

**Ultrafast electron dynamics in
strongly correlated materials:
all photon techniques using X-rays**

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Strongly correlated systems: open issues

Strongly correlated electronic materials

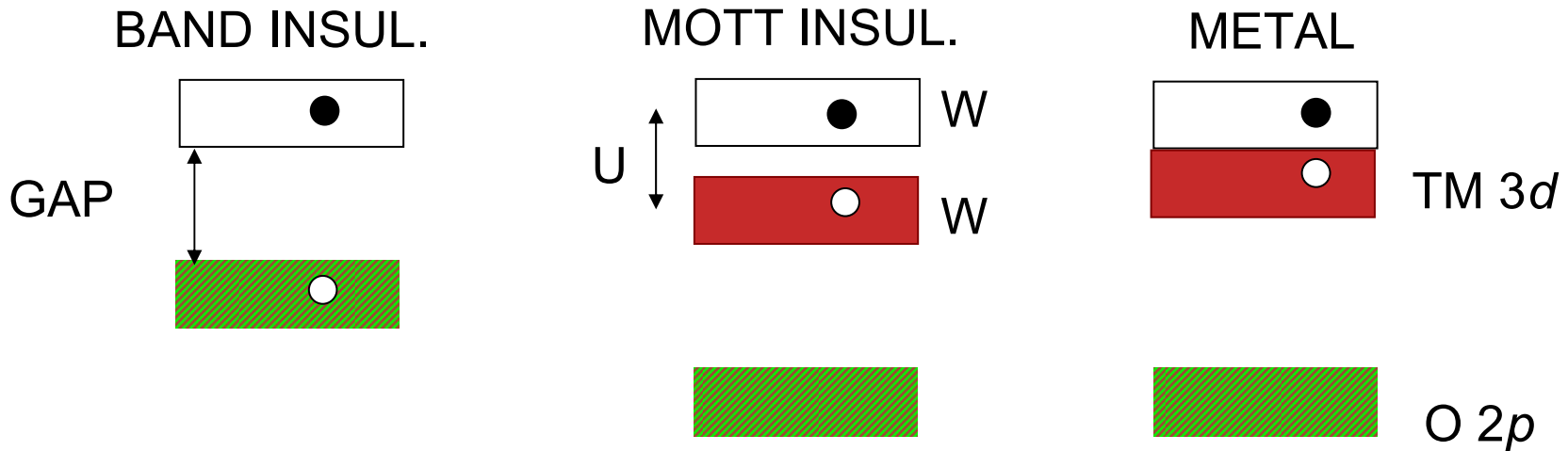
- Metal – insulator transitions
- High T_c superconductors

→ Open questions on electronic structure, accurate experimental data needed for electronic structure close to the Fermi level

- Photoemission (filled electronic states)
- X-ray absorption (empty electronic states), XAS
- X-ray scattering (low energy excitations across gap), IXS, RIXS
- XAS, IXS all photon methods, complementary to ARPES
- (R)IXS flux hungry technique – bulk sensitive, insulating samples
- with pump probe approach: different pathways for phase transitions
- fs scale: $T_{el} \neq T_{lattice}$, electron-phonon coupling...

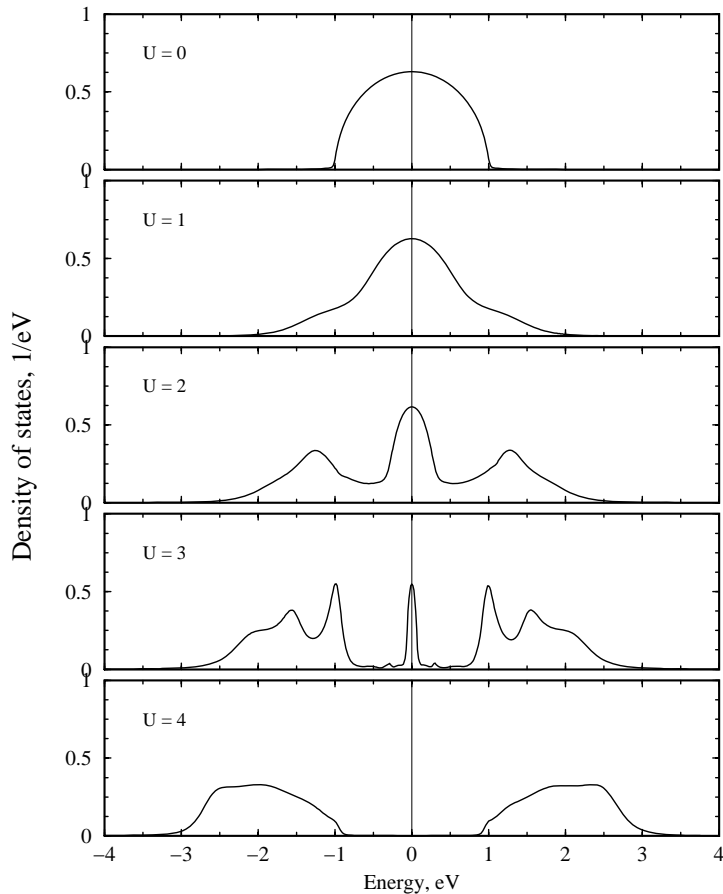
Mott transition

Metal insulator transition

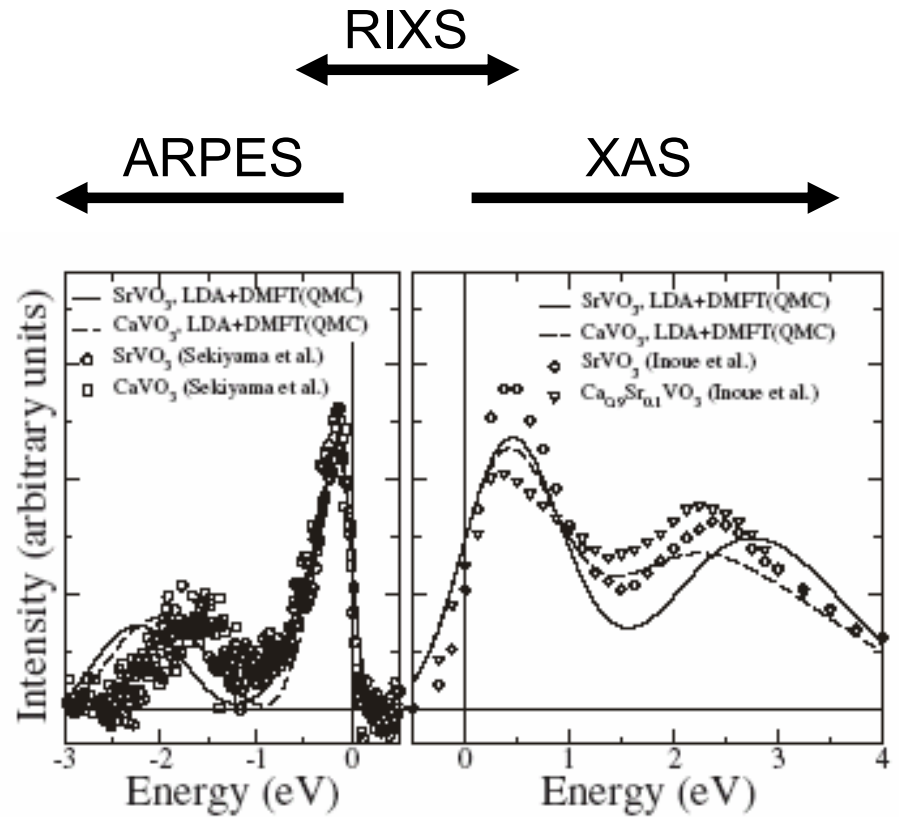


Upper and lower Hubbard band W
 $d-d$ electron Coulomb interaction U
 $\rightarrow \text{gap} \sim U - W$

spectroscopy \leftrightarrow DMFT



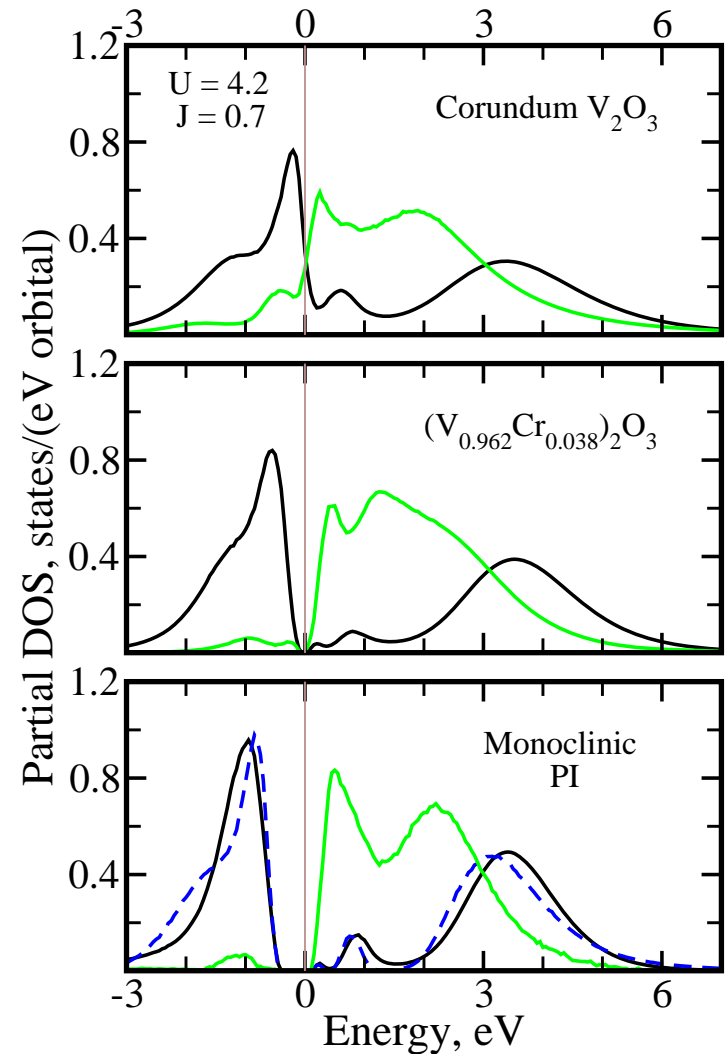
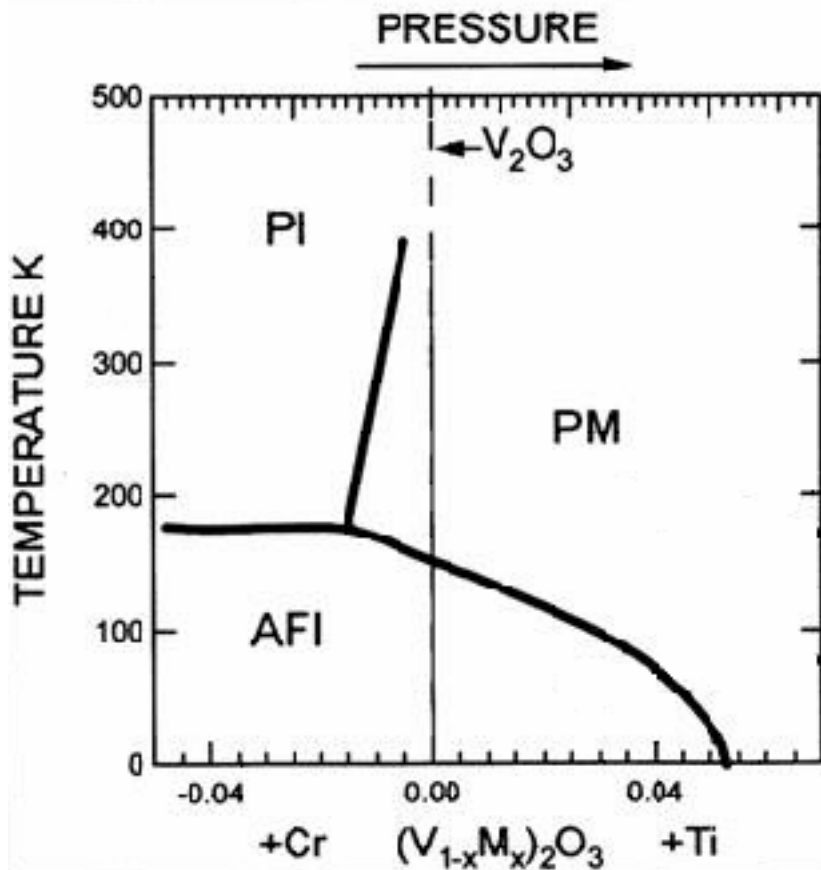
A. Georges et al. RMP (1996)



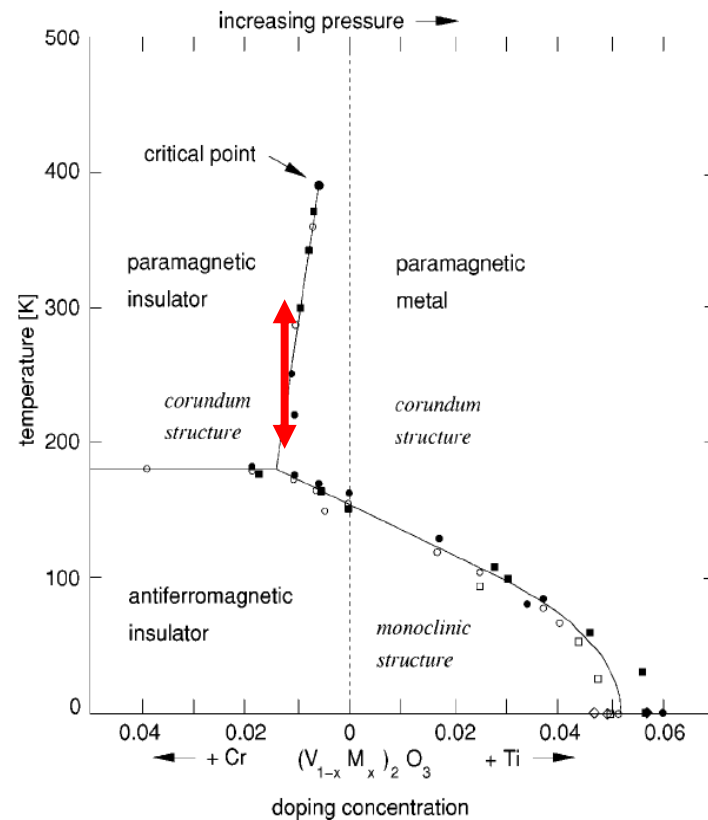
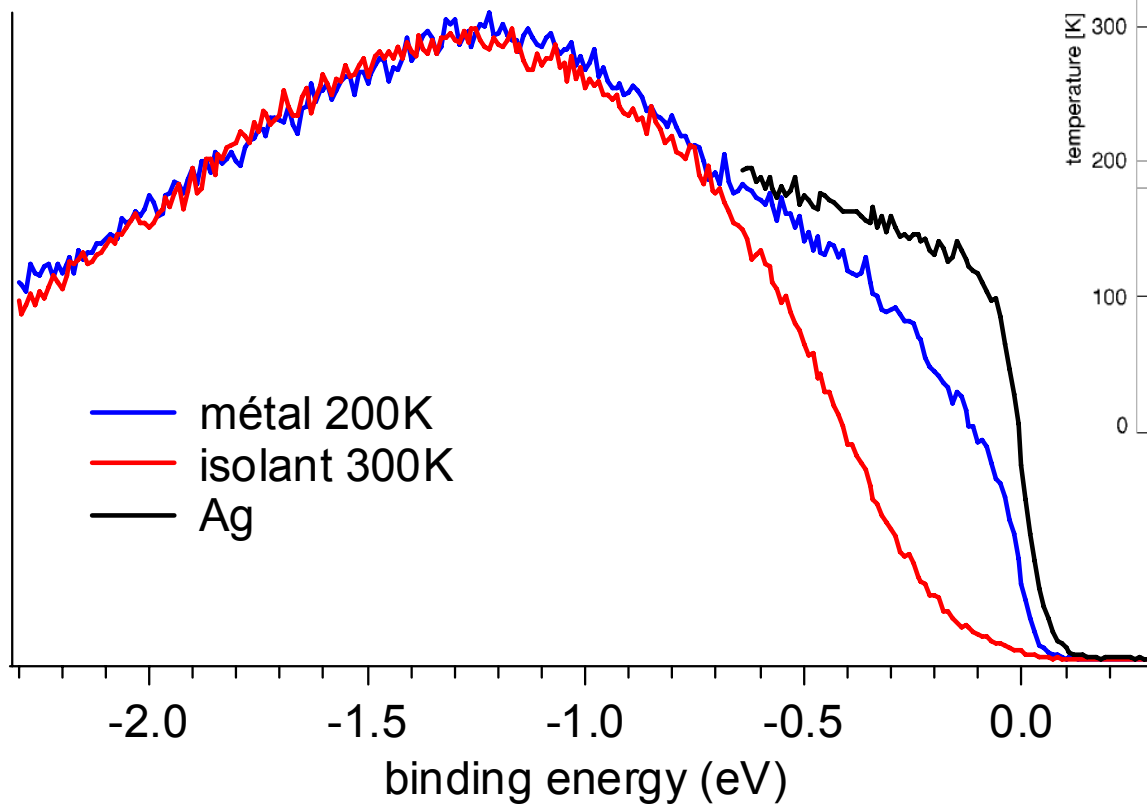
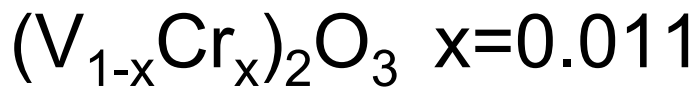
Vollhardt et al. JPSJ (2005)
LDA + DMFT

$V_2O_3 \rightleftharpoons$ prototype Mott transition

McWhan et al. (1973)



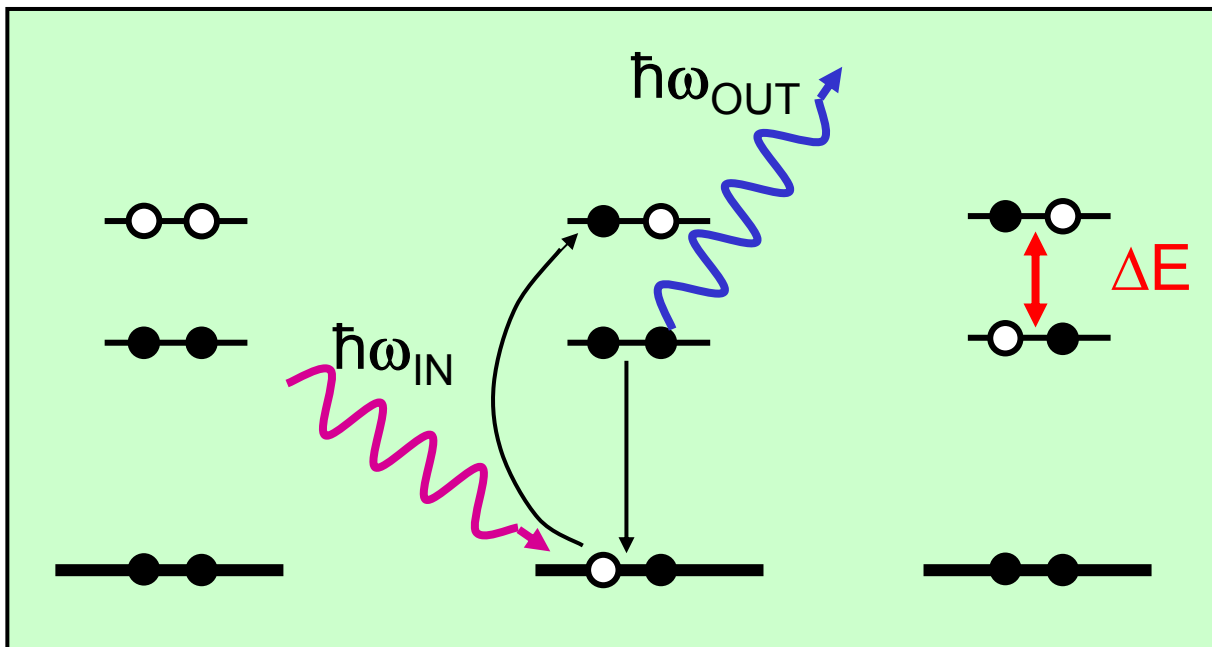
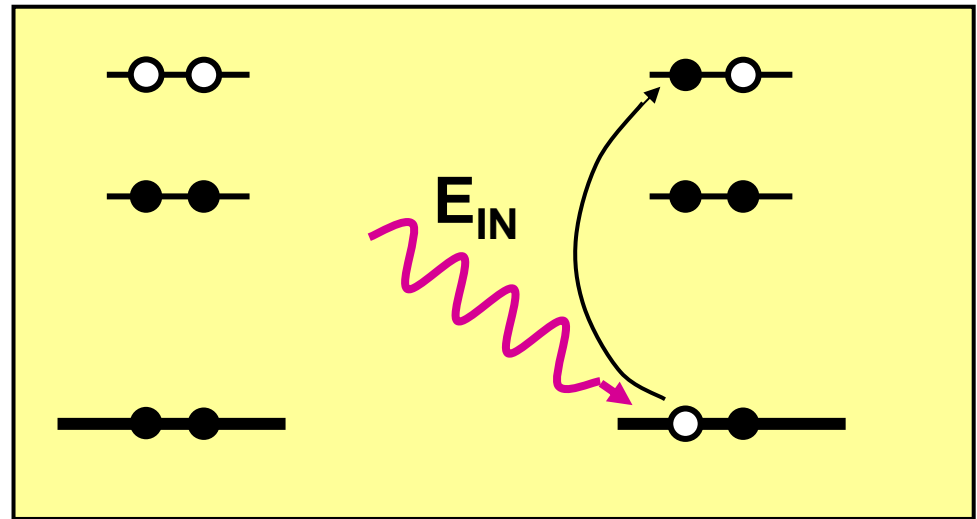
LDA+DMFT, Poteryaev *et al.*, (2006)



XAS and IXS: principle

X-ray Absorption Spectroscopy

→ empty states



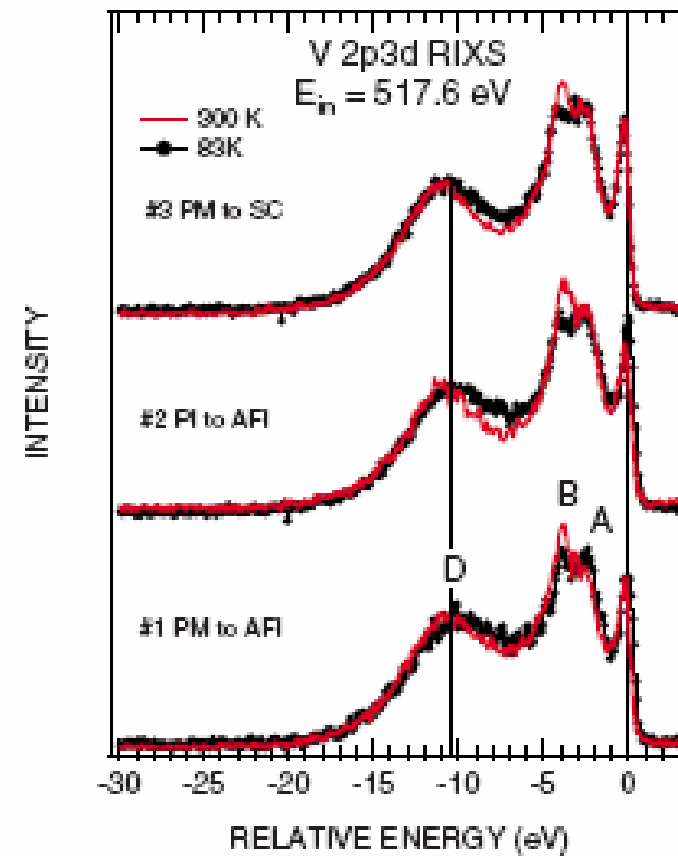
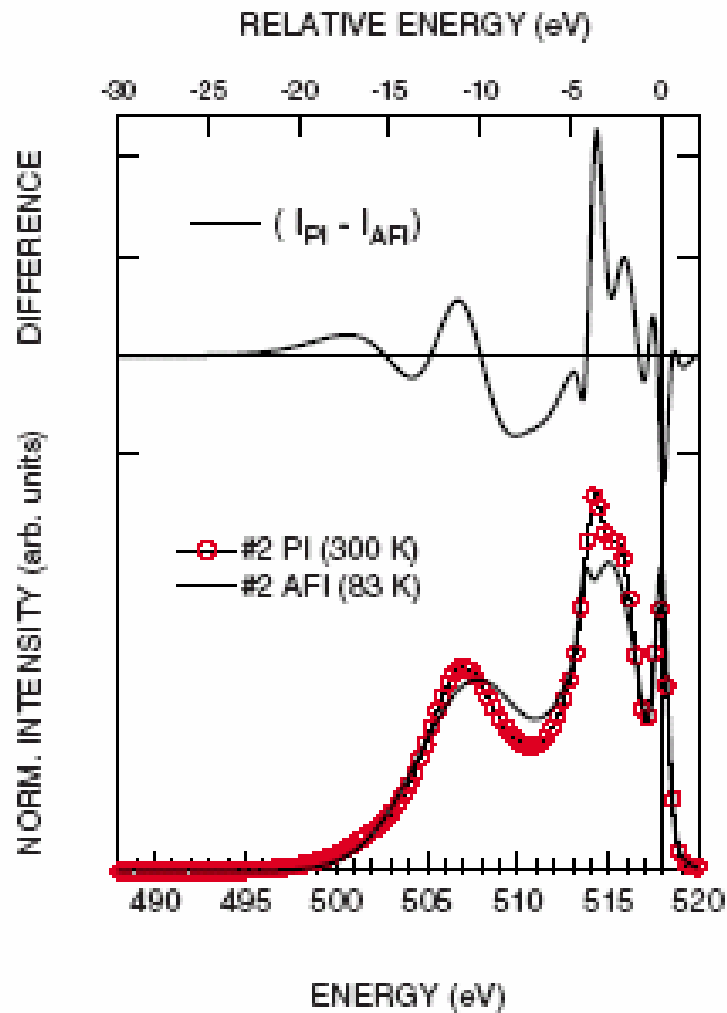
Inelastic X-ray Scattering

$$\Delta E = \hbar\omega_{IN} - \hbar\omega_{OUT}$$

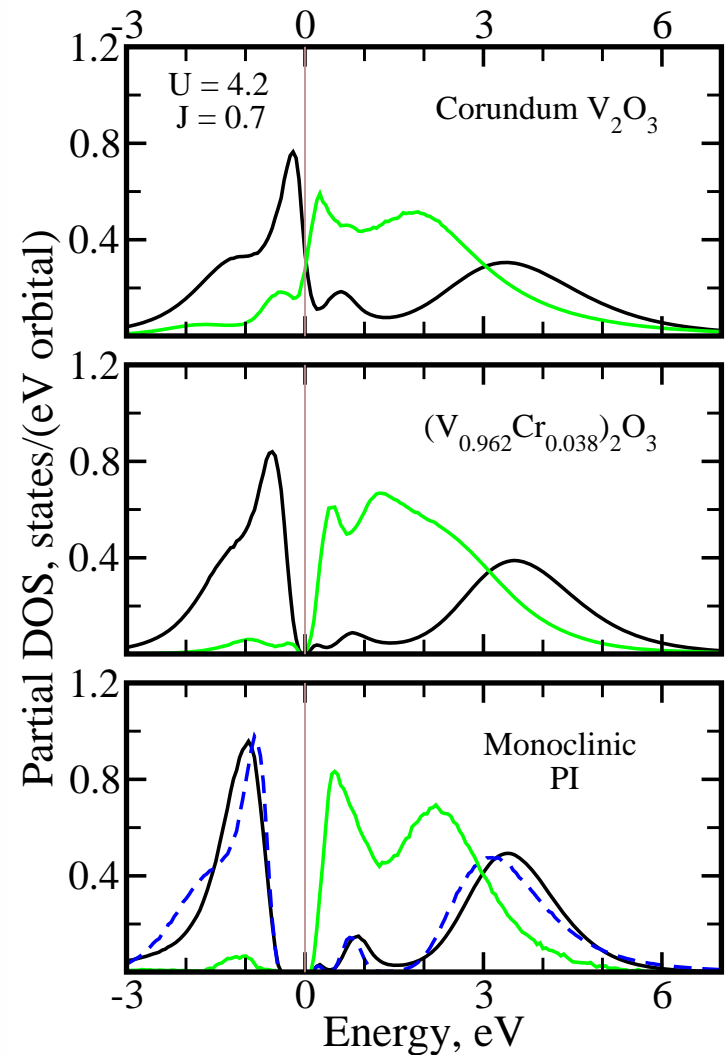
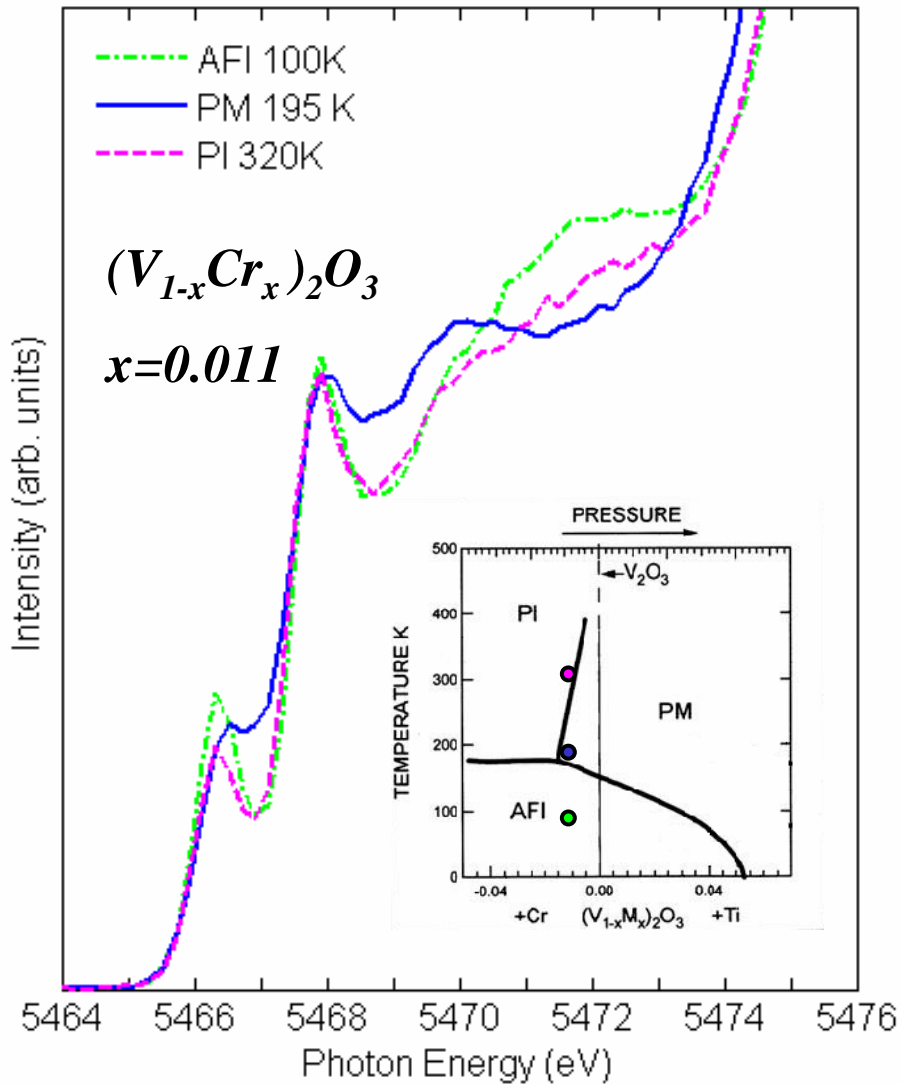
low energy excitations

→ **d-d** (dipole forbidden)

Stronger signal at
resonance (RIXS)

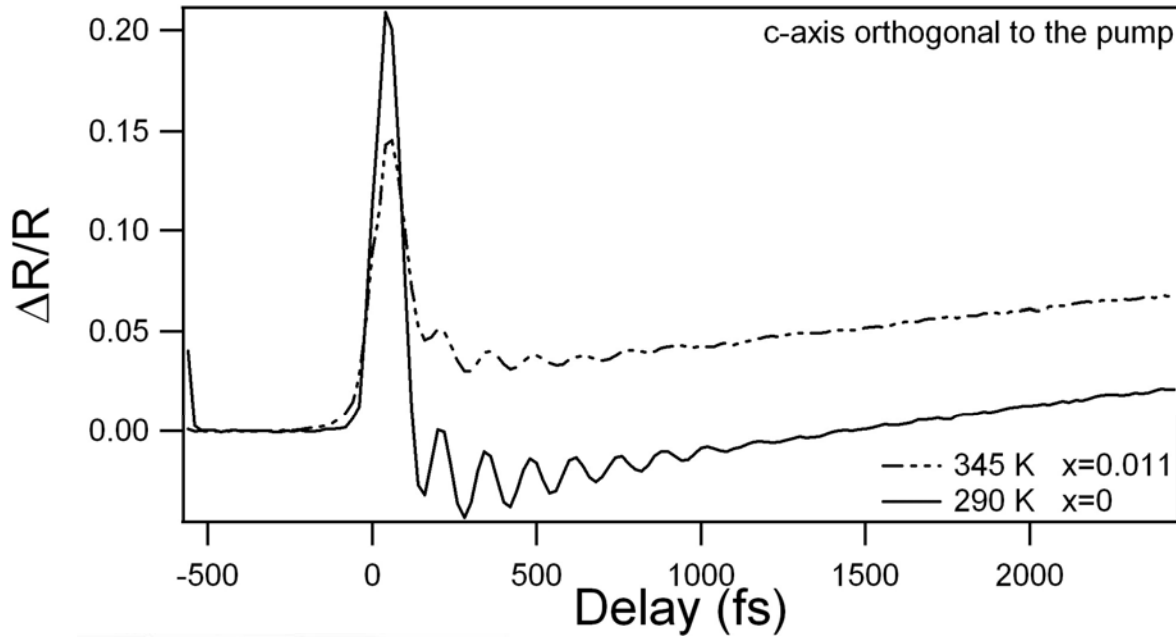


X-ray Absorption: V_2O_3



LDA + DMFT

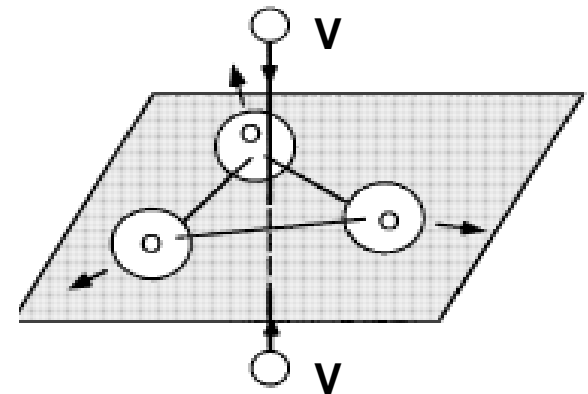
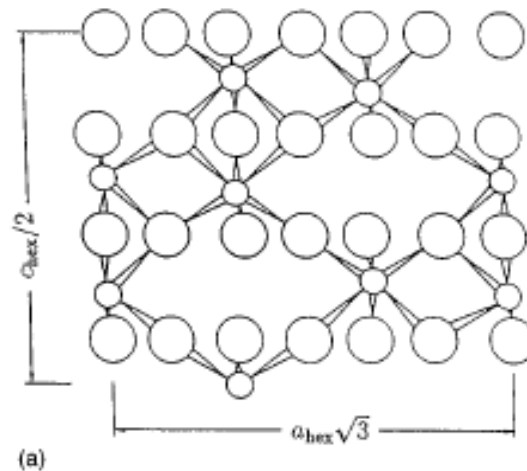
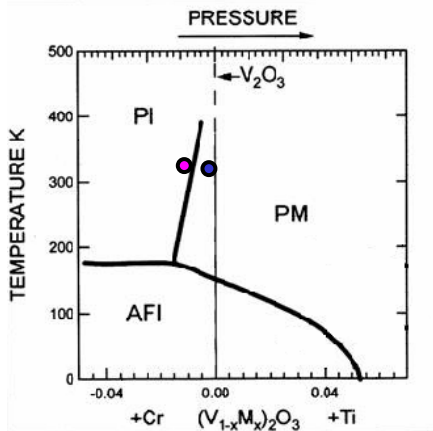
fs pump-probe IR reflectivity



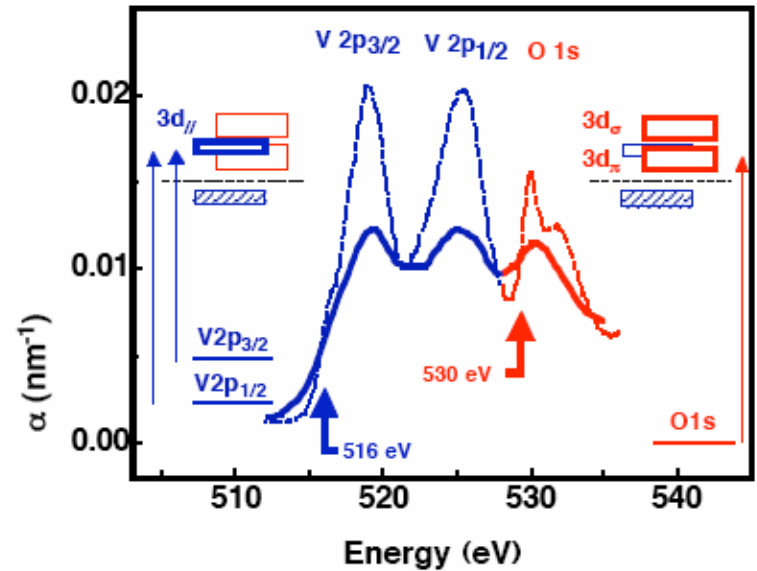
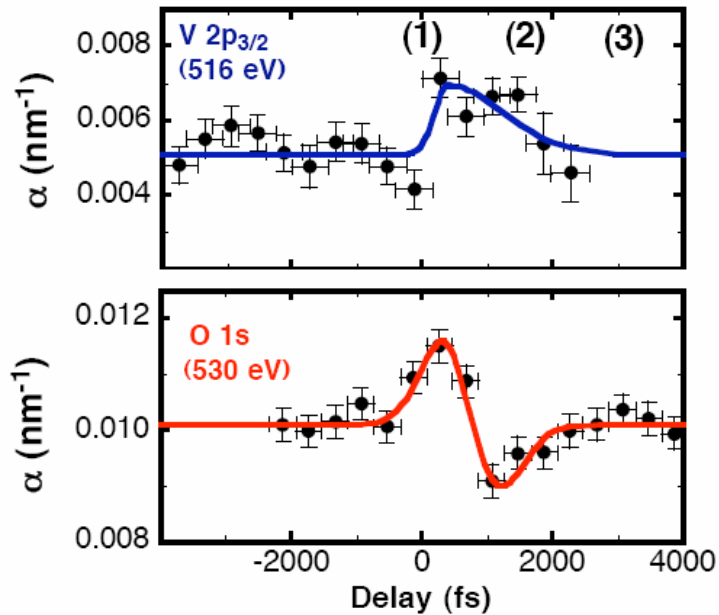
B. Mansart et al.,
in collaboration with
D. Boschetto (LOA)



A_{1g} mode

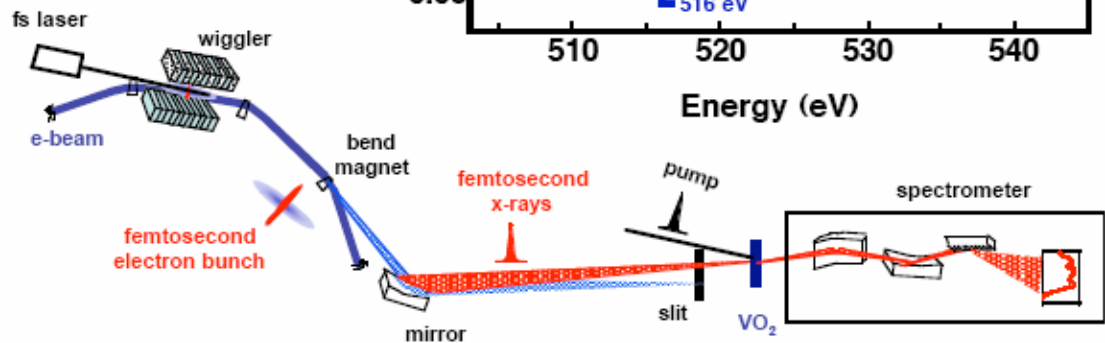


VO₂ ⇌ fs X-ray XAS



T < 320 K insulator
T > 320 K metal

(Mott vs Peierls?)



A. Cavalleri et al., PRL **95**, 067405 (2005)

Conclusions

XAS and (R)IXS \Leftrightarrow strongly correlated systems (transition metal oxides)

Complementary approach to ARPES for fermiology studies

Bulk sensitive, compatible with insulating samples

Pump-probe configuration \rightarrow phase transition dynamics (fs, e-ph coupling)

Applicable in different wavelength regions (optical pump, X-ray probe)

with sources 400-1000 eV (1-3 nm) \rightarrow L_{2,3} edges

High cross section 2p \rightarrow 3d transitions

with sources 4-10 keV (0.1 – 0.3 nm) \rightarrow K edges

transitions over the whole (P, T) phase diagram

High repetition rate (1-100 kHz) preferable, but feasible even with

low repetition rate (\sim 10 Hz) fs sources



Resonant X-Ray Raman Spectra of Cu dd Excitations in $\text{Sr}_2\text{CuO}_2\text{Cl}_2$

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(Received 20 November 1997)

We present resonant x-ray Raman scattering results on $\text{Sr}_2\text{CuO}_2\text{Cl}_2$, a model compound for high- T_c superconductors. We demonstrate that the dd excitations can be observed and show that the polarization dependence can be used to identify the dd excitations. We find the transition from the $d_{x^2-y^2}$ ground state to the d_{xy} excited state at 1.35 eV and to the degenerate d_{xz} and d_{yz} excited states at 1.7 eV. From analysis of the polarization dependence we conclude that the $d_{3z^2-r^2}$ orbital energy is at 1.5 eV and not in the midinfrared (0.5 eV) as recently suggested. We use recent theoretical arguments to show that the $d_{3z^2-r^2}$ excitation is accompanied by a local spin flip resulting in a shift upwards of 0.2 eV due to the exchange interaction with the neighboring spins. [S0031-9007(98)06273-5]

RIXS on M edge

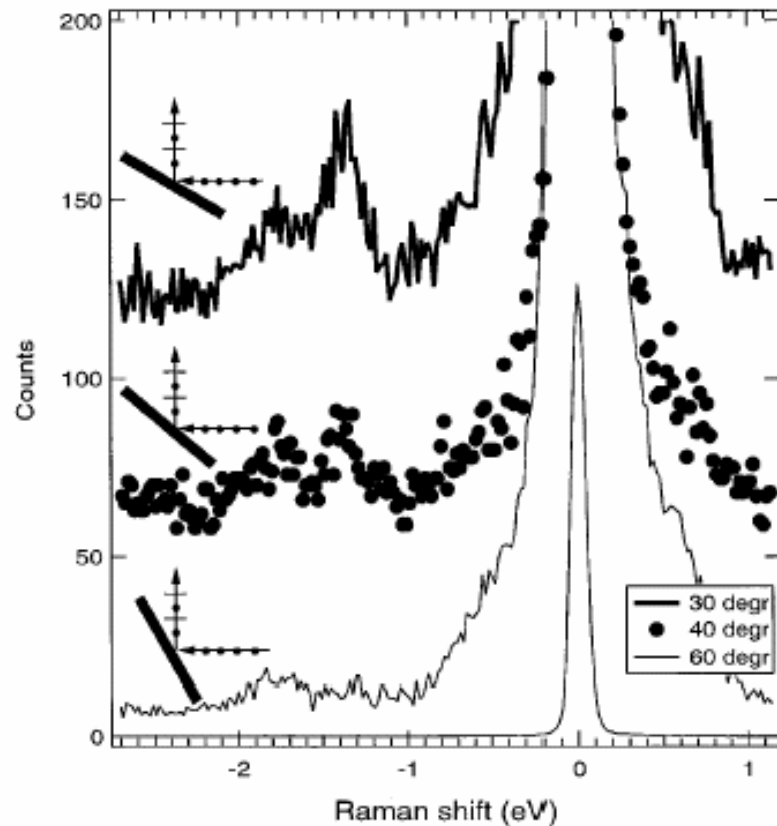


FIG. 1. Polarization-dependent x-ray resonant Raman spectra at the Cu M_3 resonance (74 eV). The angle between the emission direction and the sample normal is 30°, 40°, and 60°, from top to bottom. The last spectrum is also shown reduced by a factor of 200.

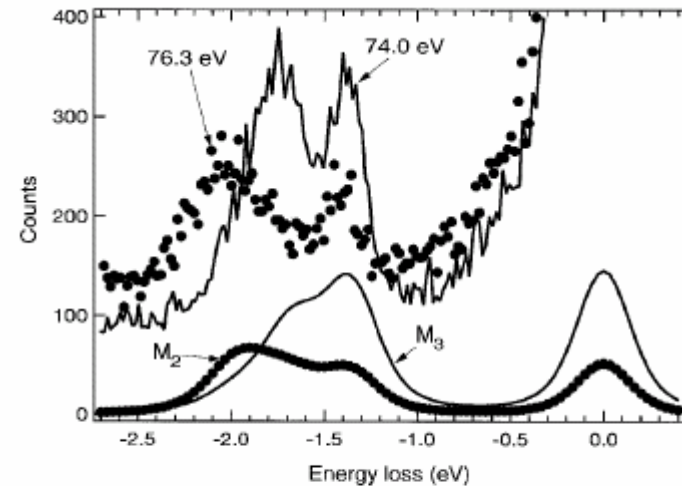


FIG. 3. X-ray resonant Raman spectra at the Cu M_3 and M_2 resonances (74 and 76.3 eV). The emission direction is in the direction of the incident polarization and makes an angle of 40° with the sample normal.

Pressure induced transitions

