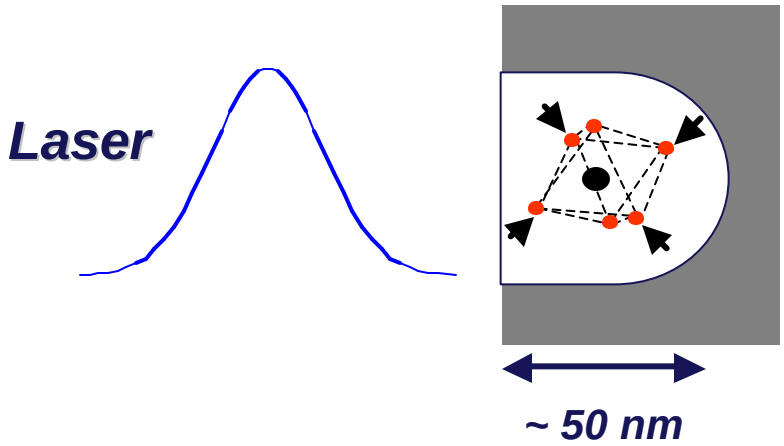


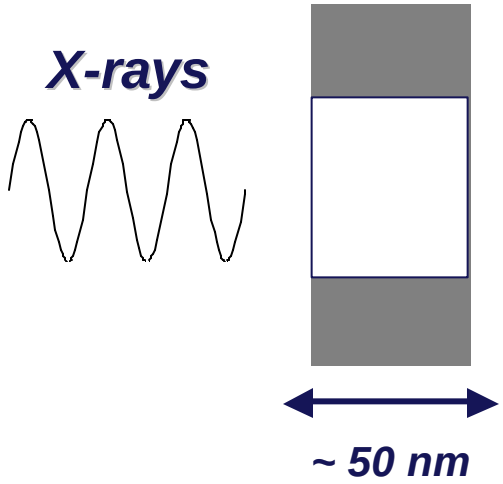
K edge (250 eV - 1 keV)

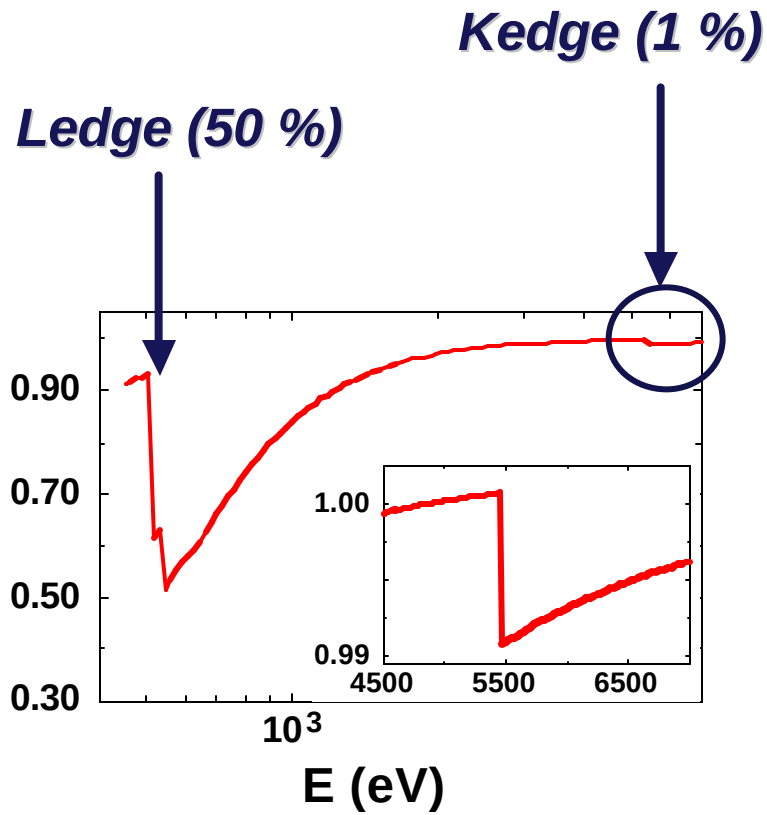
L edge (400 eV - 4.5 keV)

M_4, M_5 edge (700 - 2 keV)

L edge vs K edge



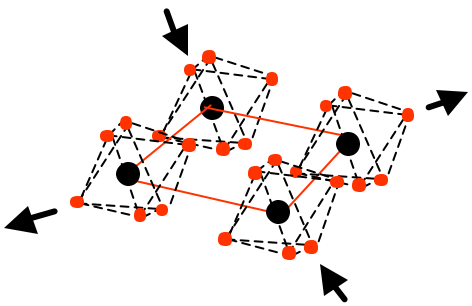




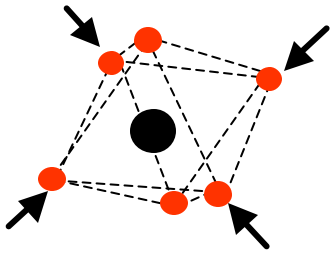
Ideal x-ray source

✓ **100 fs or shorter, synchronized to laser**

180 fs



50 fs



? 10 fs - 500 fs ?

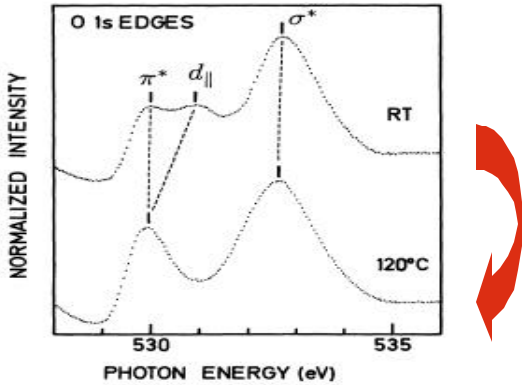
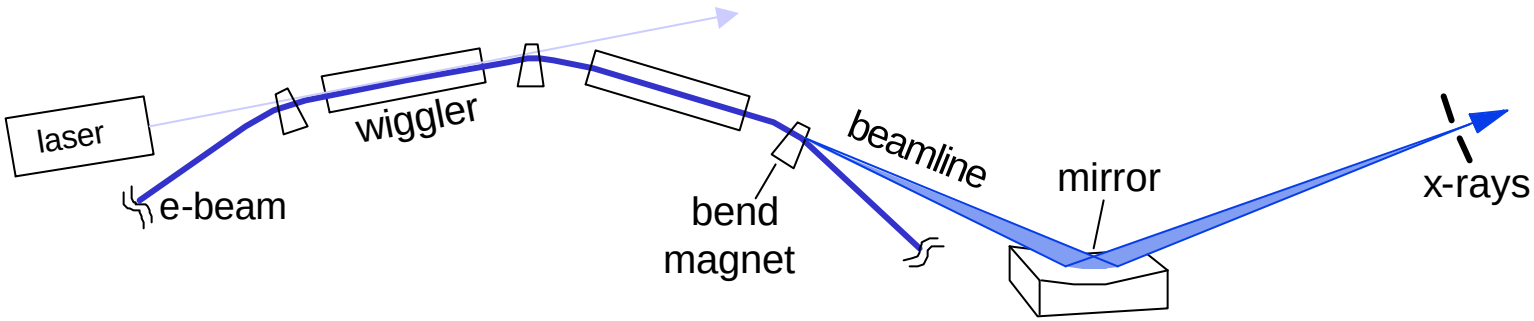


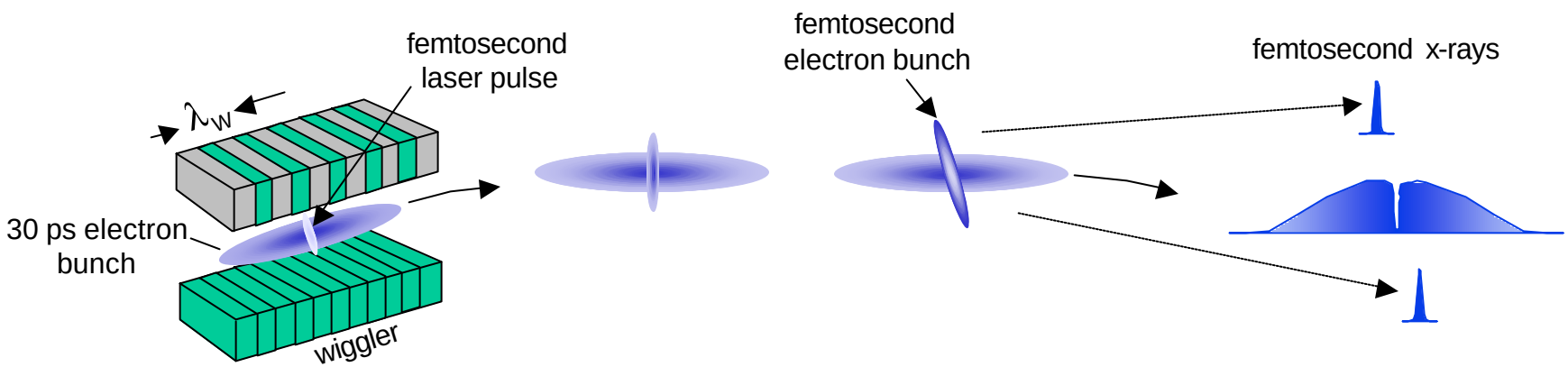
FIG. 1. O 1s absorption spectra of VO_2 taken at room temperature and $T = 120$ °C.

✓ **Rep rate: 1 Hz - few tens of KHz**

- **Sample recovery ~ 1 msec**
- **Average Heating**

Tunable femtosecond X-rays at the ALS





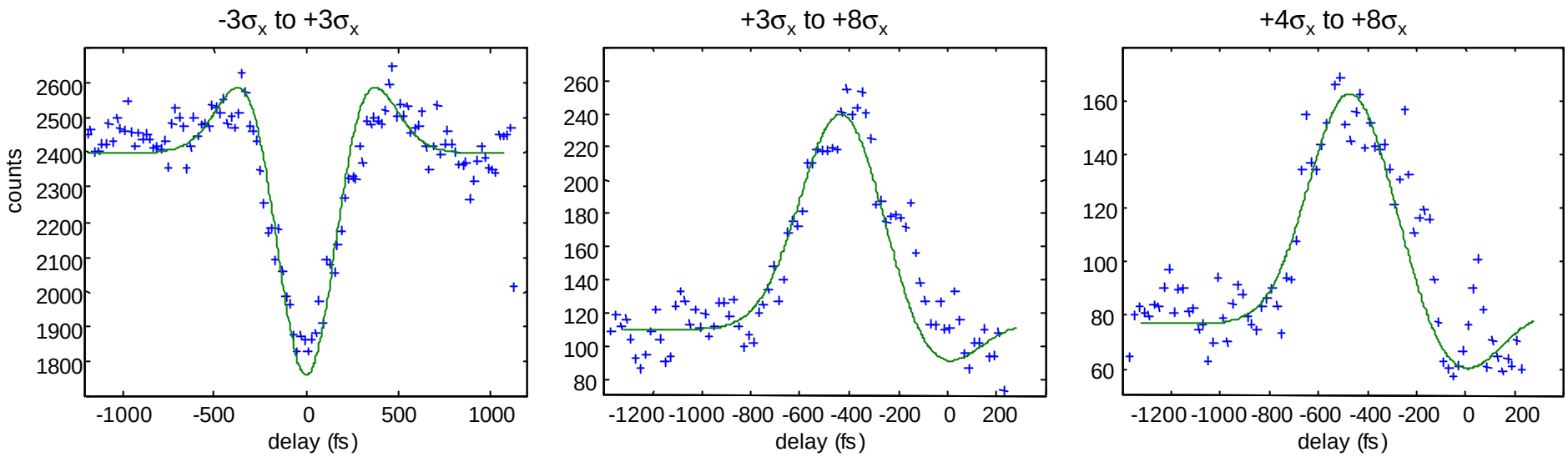
electron-photon interaction in wiggler

spatial separation dispersive bend

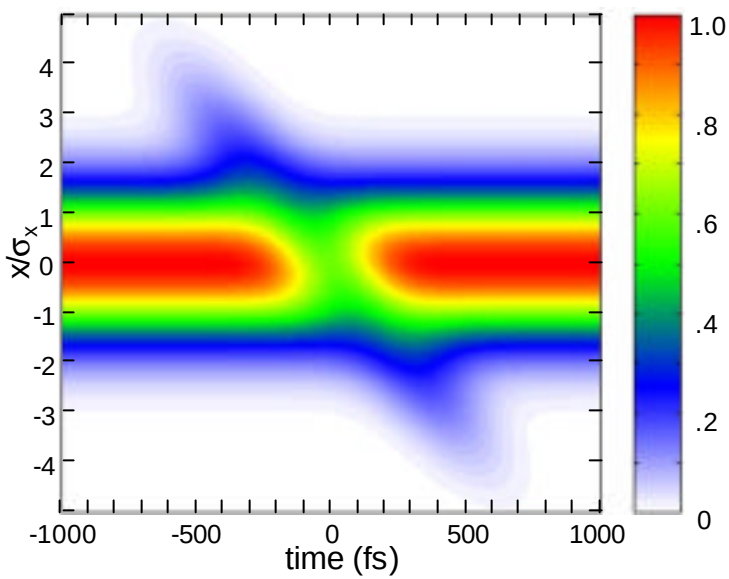
bend magnet beamline

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

Fs Pulses of Synchrotron Radiation

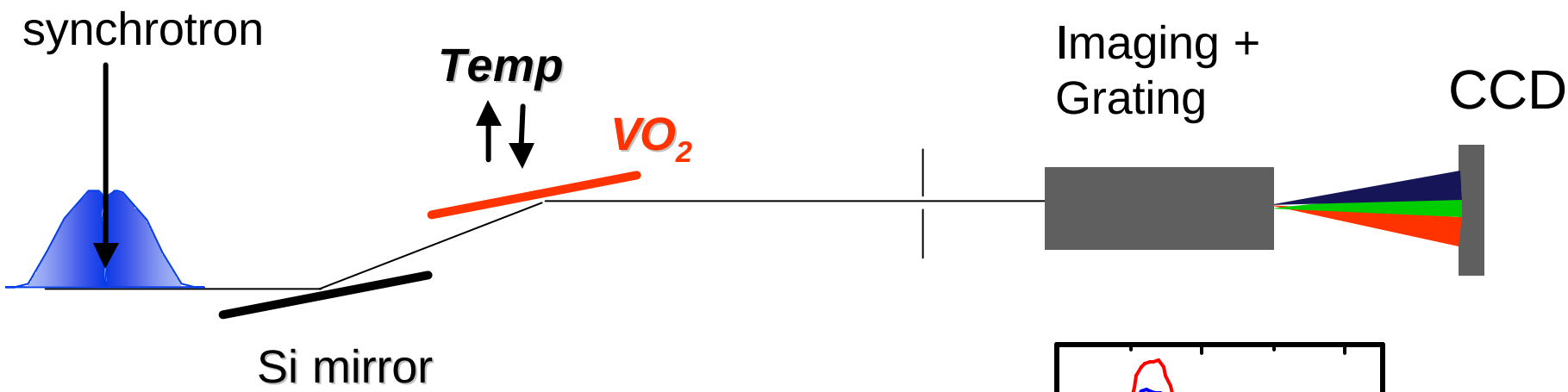


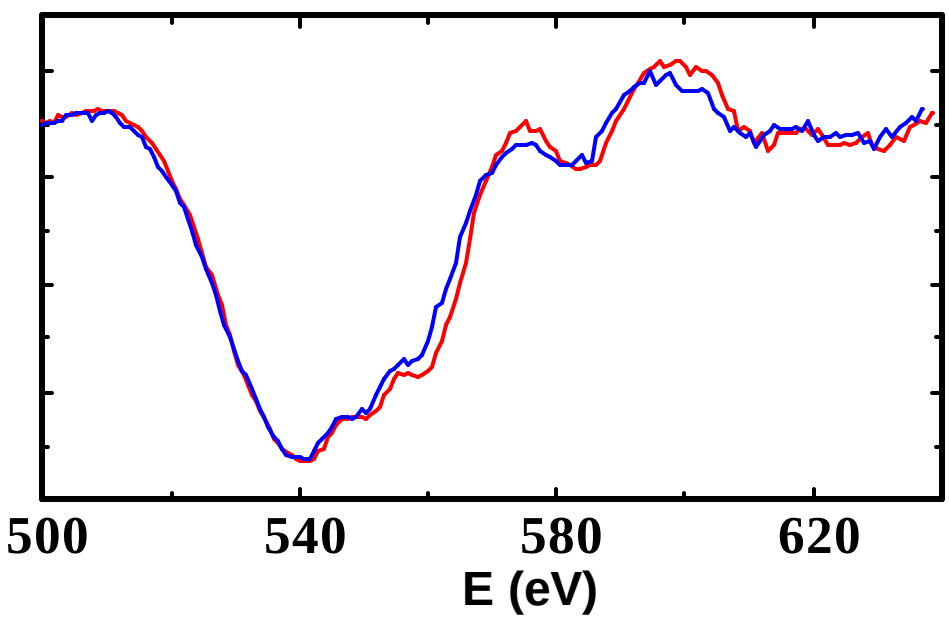
Calculated Electron Density Distribution

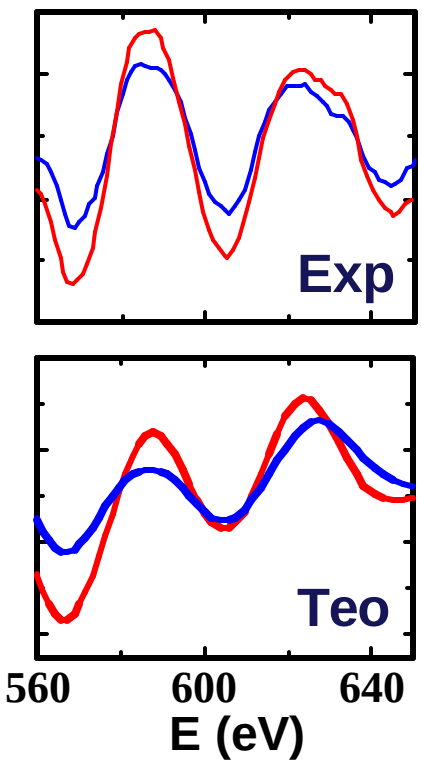


Schoenlein et al., *Science,* **287**,2000

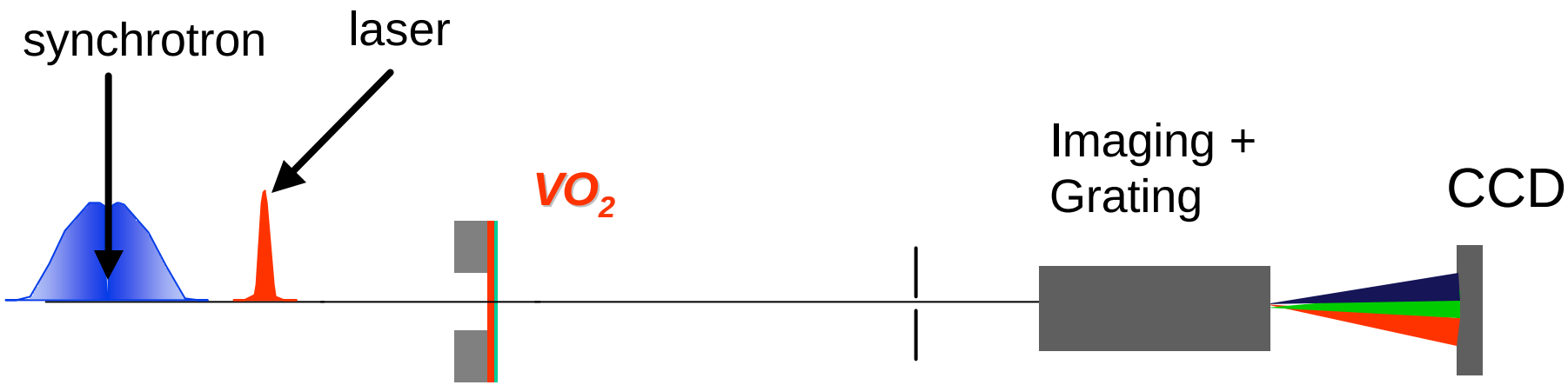
Static Reflectivity EXAFS: Preliminary

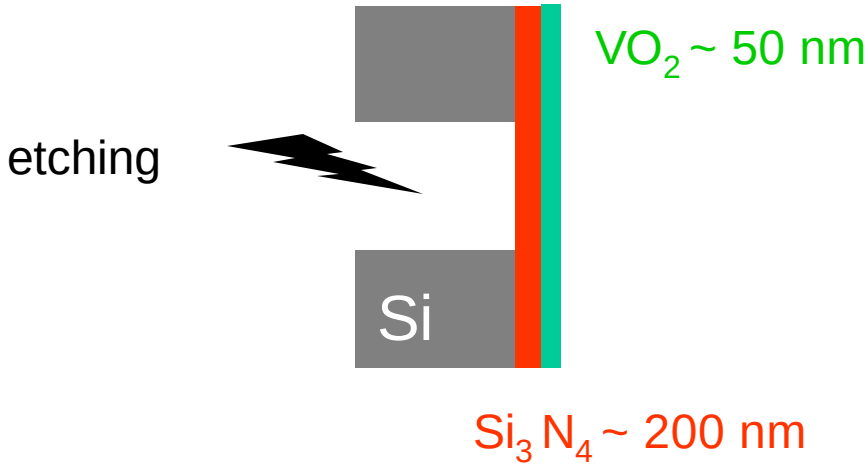






Needed: transmission sample





Apparatus requirements

✓ **Flux requirements**

EXAFS (S/N ~10)	10^4 photons / 0.1 % BW
EXAFS (S/N ~ 100)	10^6 photons / 0.1 % BW

↓

Detected photons / time point

↓

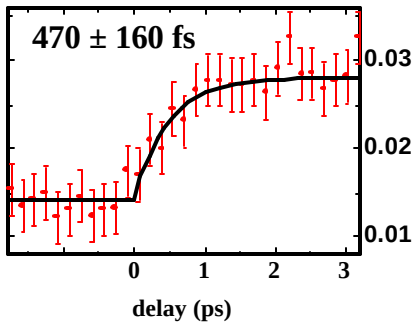
Minutes to few hours for whole experiment

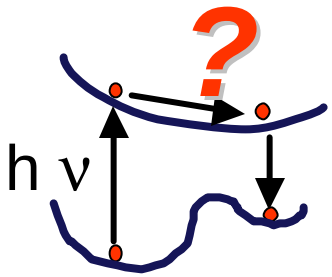
✓ **Spectrometer Resolution**

EXAFS	~ 2 eV
NEXAFS	< 500 meV

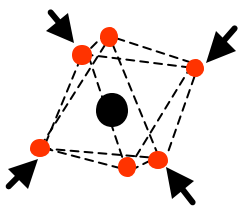
Conclusion

✓ ***Photo-induced phase transitions are ultrafast***





✓ ***Role of distortions in the electronic transitions***



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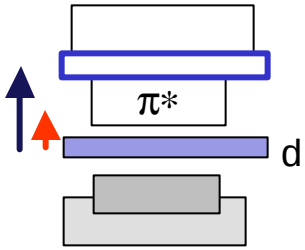
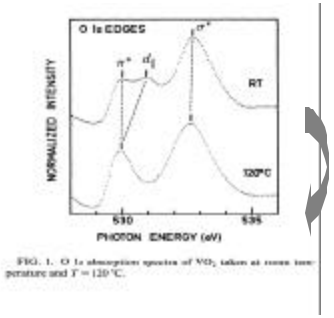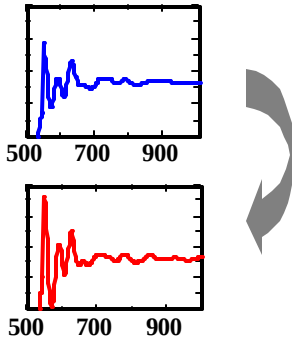
✓ ***Femtosecond x-ray absorption spectroscopy***