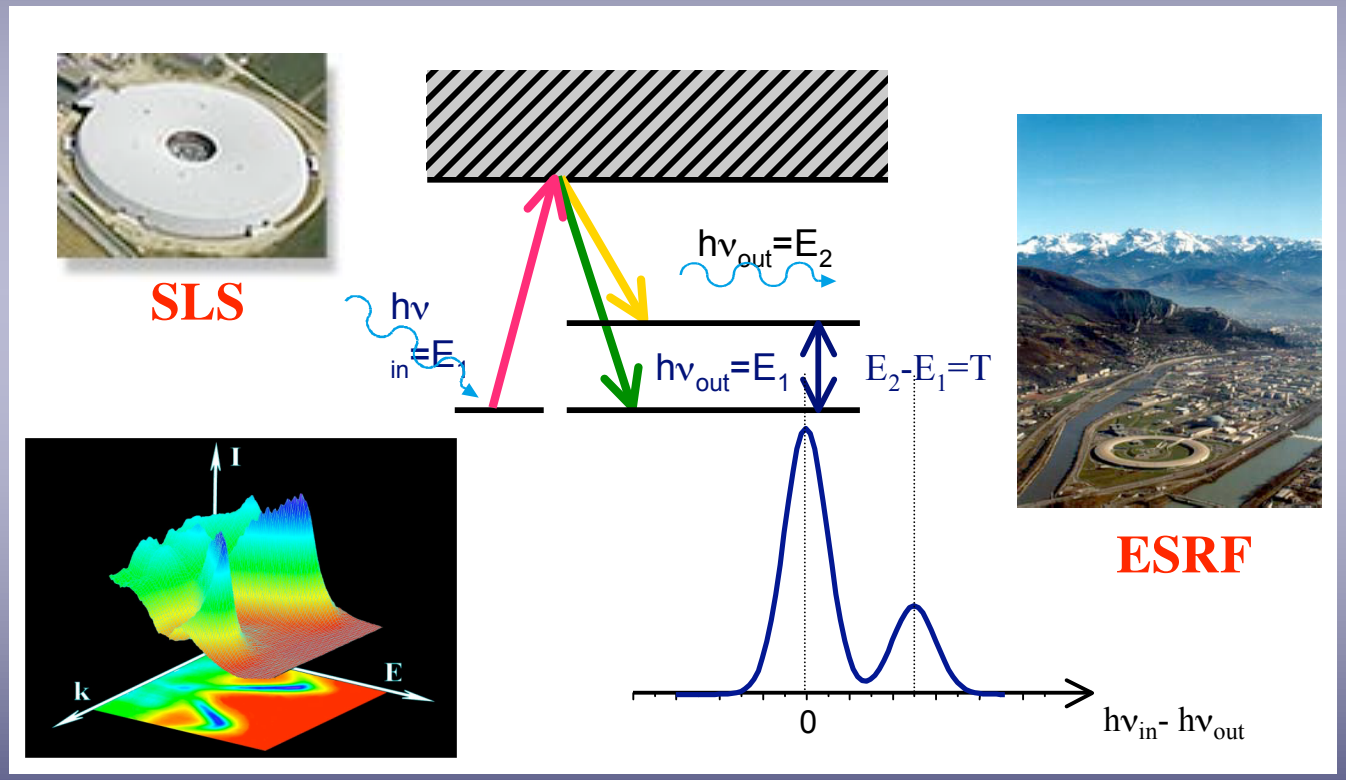
BULK INFORMATION FROM HIGH- (AND HIGHER) ENERGY SPECTROSCOPIES

Marco Grioni



ÉCOLE POLYTECHNIQUE FÉDÉRALE DE LAUSANNE

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MaNEP SWITZERLAND

Acknowledgments

L. Perfetti
D. Pacile
M. Papagno
H. Berger — EPFL

L. Braicovich
C. Dallera
G. Ghiringhelli — Politecnico Milano

G. Panaccione — Trieste

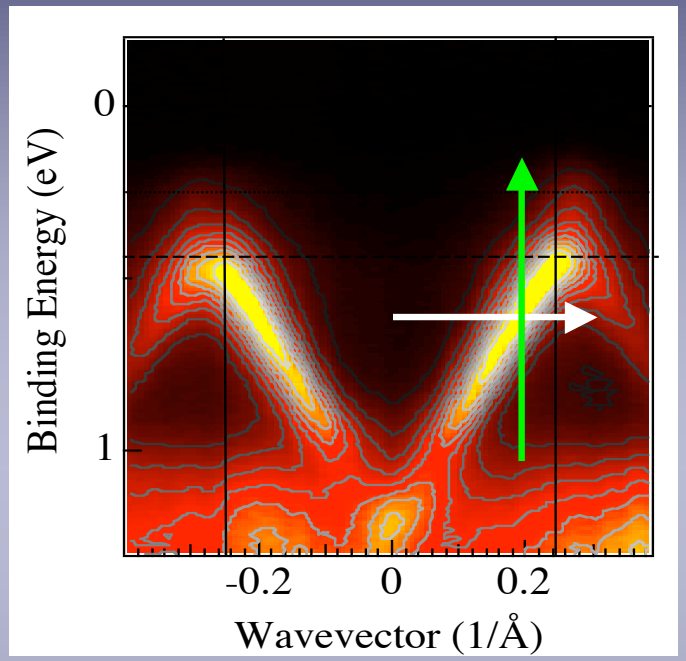
G. Monaco — ESRF

Outline:

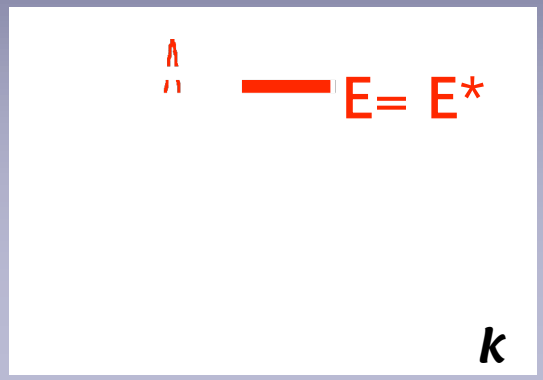
- ARPES: what we *would like* to do: a case study
- Bulk sensitivity : soft x-rays
- *Real* bulk sensitivity : photons and hard x-rays

ARPES: E, *k* selectivity

ARPES
intensity map



MDC (k, E*)



$\Delta k = 1/l$

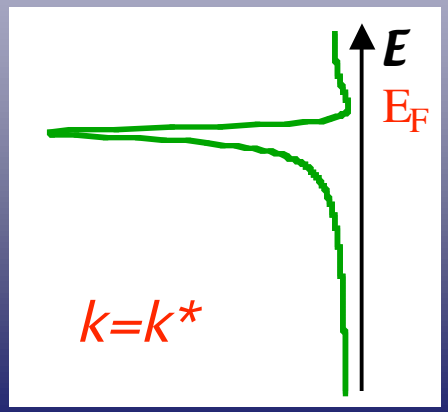
l = QP coherence length

Probe the nature of the quasiparticle

EDC (k^*, E)

$\Delta E \sim 1/\tau$

τ = QP lifetime

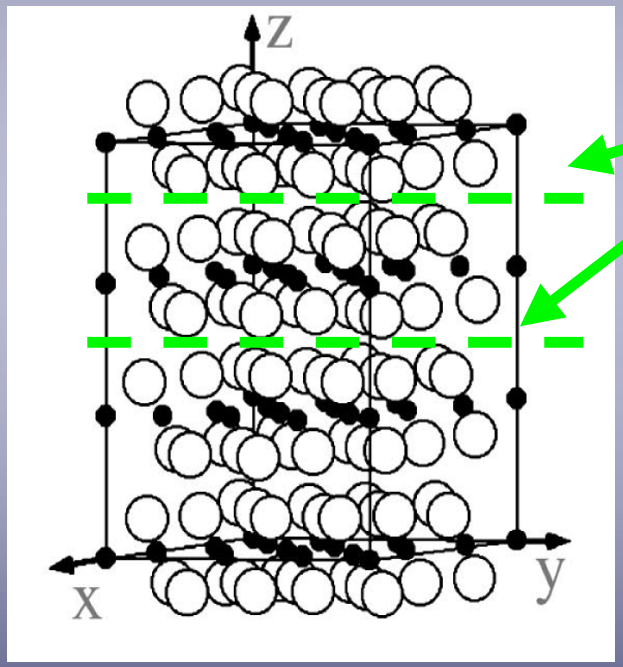


What we want to do with ARPES

Case study - a 2D metal-insulator transition: TaS_2

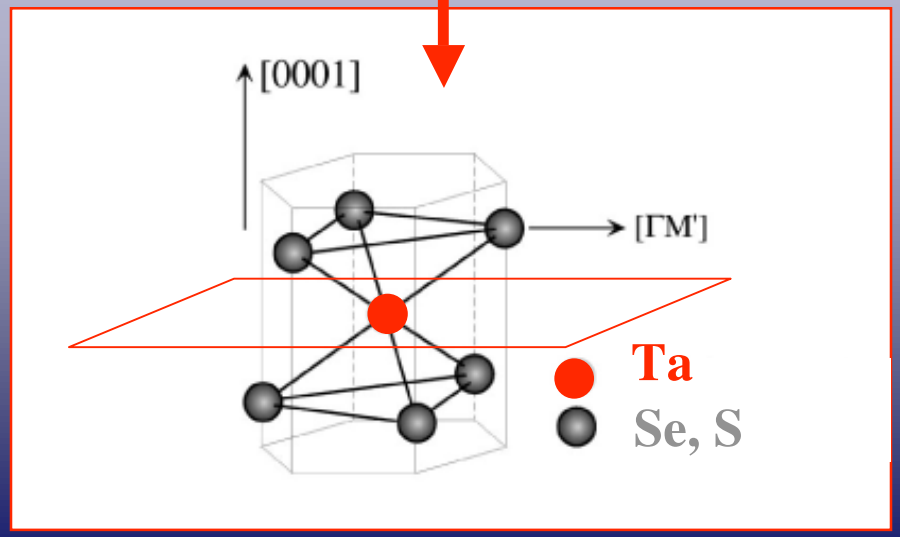
Layered TM dichalcogenides: "model" *quasi*-2D metals

1T-TaX_2 (X = S, Se)

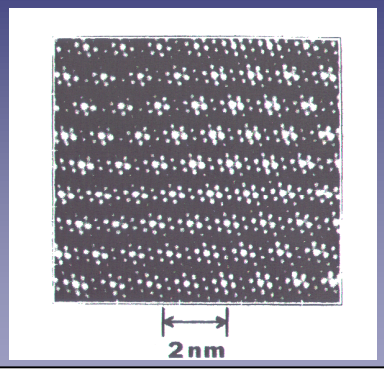


van der Waals gap

2D physics in Ta layer

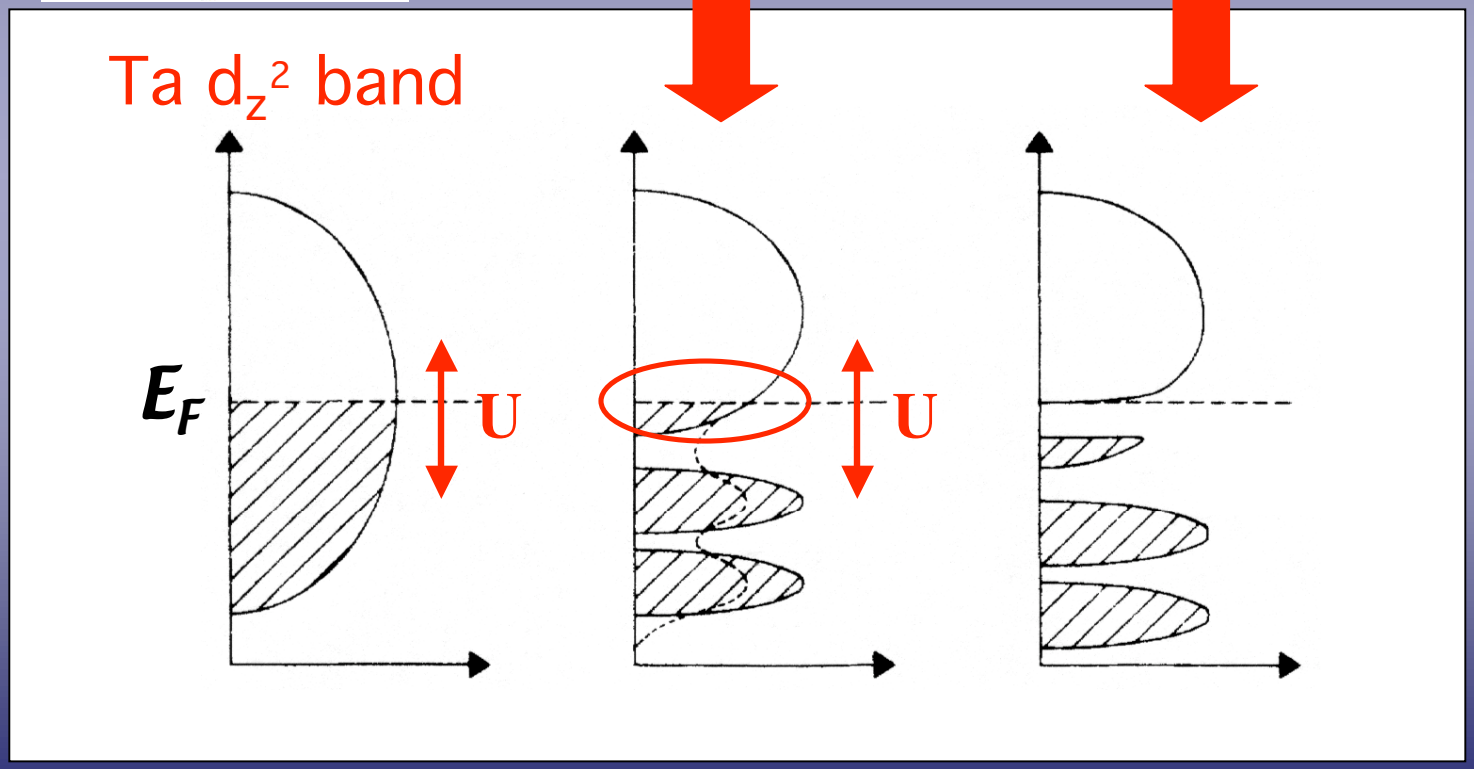


CDW + correlations → MI transition



CDW potential

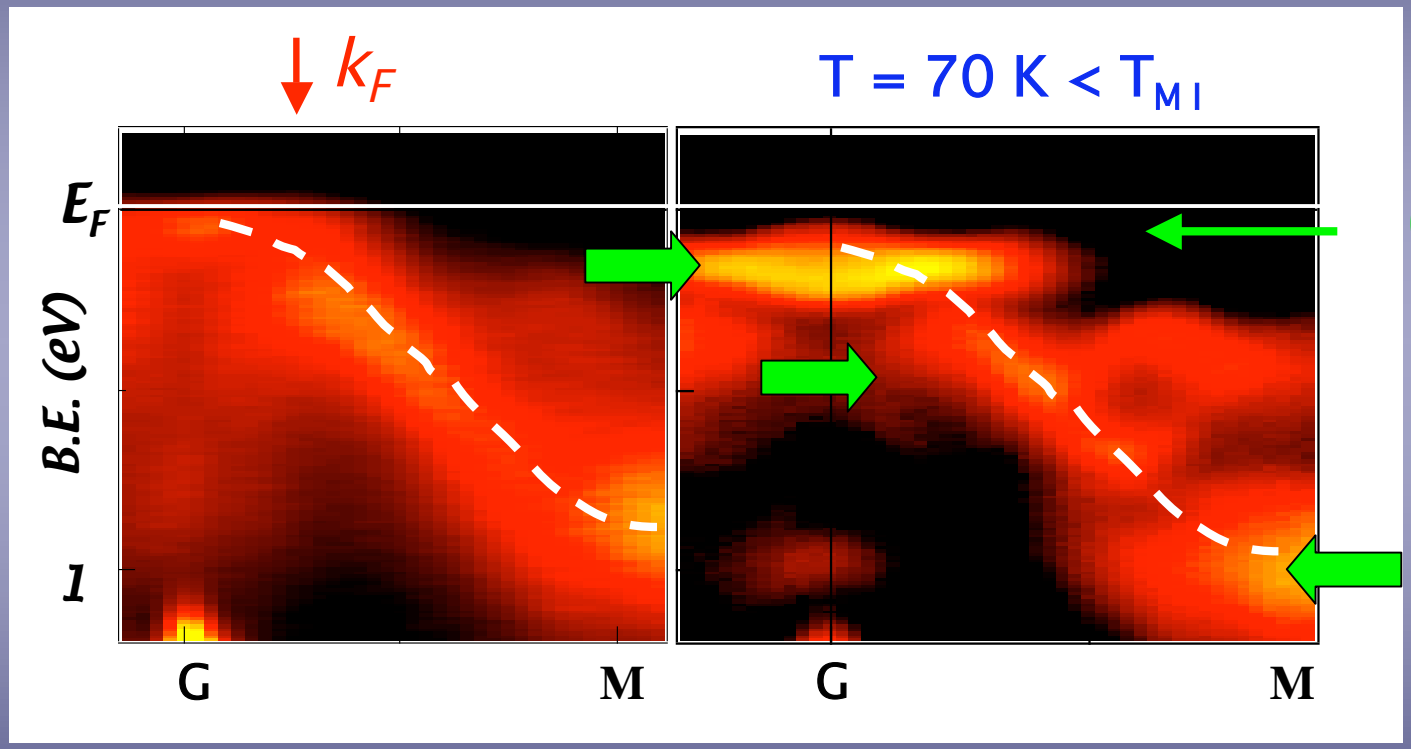
Electronic correlations



Tosatti & Fazekas

1T-TaS_2 - CDW manifolds and Mott gap

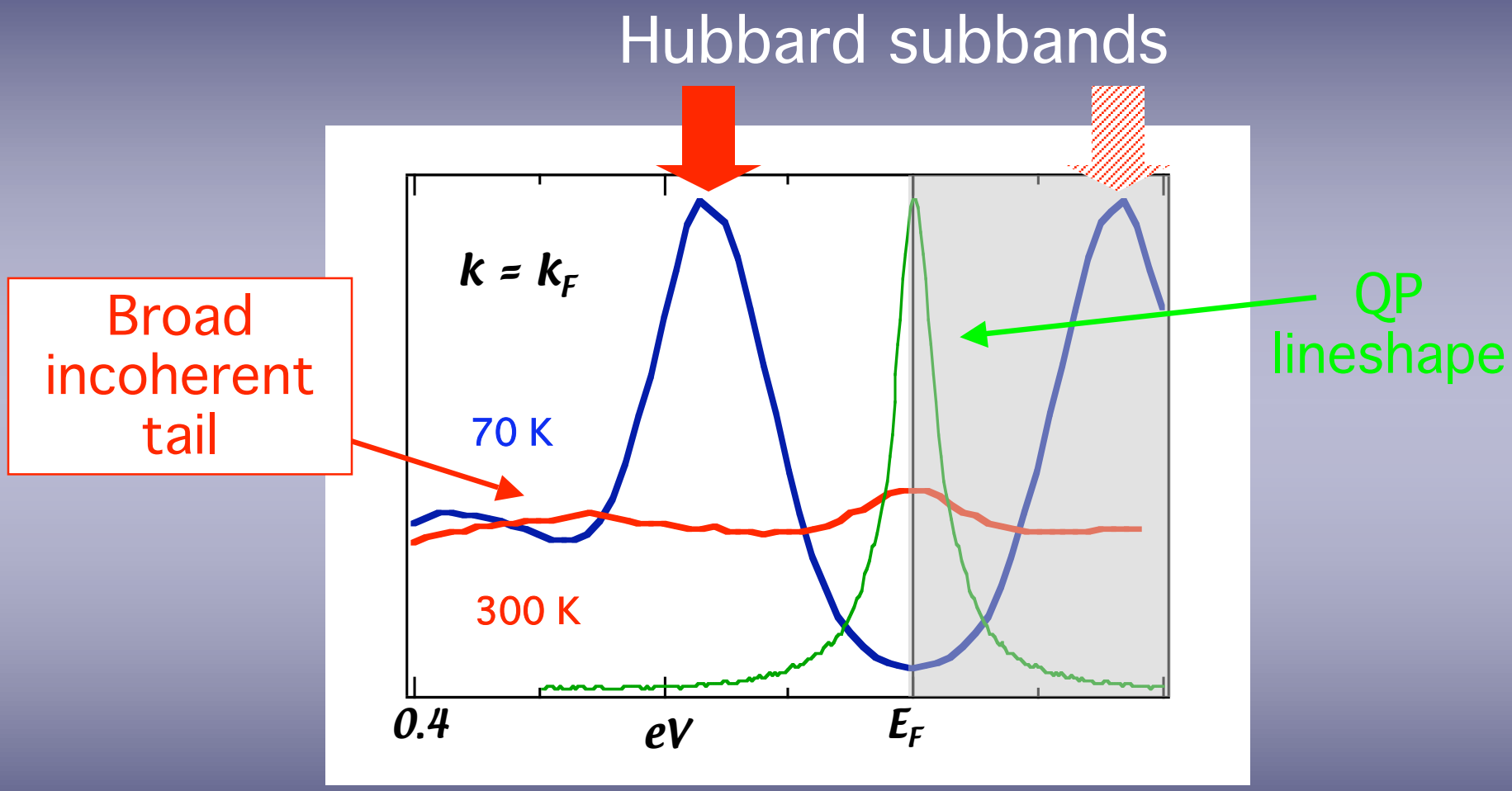
ARPES intensity map



metal insulator

Spectral weight distribution follows "main" band

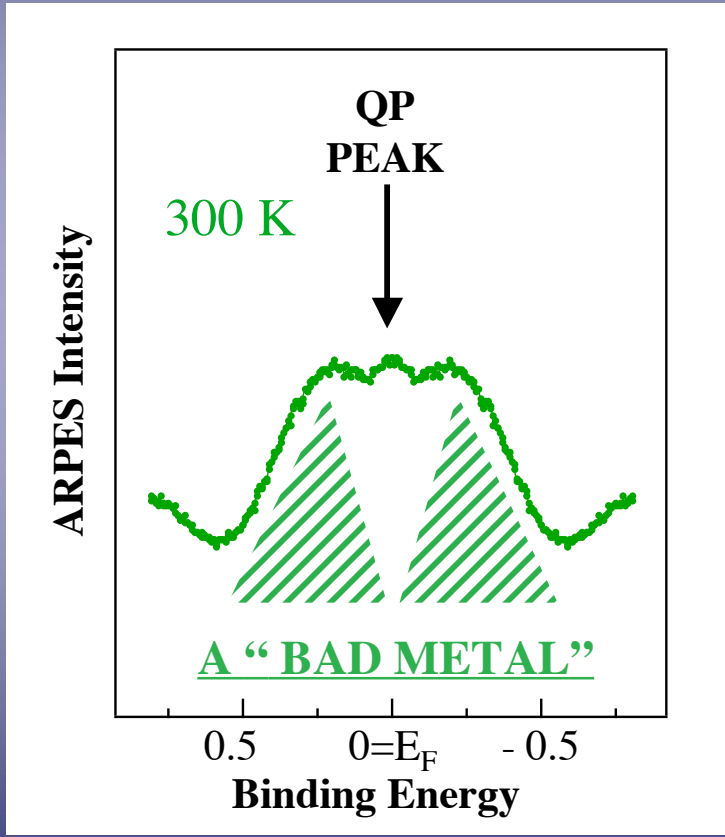
1T-TaS_2 : the Mott transition



after symmetrization around E_F

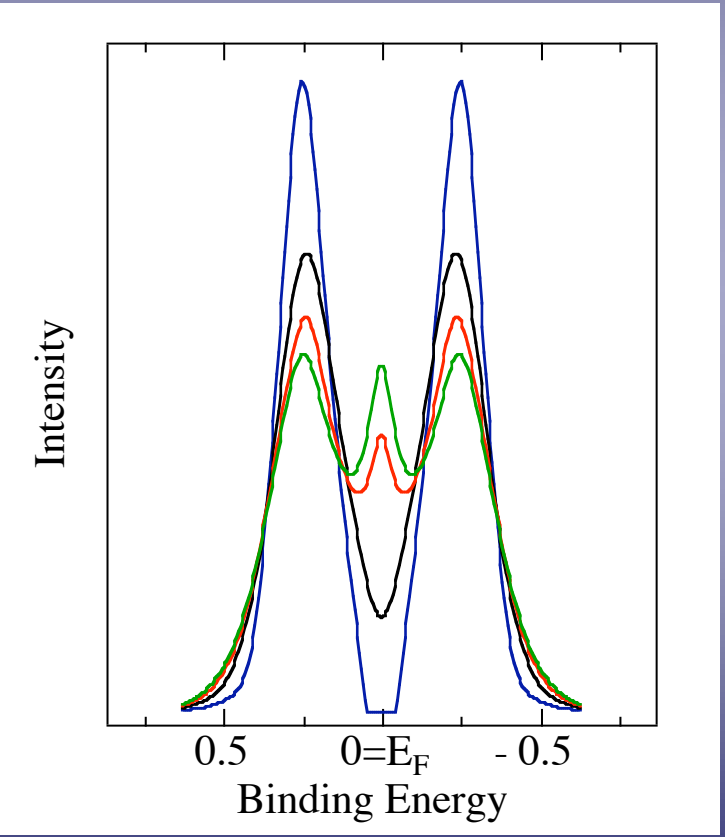
1T-$TaSe_2$: from bad metal to insulator

The disappearing quasiparticle



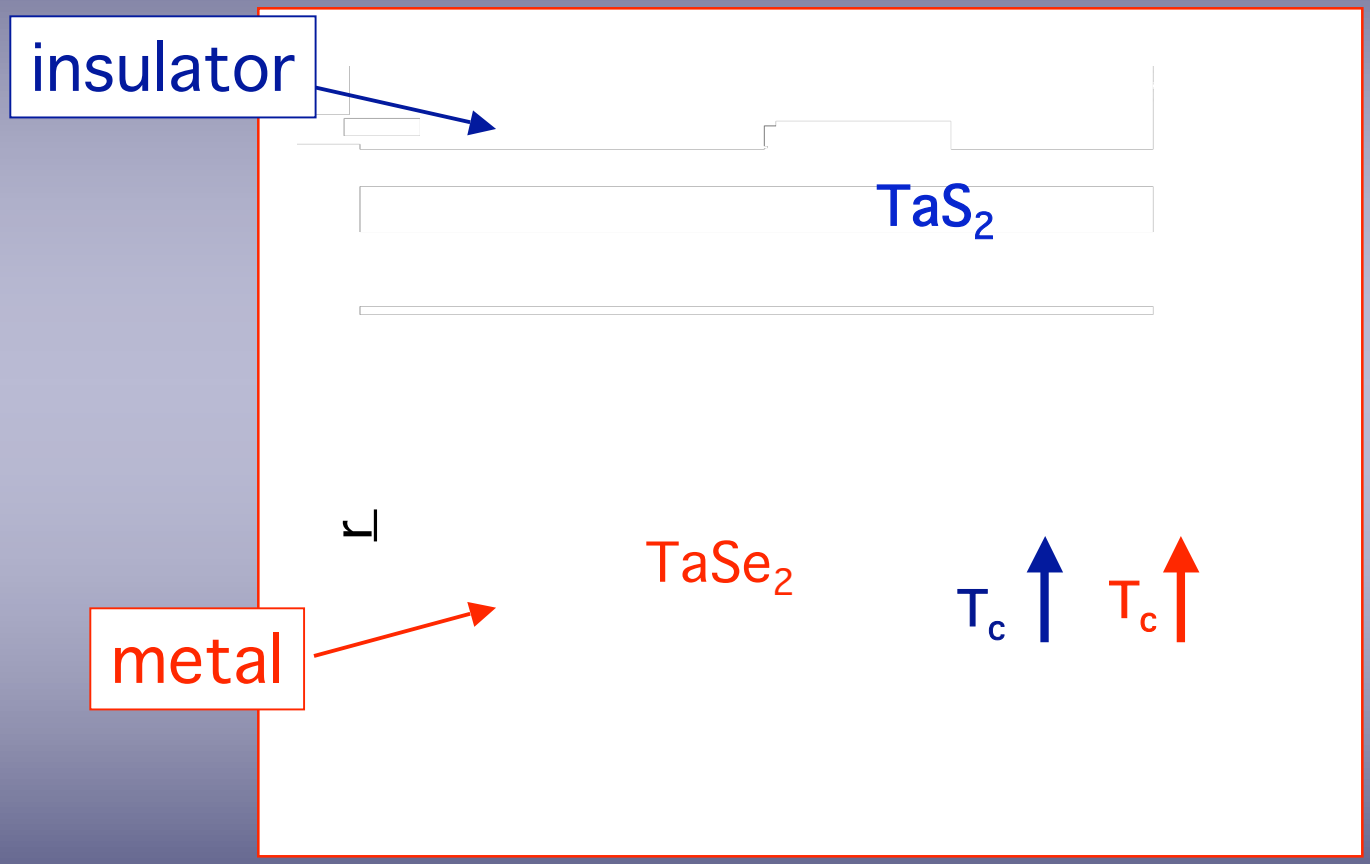
at $k = k_F$

DMFT calculation for the 1/2-filled Hubbard model
(A. Georges & coworkers)



L. Perfetti *et al.*, PRL (2003)

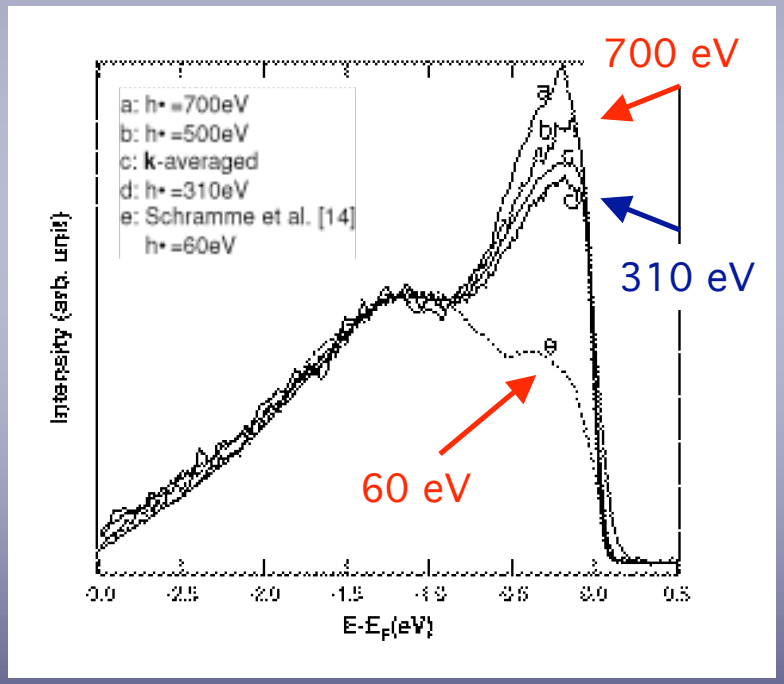
1T-TaX_2 (X = S, Se)



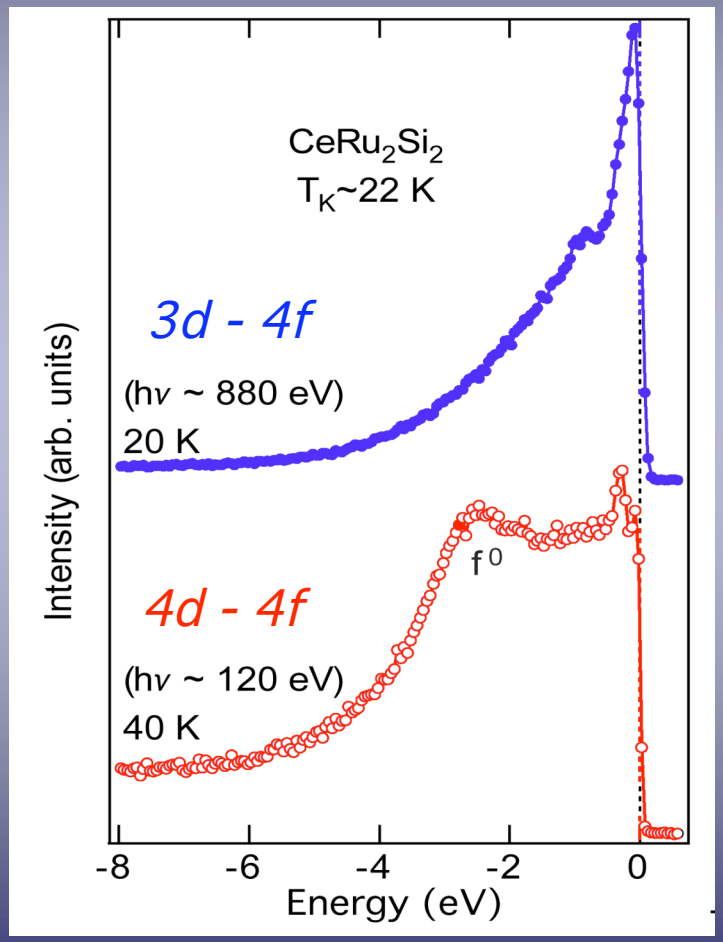
Only surface transition in $TaSe_2$!!

Surface and bulk are often quite different

V_2O_3



Mo et al., PRL (2003)

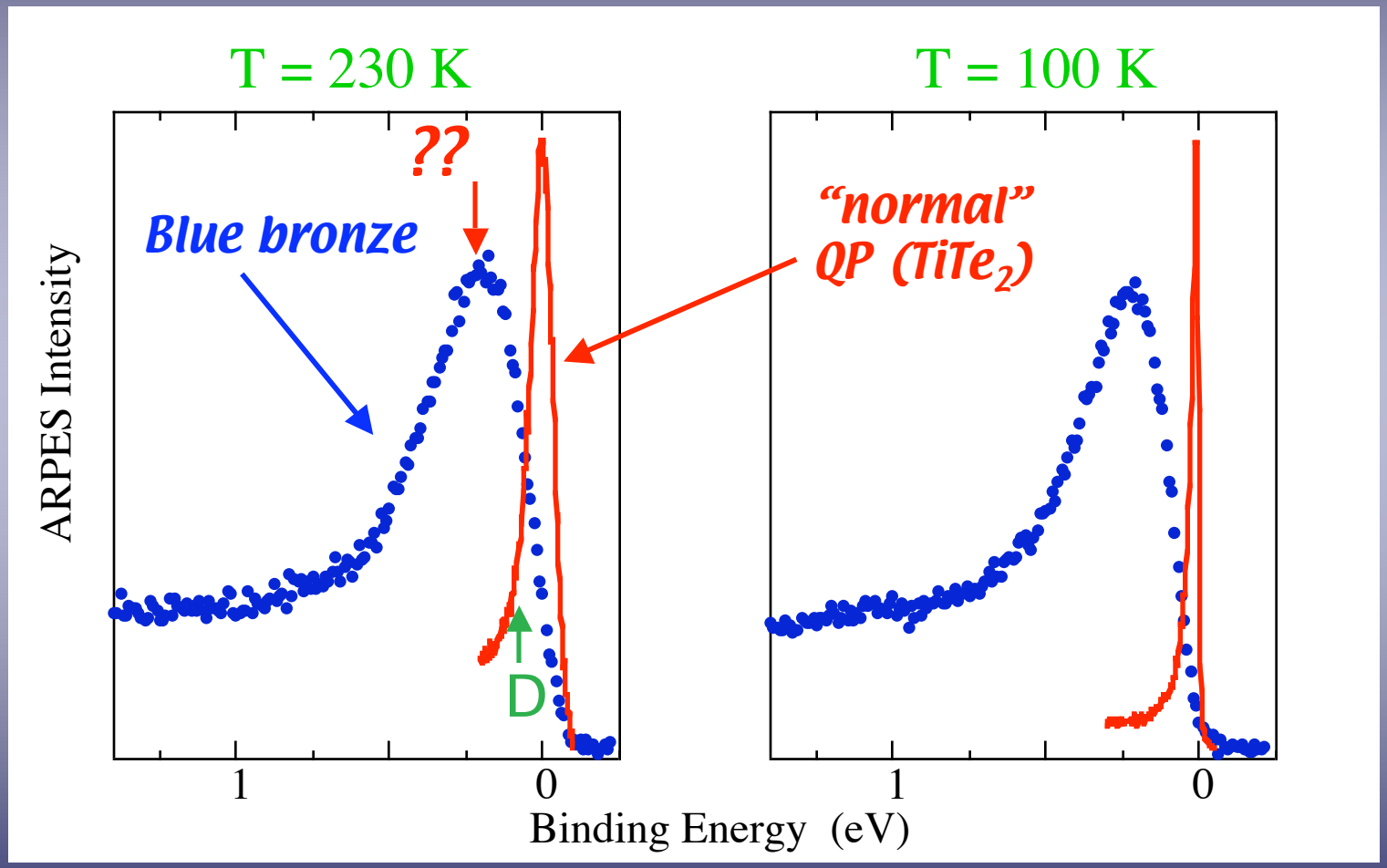


880 eV
bulk

120 eV
surface

Sekiyama et al., Nature (2000)

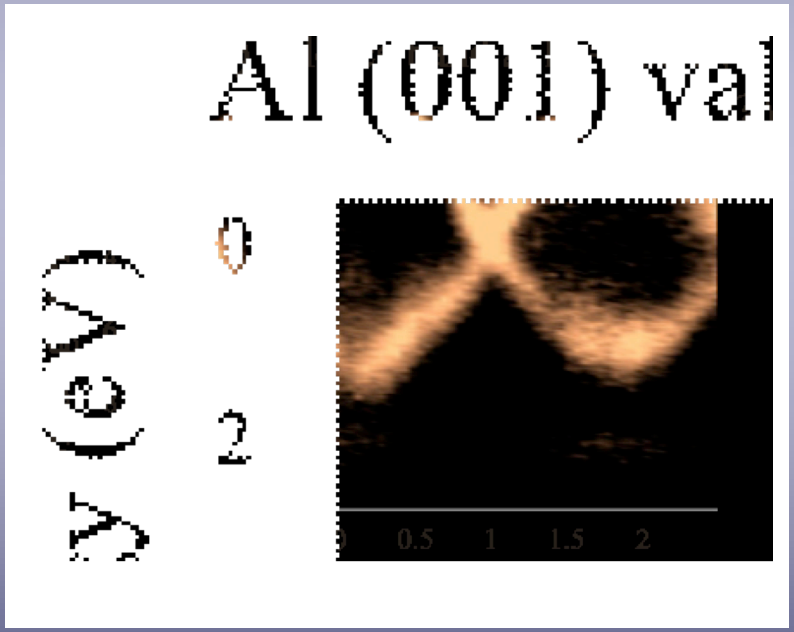
Non-Fermi liquid ARPES lineshapes in 1D: $k = k_F$

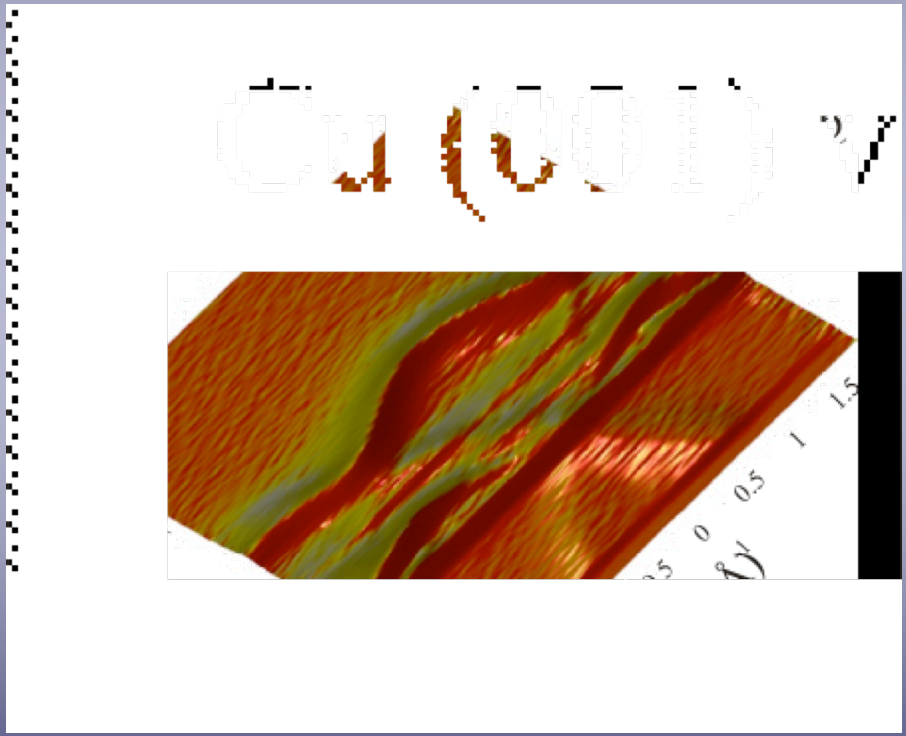


Is this a truly intrinsic feature ?

The european reference: ID8 at ESRF

Band mapping with soft x-rays





Data: O. Tjernberg, C. Dallera, N.B. Brookes

Open questions: technical, vertical transitions, phonons...

The ADRESS project: advanced soft x-ray spectroscopies

The Swiss Light Source

L. Patthey, V. Strocov

J. Mesot
PSI - Zurich

M. Grioni
EPFL

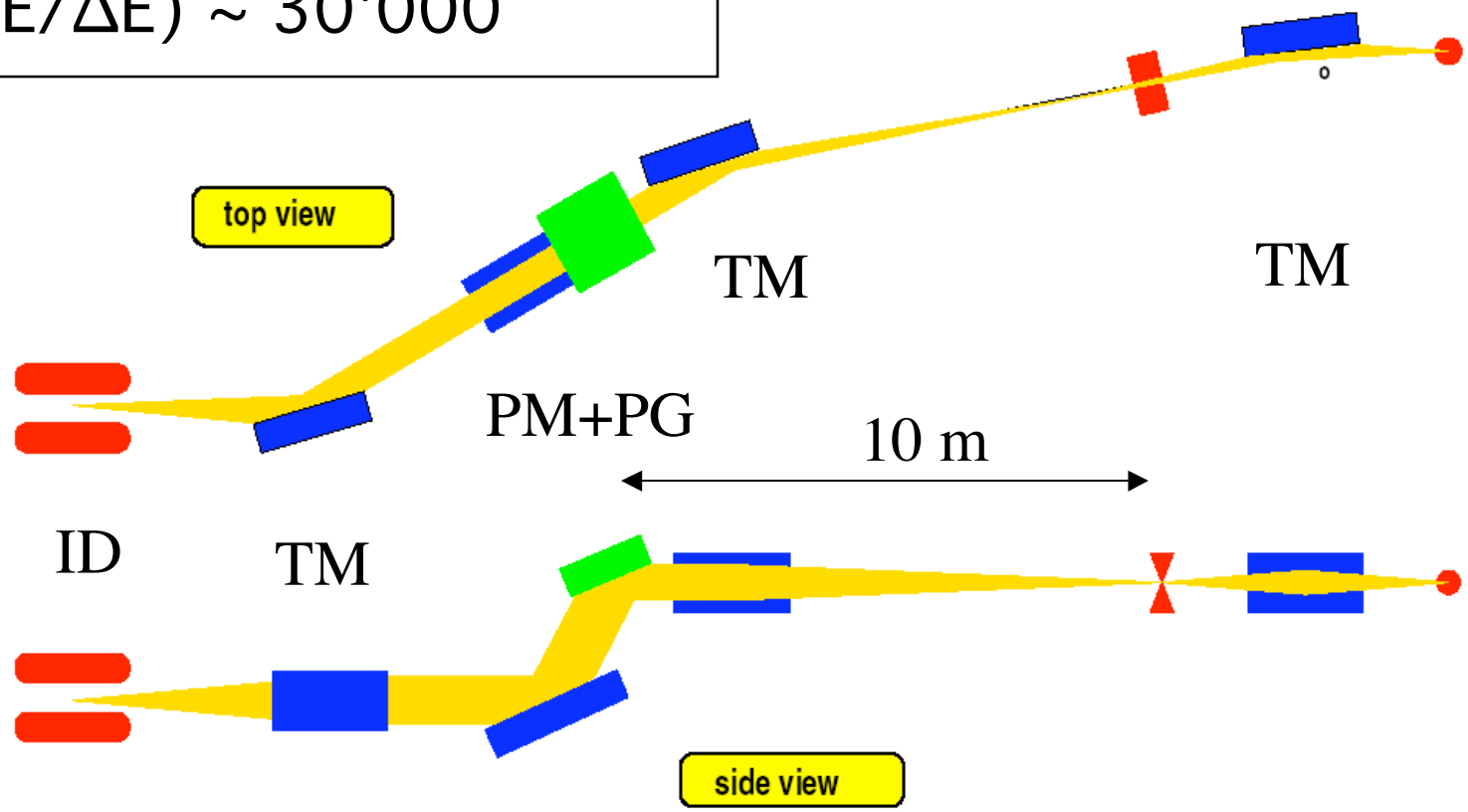
L. Braicovich
C. Dallera
G. Ghiringhelli

Milan

Input and suggestions from J.C. Campuzano
+ crucial support from the “SC” community

The ADRESS beamline:

Range: 400 - 1'500 eV
(E/ΔE) ~ 30'000

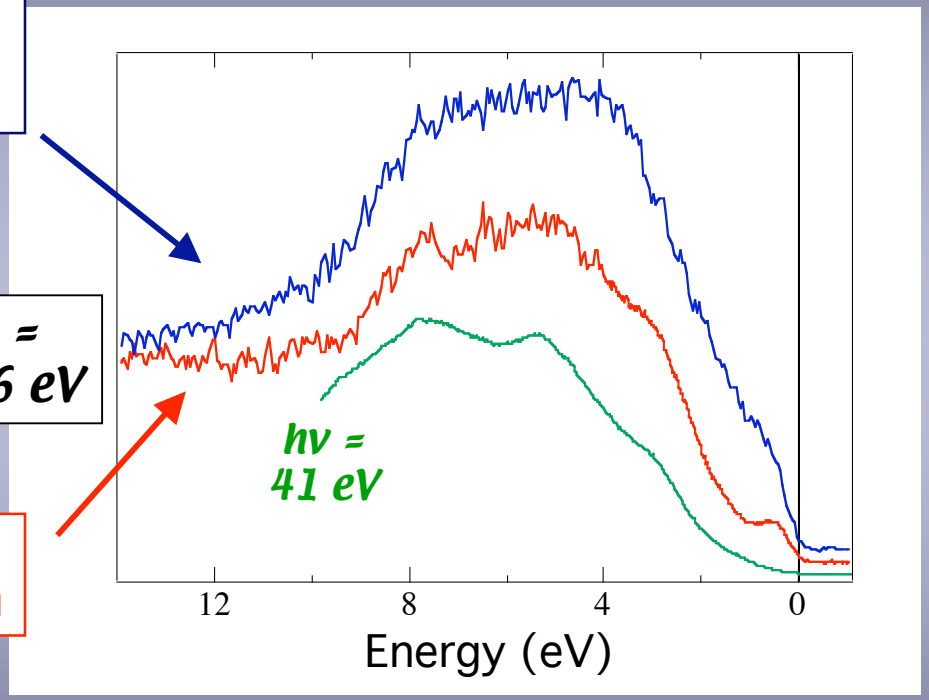


Bulk sensitivity: Fe_3O_4 (111)

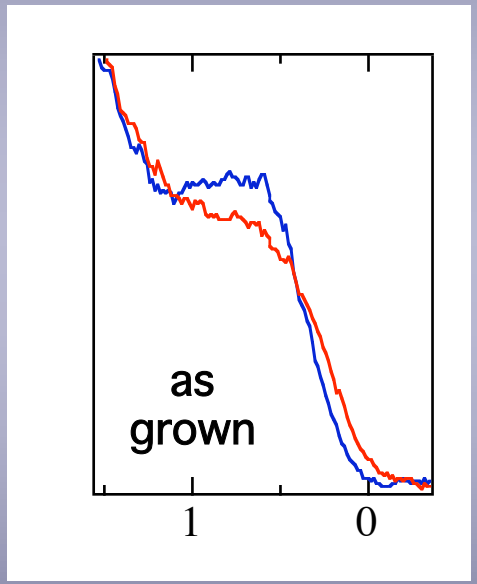
"gently" sputtered

hν = 1486 eV

As grown



Verwey transition



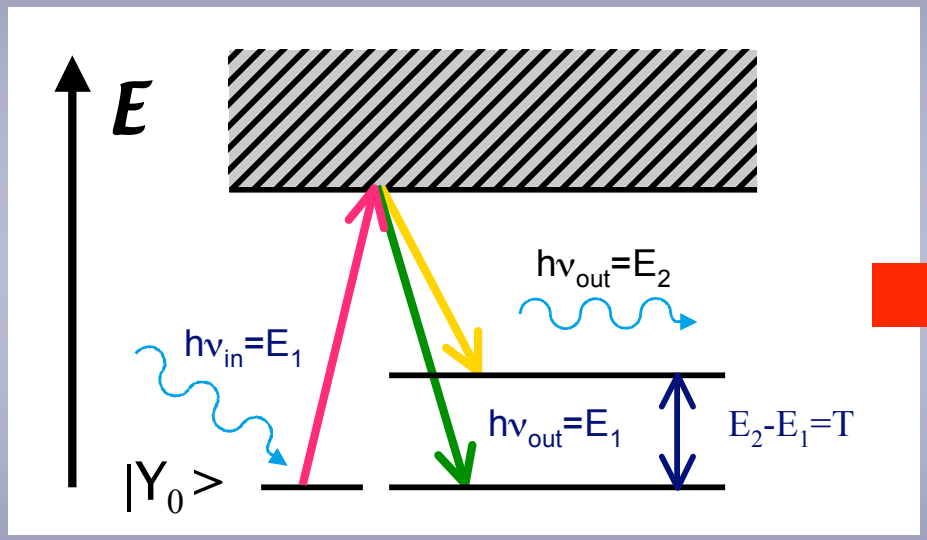
Message :

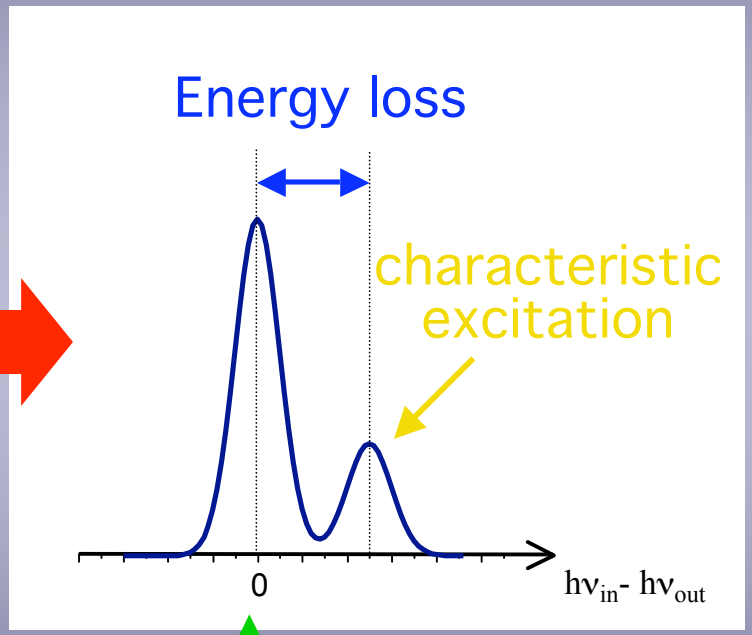
PES at ~ 1 keV *somewhat* bulk sensitive

How can we probe deeper ?

1. Use photons

RXES: '*Resonant* Raman' with x-rays





resonant absorption

coherent 2nd order optical process

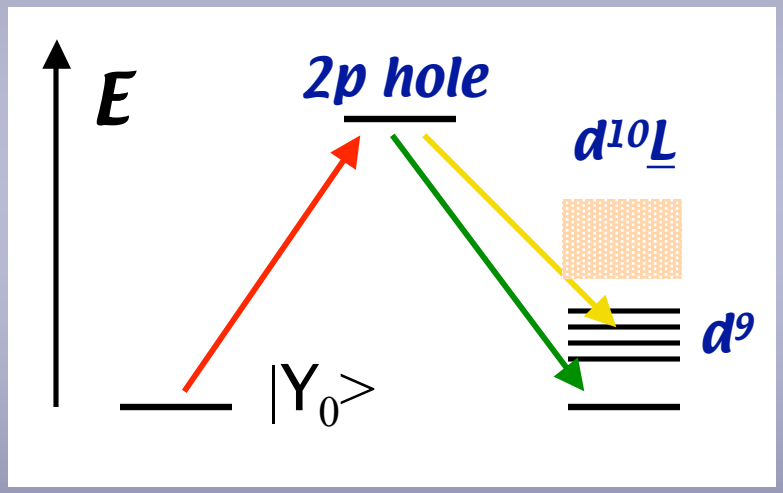
emission

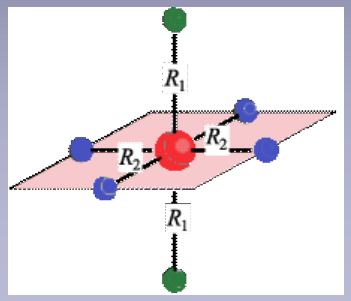
Elastic peak

‘*d-d*’ excitations in the cuprates

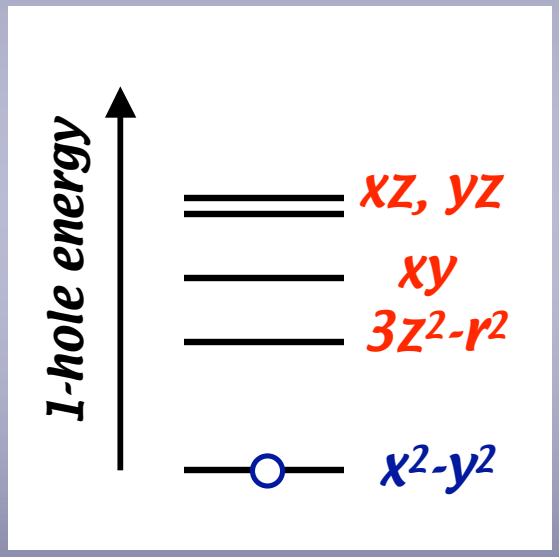
Cu 2p edge (2p → 3d; hn = 930 eV)

Cu L_3 RXES



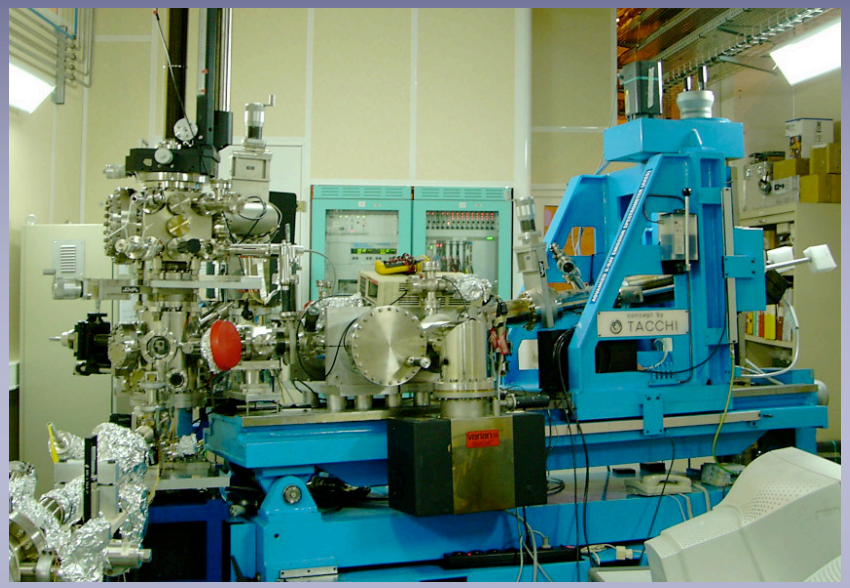


hole excitations



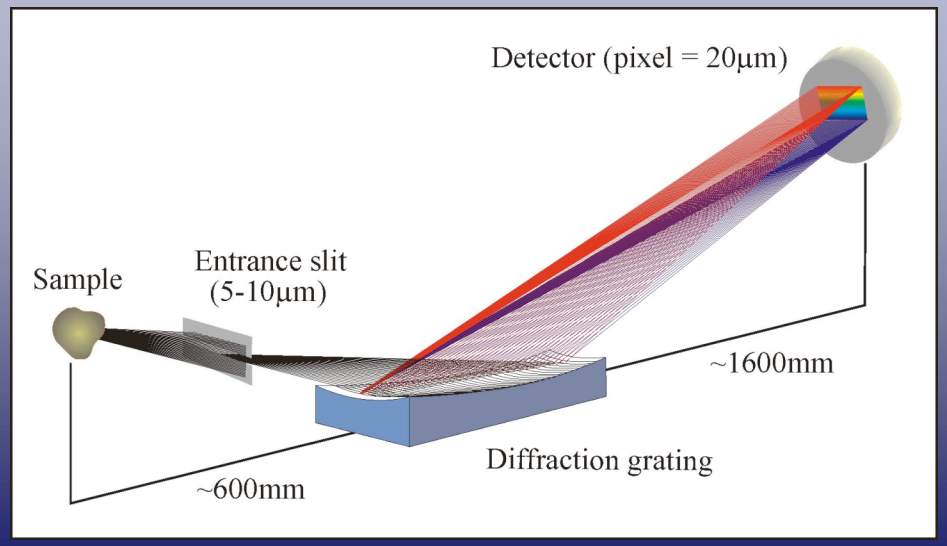
- neutral excitations (# ARPES)
- dipole forbidden (# optics)

Our reference: AXES @ ESRF



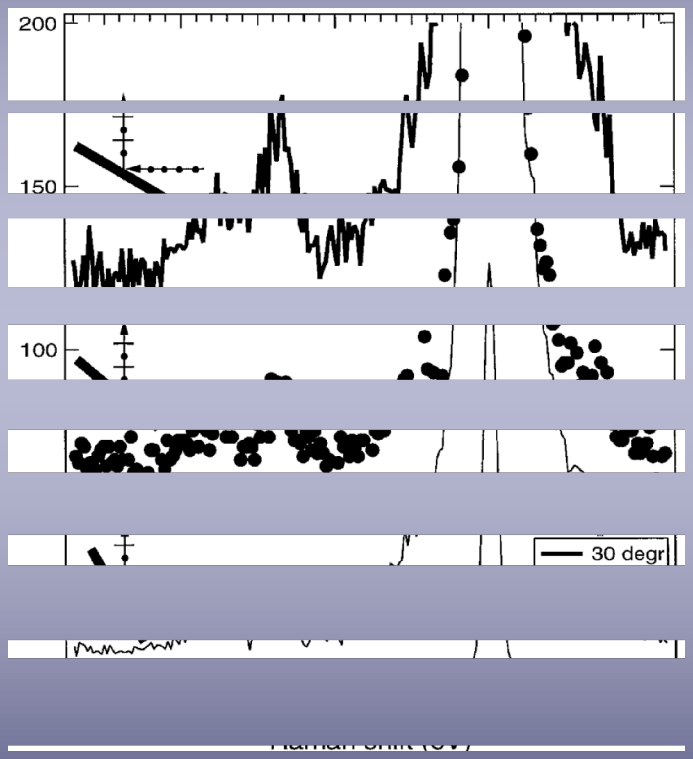
OFF-ROWLAND MOUNT
FIXED GRATING
FLAT FIELD

INFM - Politecnico di Milano
BL ID08 - ESRF

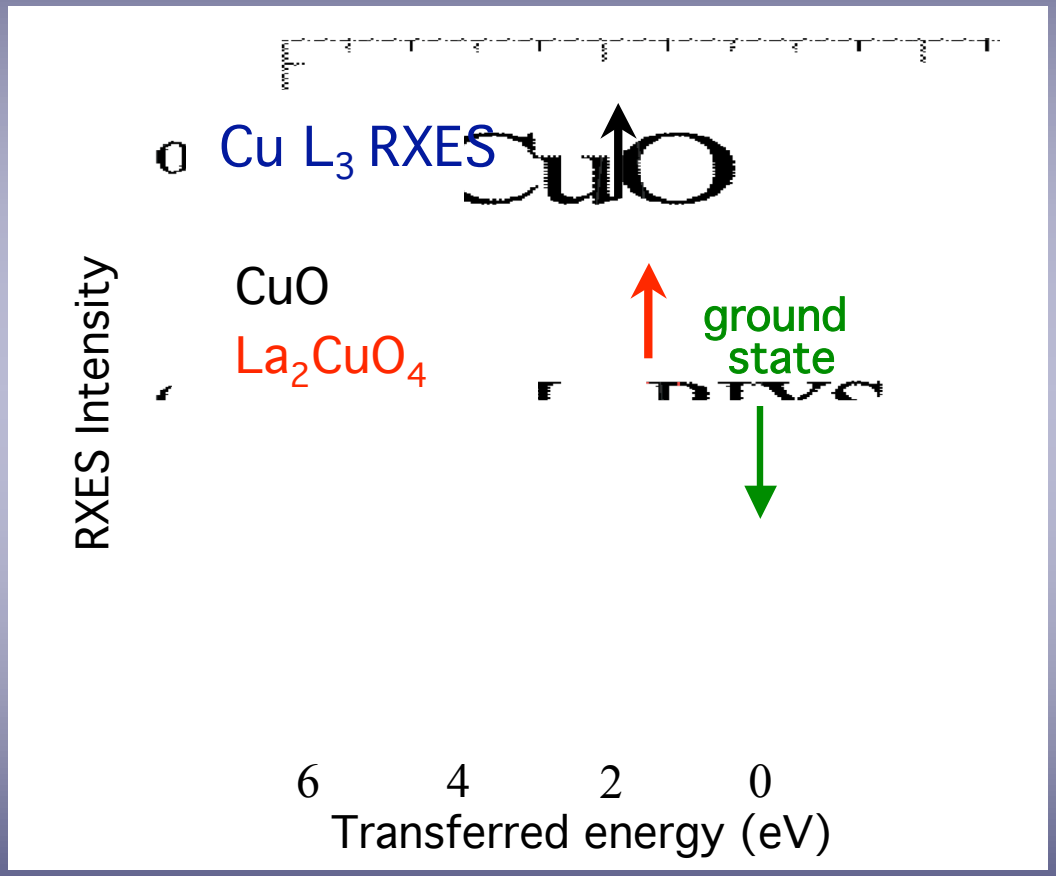


First experiment at the ESRF

$Sr_2CuCl_2O_2$ - $M_{2,3}$ edges



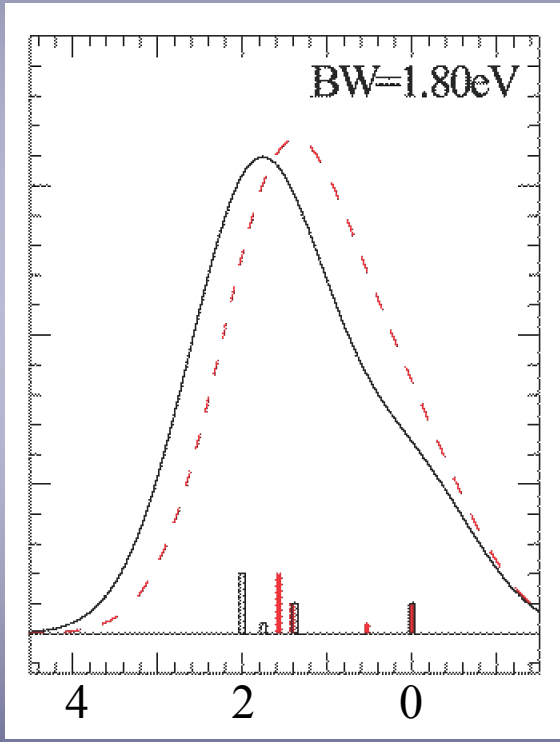
P. Kuiper *et al.*, PRL (1998)



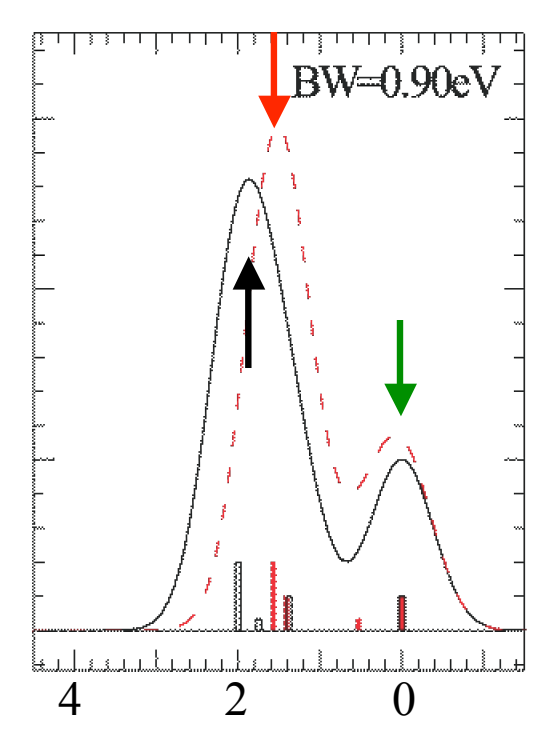
G. Ghiringhelli *et al.*, PRL (2004)

Pushing the resolution

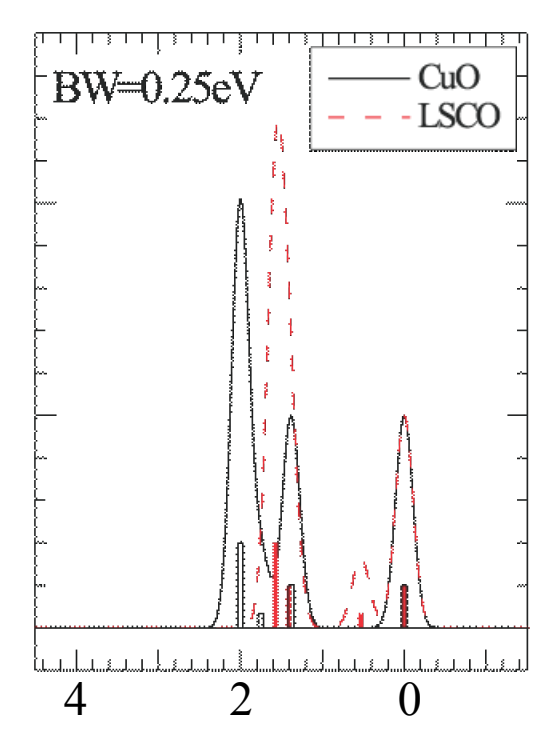
Past



Present (ESRF)



the future

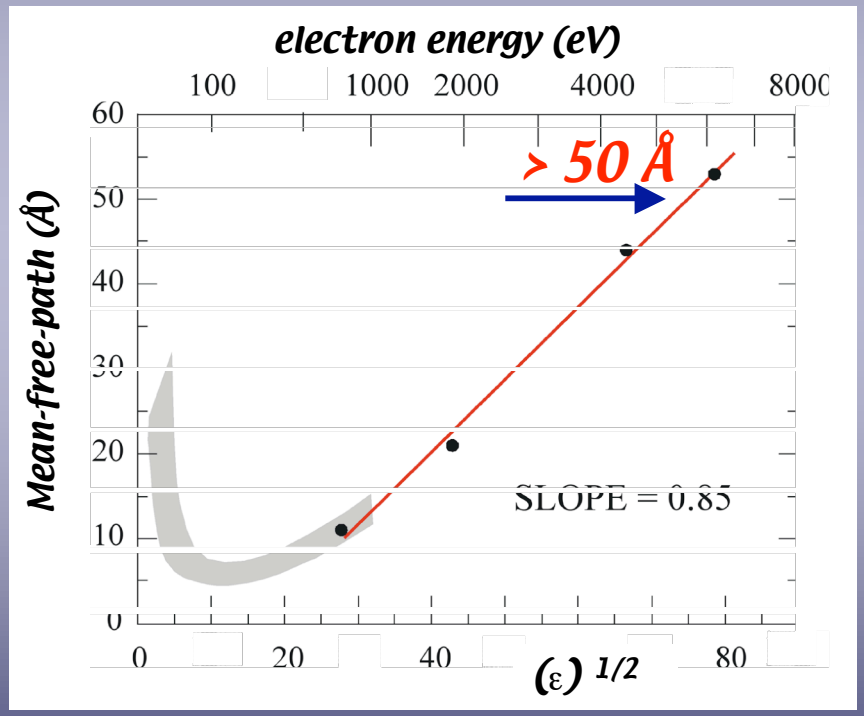


Transferred energy (eV)

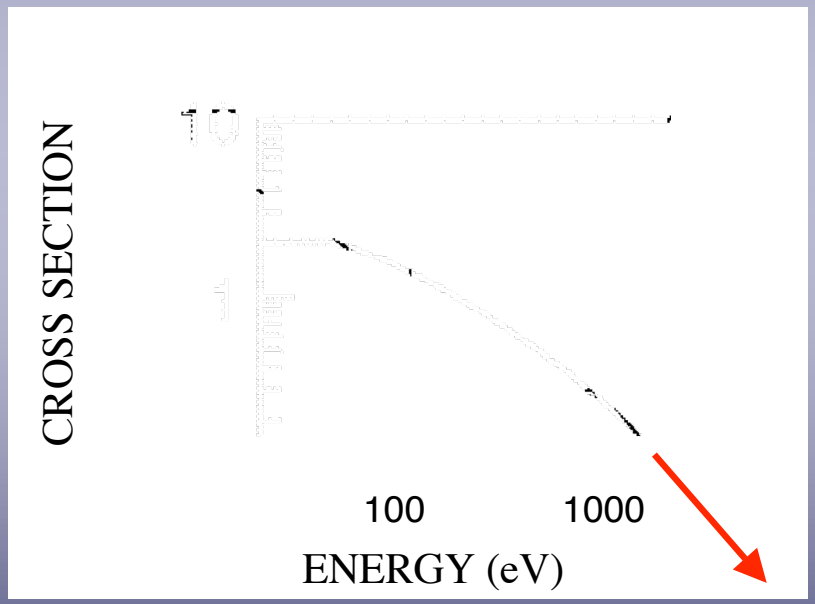
How can we probe deeper ?

2. PES with harder x-rays

Real bulk sensitivity with hard x-rays

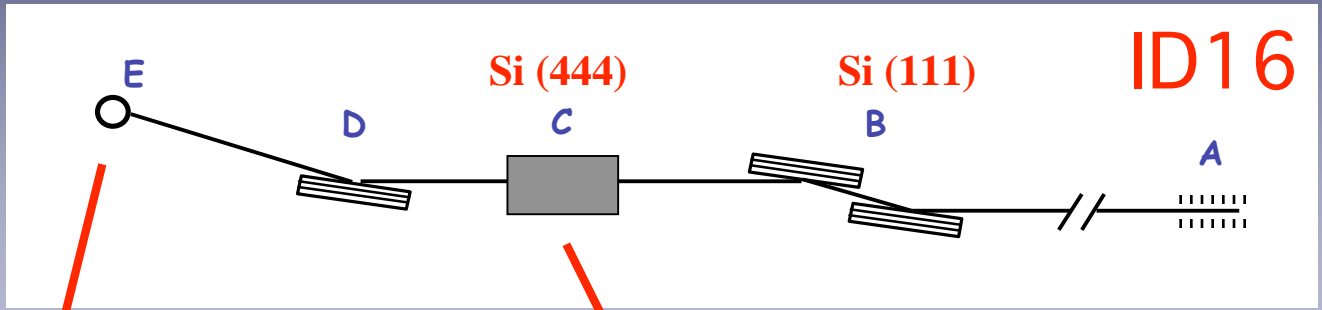


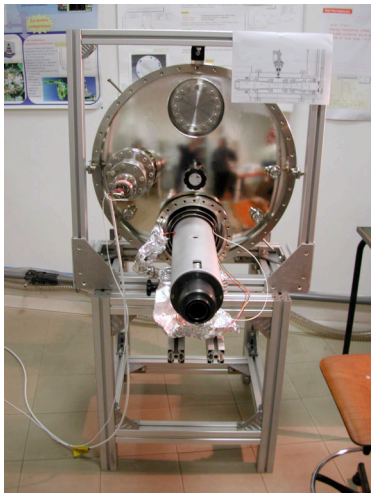
cross section



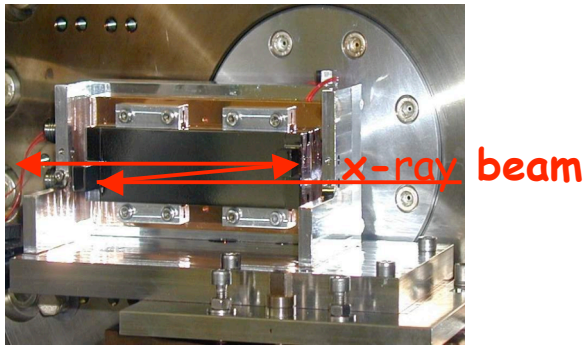
Dallera et al., (2003)

The european VOLume PhotoEemission (VOLPE) project





dedicated PES spectrometer



channel-cut post-mono in backscattering

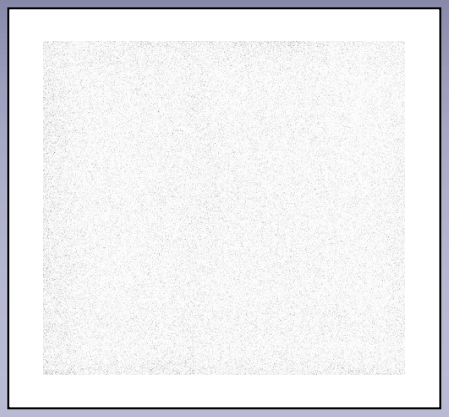
hn ~ 6 - 8 (10) keV

ESRF

Main features of VOLPE

high RR electrostatic lens (Rome)

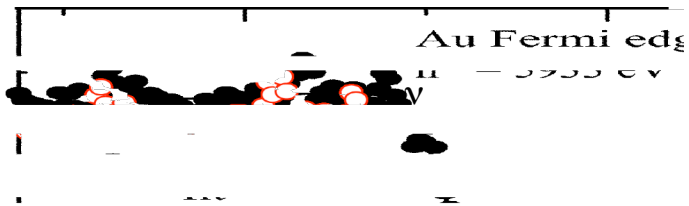
Low-noise detector (Trieste)



500 x 500 pixels
30 x 30 mm^2
20 hours

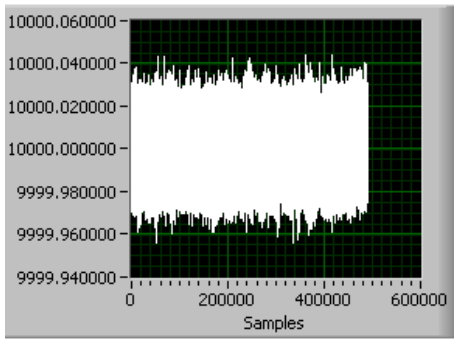
< 0.3 cts /(s cm^2)

Performances (june '04)



Δ

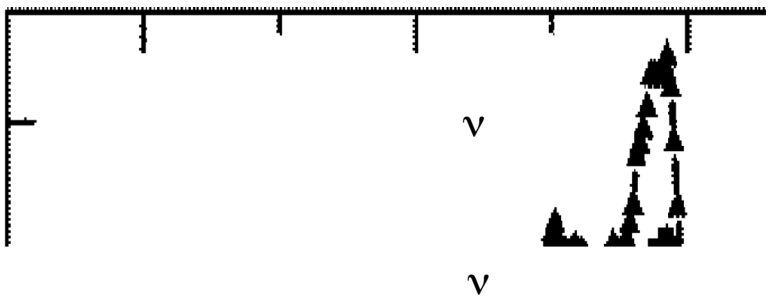
High-stability HV supplies



8 meV (rms) @ 10 keV / 24 hours

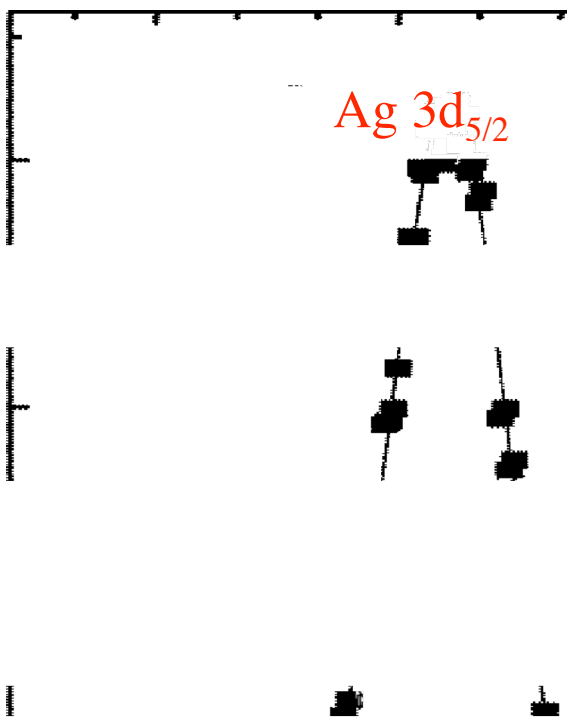
VOLPE: first results

counts !



s - band

Only 1 (bulk) component



hn = 7897 eV

Binding energy (eV)

How can we probe deeper ?

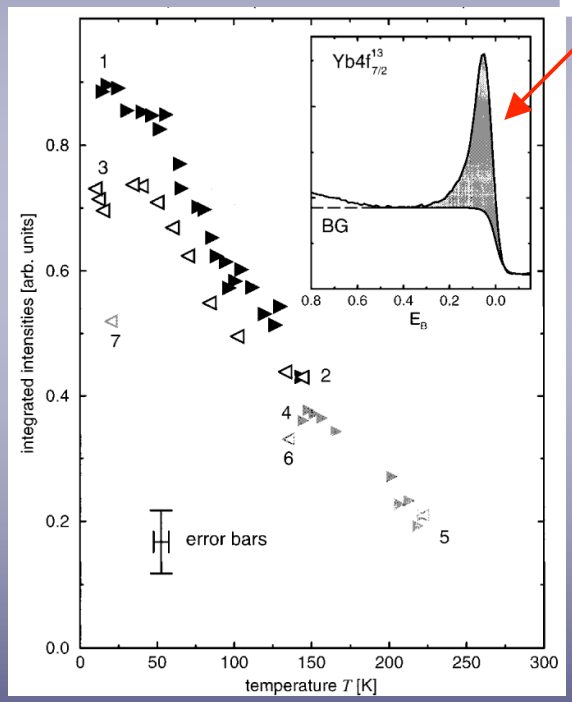
3. (Hard) x-ray emission

Valence transition in $YbInCu_4$

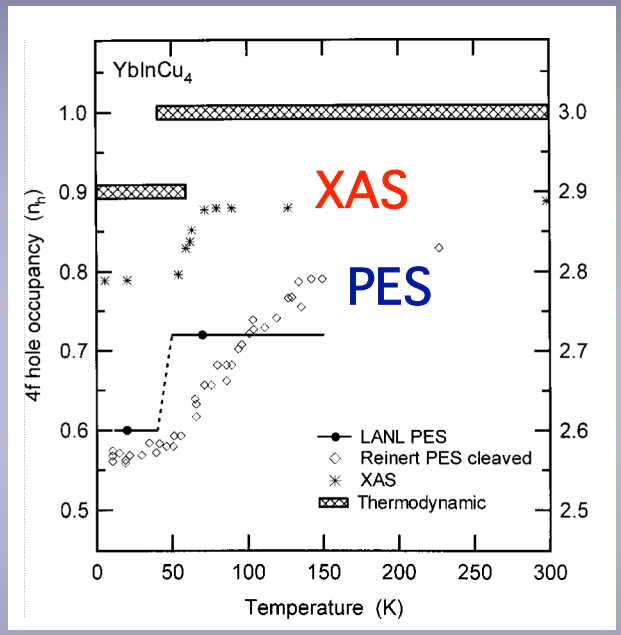
$v = 2.83 \rightarrow 2.96$; $T_c = 42$ K

PES

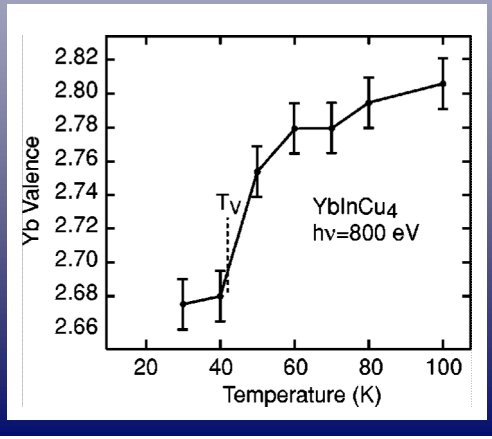
Kondo peak



F. Reinert *et al.* (1998)



D.P. Moore *et al.* (2000)



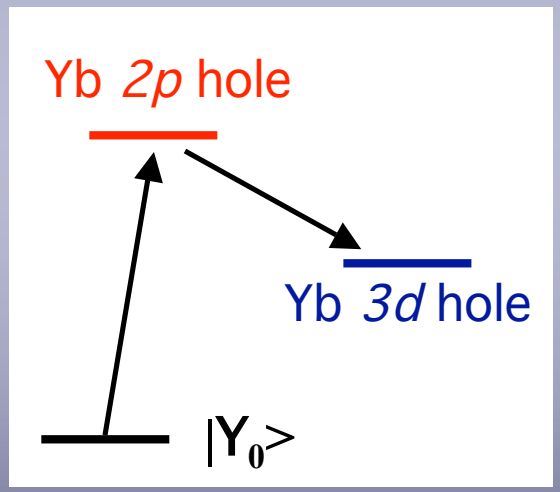
H. Sato *et al.* (2004)

Valence transition in $YbInCu_4$

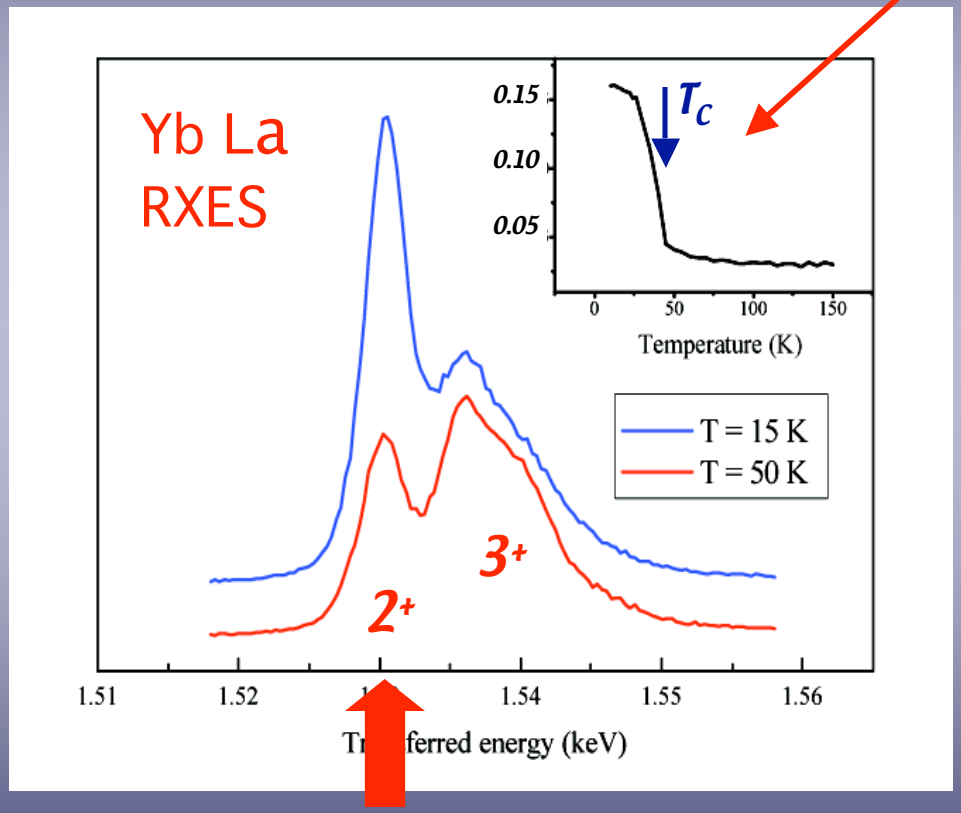
$v = 2.83 \rightarrow 2.96$; $T_c = 42$ K

Yb^{2+} weight ~ (3-v)

Yb La RXES



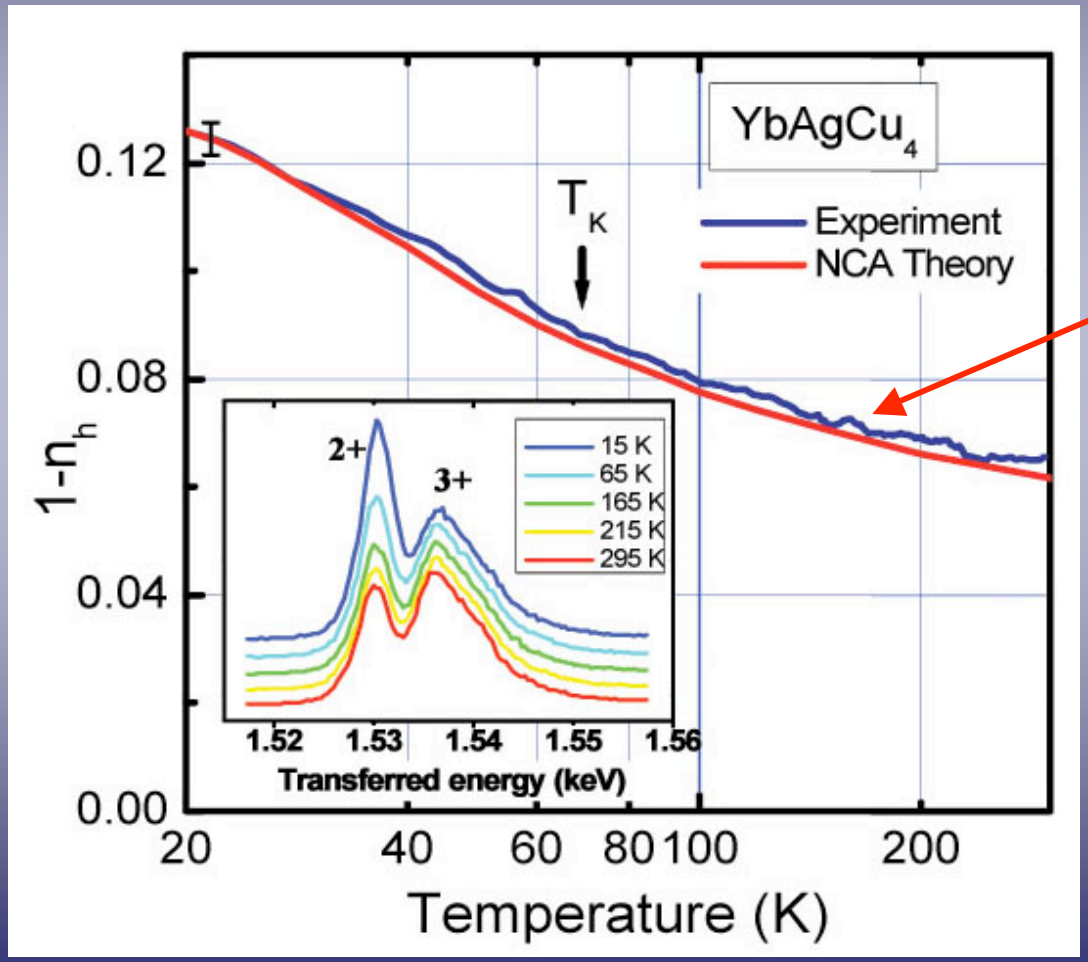
$h\nu_{out}$ ~ 7.4 keV



the small Yb^{2+} weight in the ground state is enhanced in the RXES transition

$YbAgCu_4$: Kondo scaling revealed

Yb La RXES

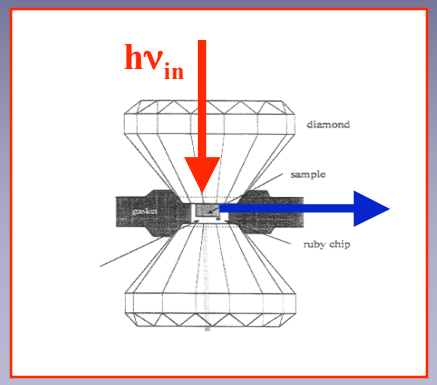


Direct evidence of the 'Kondo' temperature dependence (T_K=70 K)

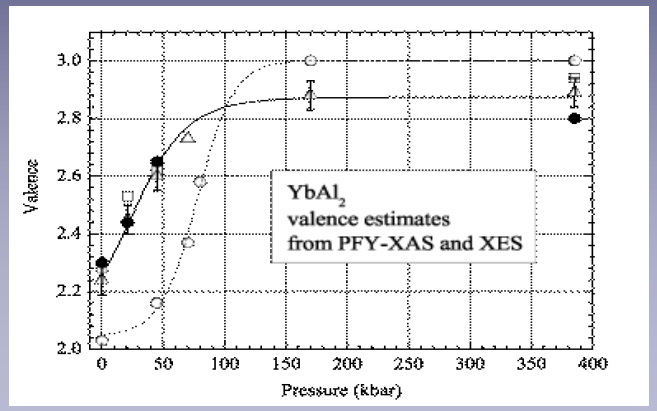
$k_B T_K$ = 6 meV
$h\nu_{in}$ ~ 9 keV !!

C. Dallera *et al.*
Phys. Rev. Lett. (2002)

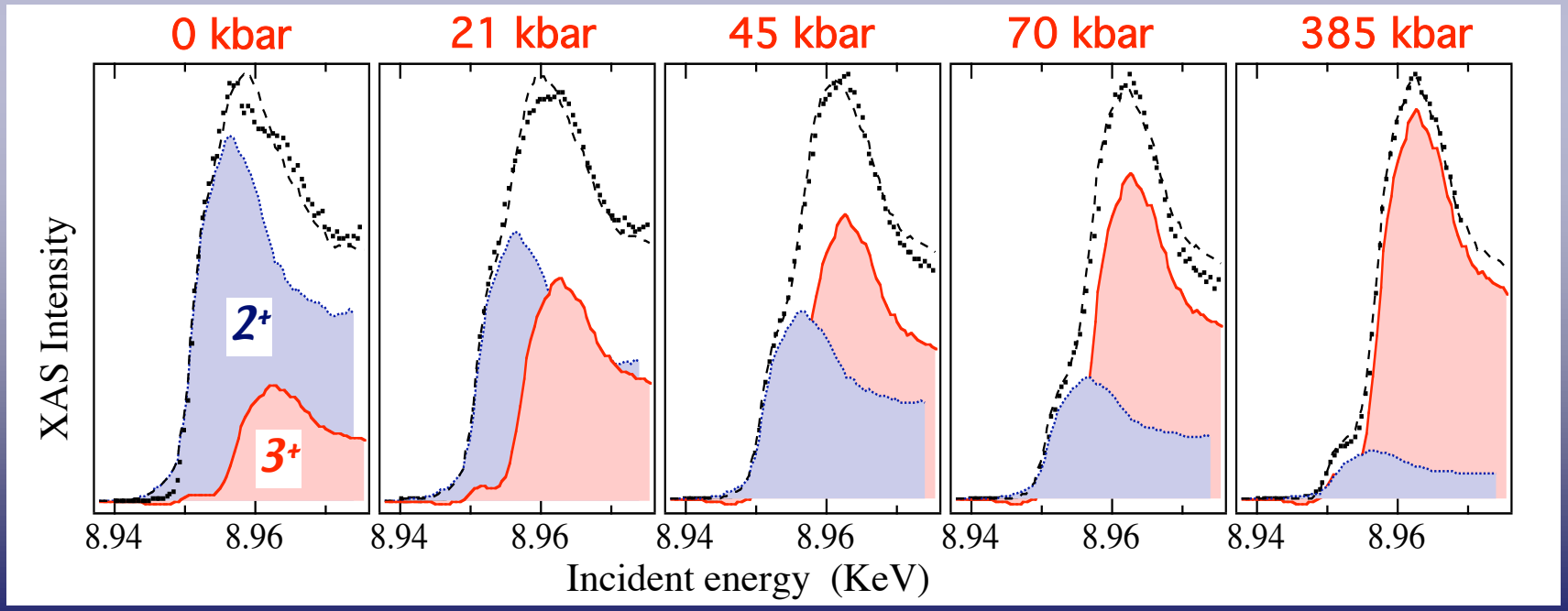
High-pressure studies with hard X-rays: "high-resolution" XAS in $YbAl_2$



Yb valence:
2.2 → 2.9



Yb La PFY-XAS



Dallera et al. (2003)

Summary

- New spectroscopic probes based on SR for strongly correlated systems
  (altius, citius, fortius)
- Photoemission
  - ARPES @ 1 keV
  - bulk-sensitive PES @ 8 keV
- Photon in - photon out spectroscopies