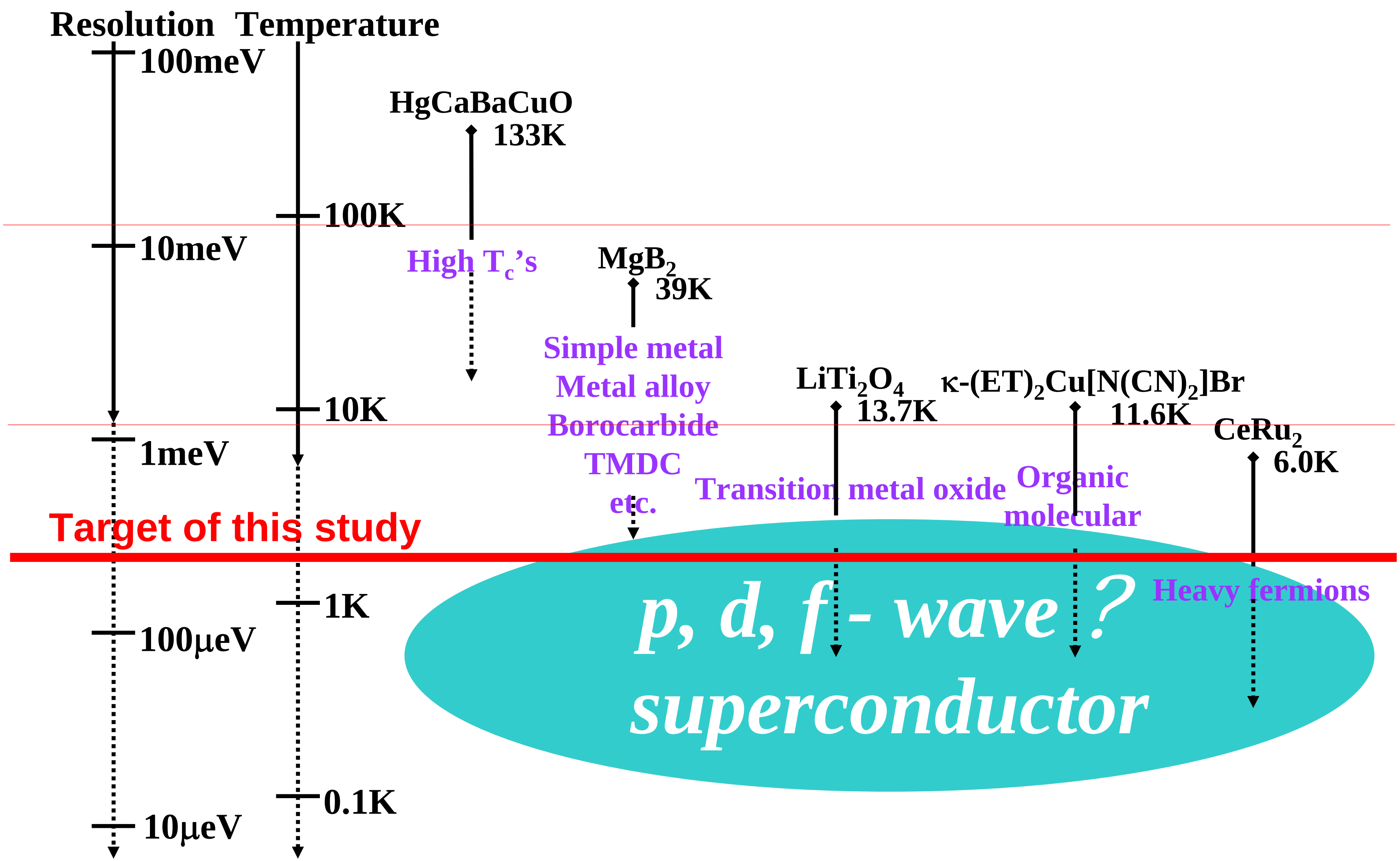
Ultrahigh-resolution photoemission study of superconductors and strongly correlated materials using quasi-CW VUV laser

S. Shin,[1,2] T. Kiss,[1] S. Tsuda,[1] K. Ishizaka,[1] T. Yokoya,[1]
and
T. Togashi,[2] C. Chen,[3] S. Watanabe[1]

[1]*Institute for Solid State Physics, University of Tokyo*
[2]*RIKEN*
[3]*Beijing Center for Crystal R&D, Chinese Academy of Science*

Research of Superconductors by PES



contents

1. Development of Laser photoemission system
   - High resolution Photoelectron analyzer
   - Quasi-CW VUV laser
   - Low temperature Cooling system

2. High resolution PES study on superconductors

   Nb ; simple metal superconductor

   $CeRu_2$; 4f electron superconductor with Onuki et al

   MgB_2 ; intermetallic compound with Tajima et al

   κ-$(ET)_2Cu[N(CN)_2]Br$; organic superconductor with Kanoda et al

   κ-$(ET)_2Cu(SCN)_2$

3. High resolution PES study on Kondo material

   LiV_2O_4 ; Kondo state in TMO with Ueda et al

4. Bulk sensitive PES in low photon energy region

   $SrVO_3$; comparison with DMFT with Inoue et al

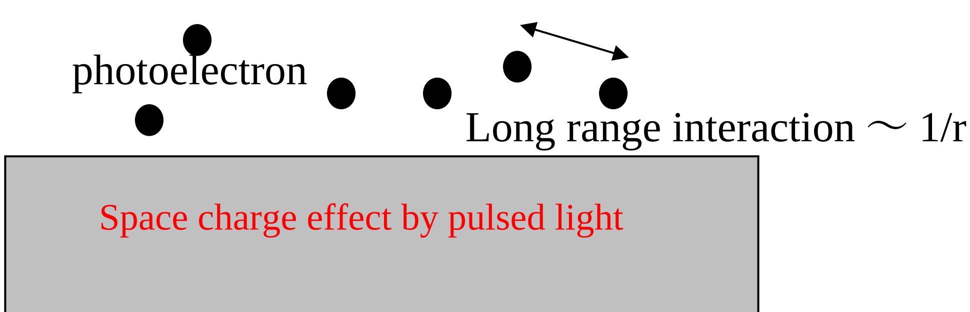
5. Conclusion and Future of Laser-Photoemission

What limits the photoemission resolution ?

1. Line width of light source → It will be settled by developing laser
2. Electron analyzer
   - Slit width
   - Engineering precision
   - Disorder of work function
   - Instability of power source
3. Residual magnetism
4. Ripple in ground level

(Items 2–4) → They have been resolved. Recent progress is in energy region below 1 meV

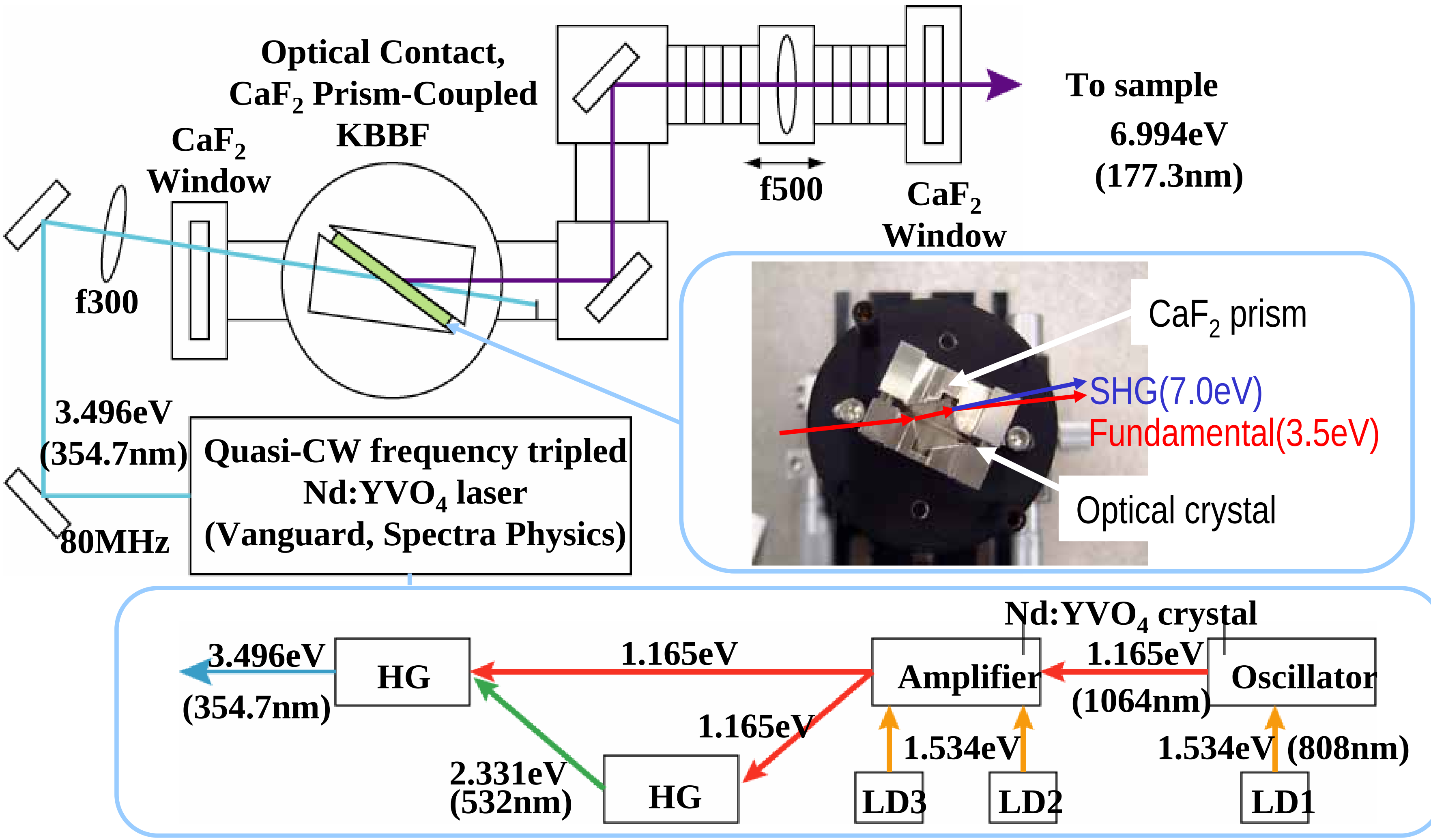
5. Space charge effect → CW or quasi-CW light source should be necessary (= weak peak intensity, but strong average intensity)



Laser system

6^{th} harmonic (6.994eV) of $Nd:YVO_4$ laser using KBBF crystal

T.Togashi ,*et al*., Opt. Lett. **28**, 254 (2003)



Specification of new Laser

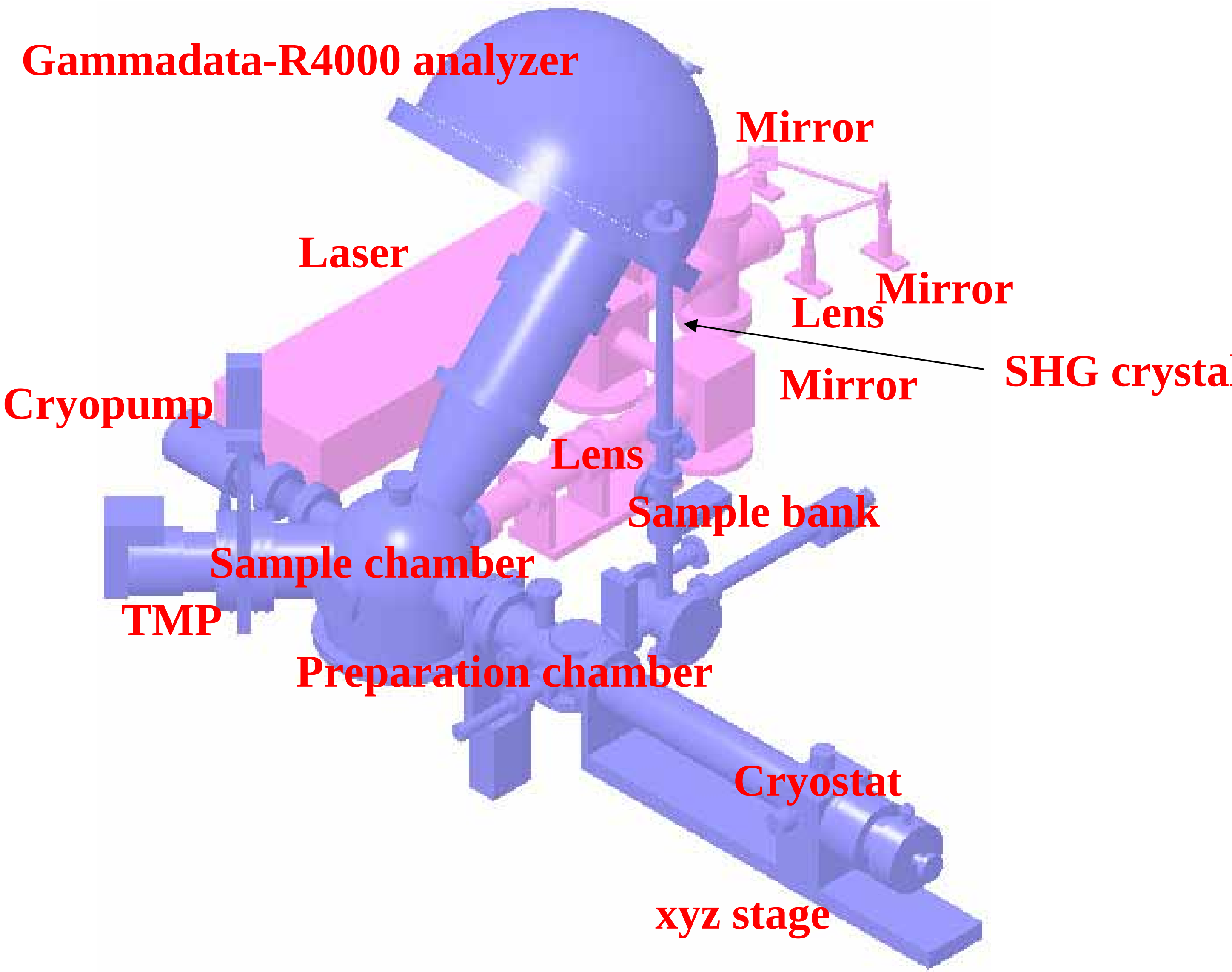
Highest photon energy quasi CW Laser

	Laser	HeIα
Photon energy	6.994eV	21.218eV
FWHM	0.26meV(0.09meV with etalon)	~1meV
Frequency	80MHz (quasi CW)	CW
Total Photon flux	2.2x10^{15} photons/sec	~10^{13} photons/sec
polarization	vertical, horizontal, circular	none
Size of spot	0.2μm – 0.5mm	6~8mm

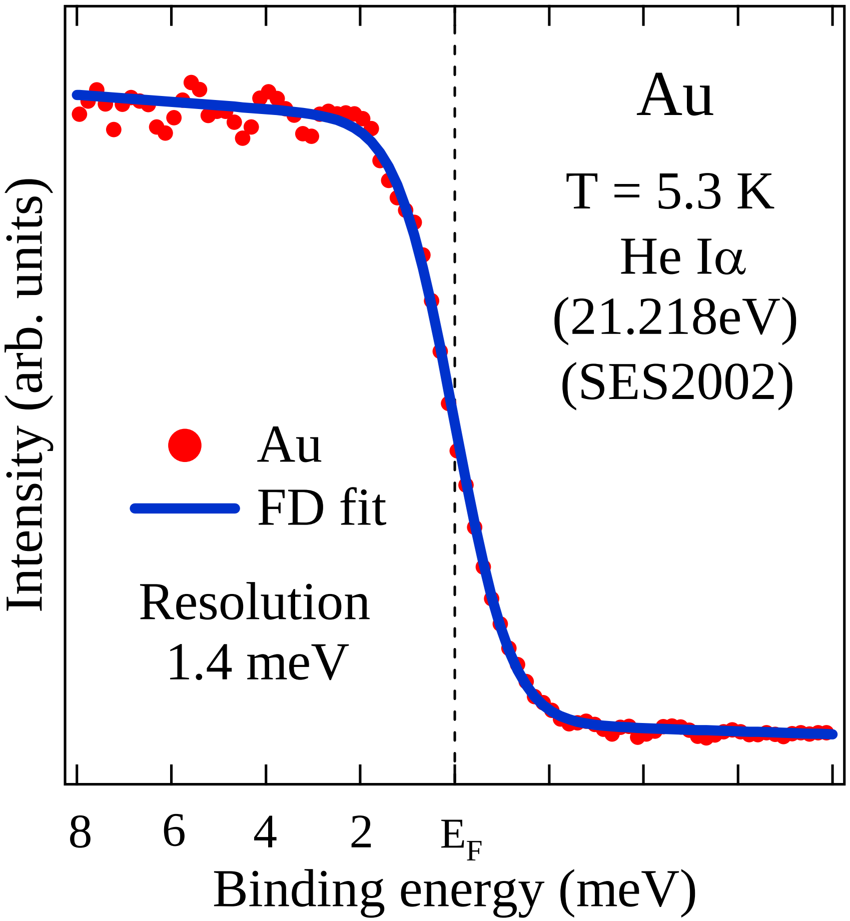
Microscopy
Bulk sensitive
High energy resolution
Polarization dependent
Low damage of sample

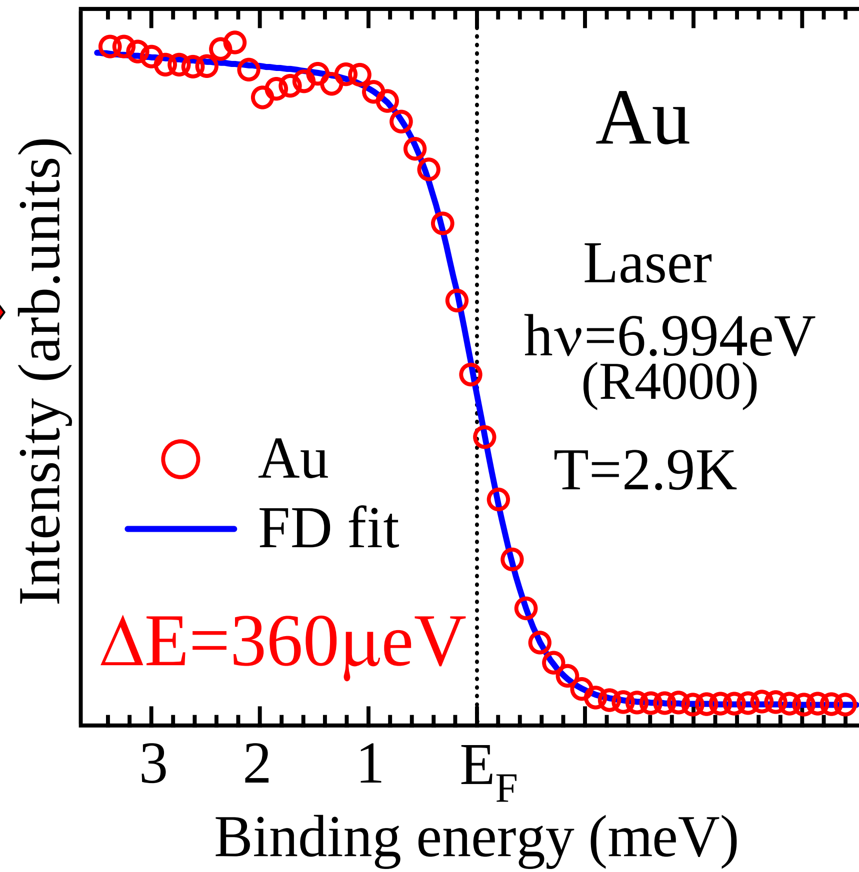
Photon flux of SR is weak,
and becomes weak in proportion to the increase of the monochromator resolution
Cf. 〜10^9 phs./sec (1 meV resolution at 100eV)

Laser excitation photoemission spectrometer



Performance of the experimental system





$$0.36^2_{\text{total}} = 0.25^2_{\text{analyzer}} + 0.26^2_{\text{linewidth}}$$

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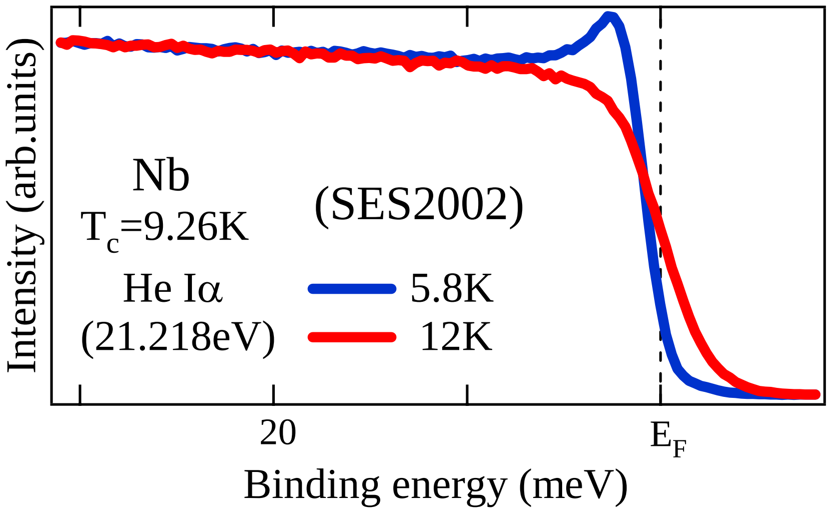
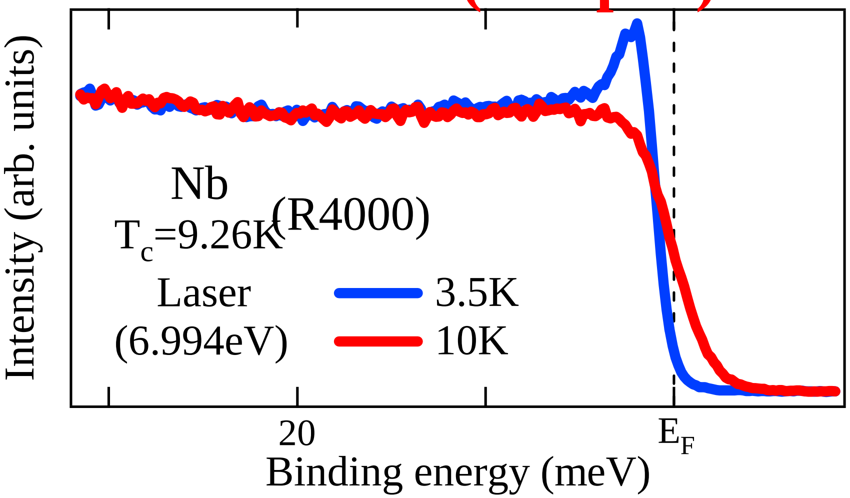
Superconducting gap of Nb

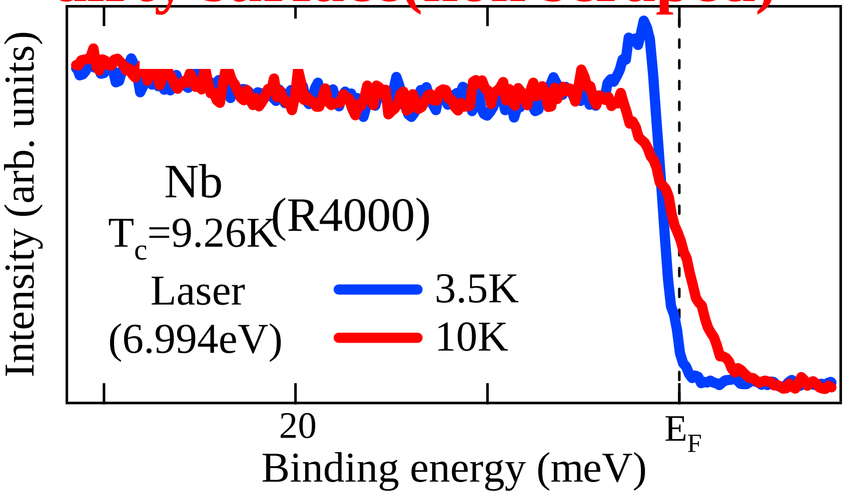
Chainani et.al., PRL, 85(2000)1966



clean surface(scraped)



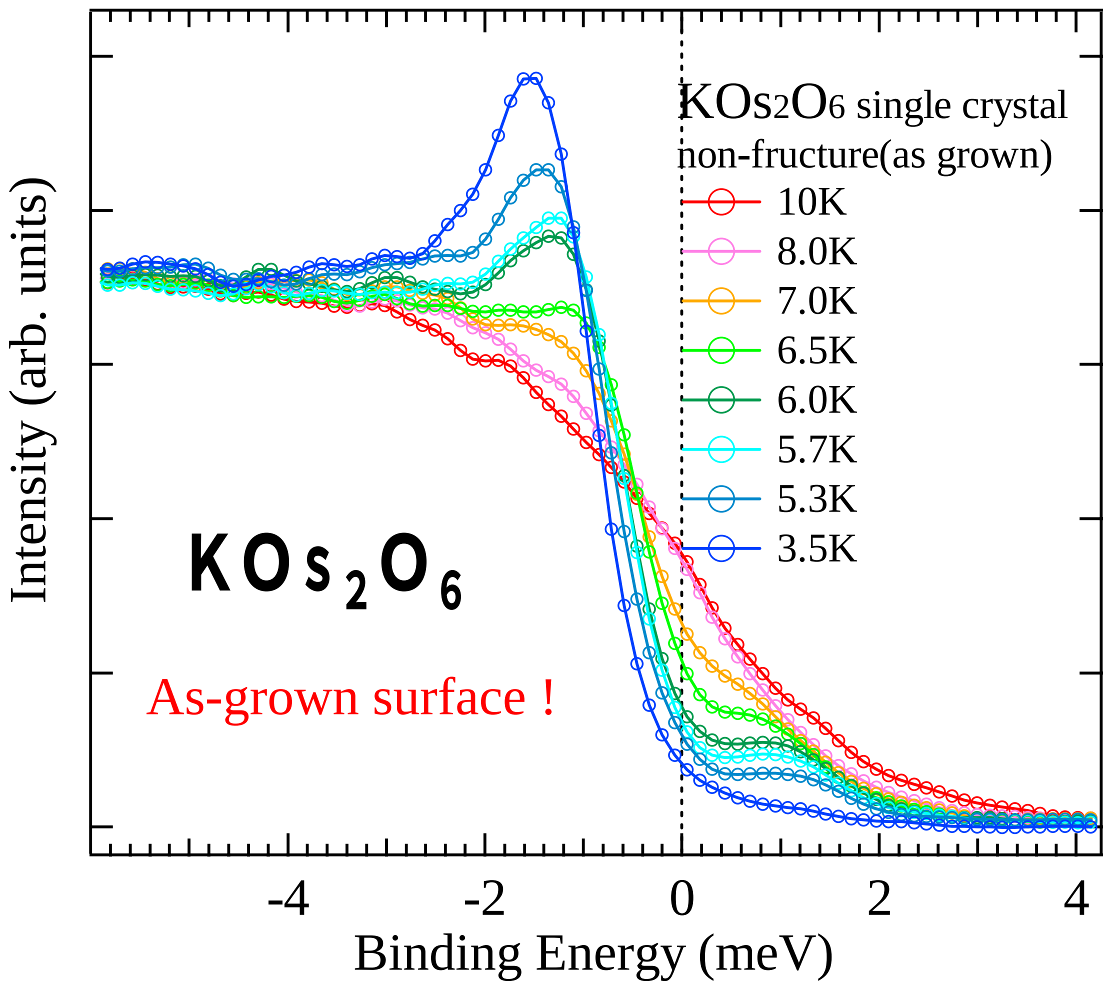
dirty surface(non scraped)



We can measure superconducting gap without clean surface

Another example

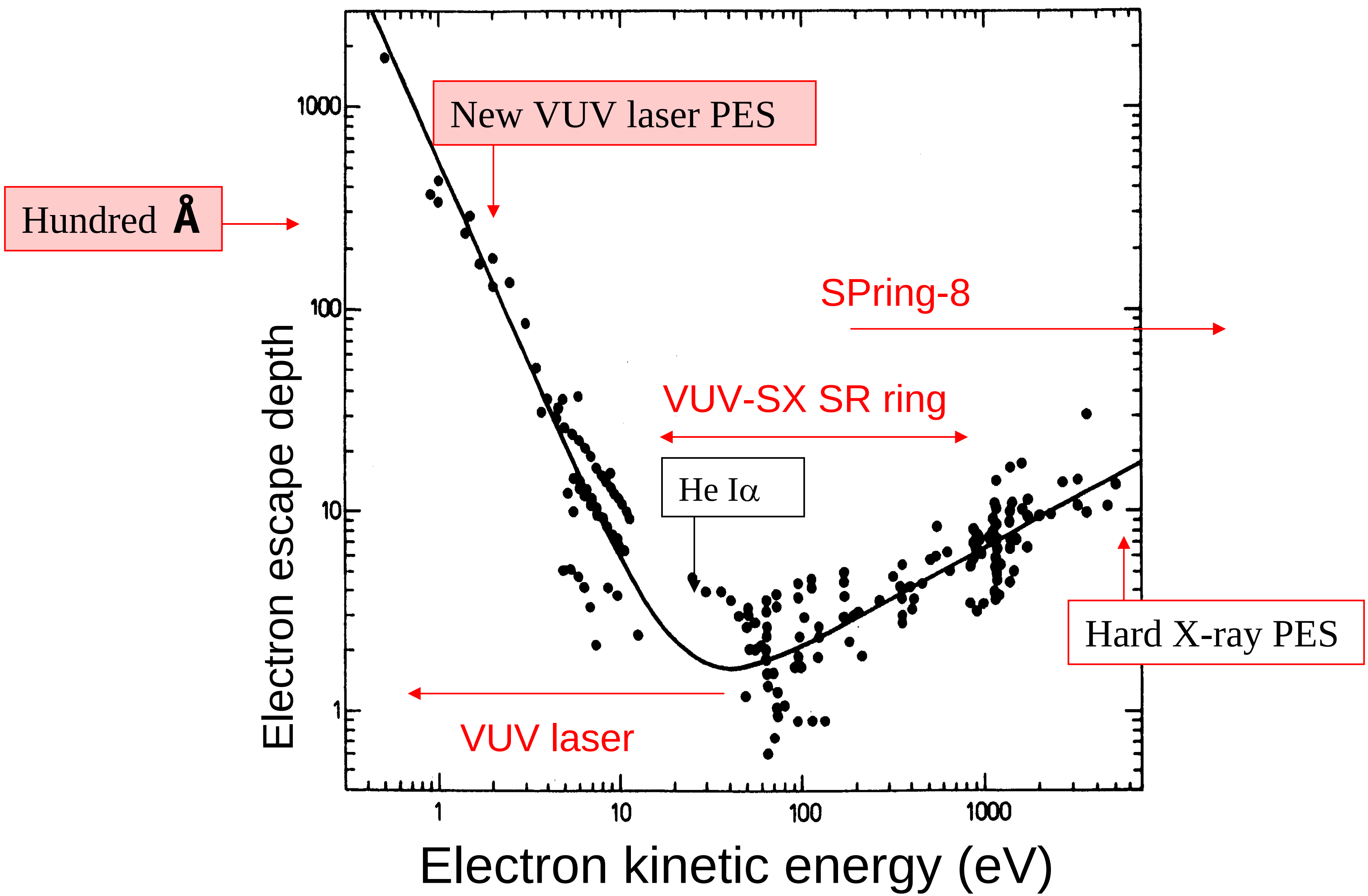
Without surface cleaning, we can measure superconducting gaps of new materials whose crystal size is quite tiny



Hiroi Group(2003)

- Geometrically Frustrated Pyrochlore
- Unconvetional superconductor ?

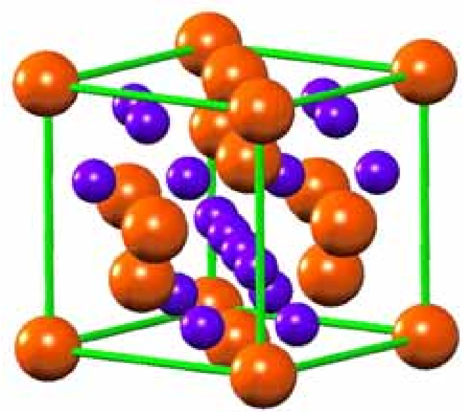
Laser-PES is the most bulk sensitive PES



$CeRu_2$

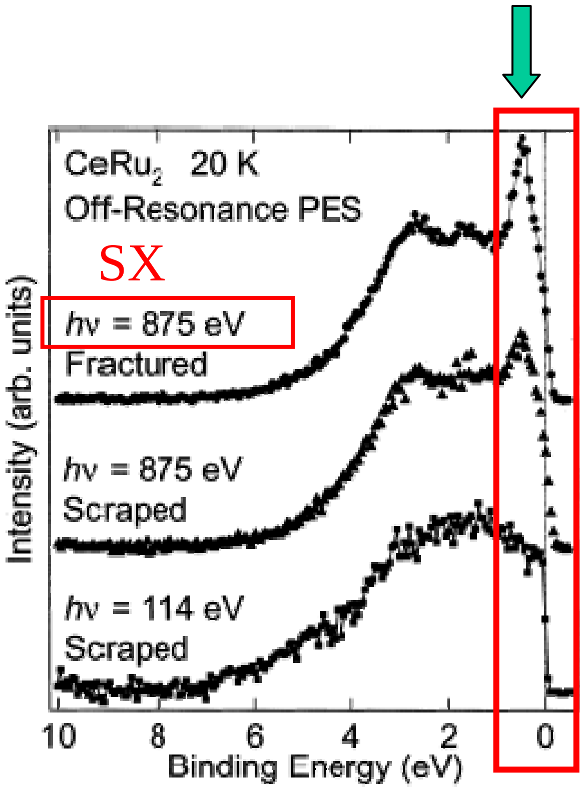
1. Valence fluctuation　(T_K>1000K、Ce valence ~ 3.7)
2. Superconductor that has the highest Tc =6.2K in Ce compounds

Experiment	$2\Delta(0)/k_BT_C$	Symmetry
Break-junction [1)]	**4.1-4.4**	**s-wave**
NMR [2)]	**4**	**s-wave(anisotropic)**
Specific Heat [3)]	**3.7**	**s-wave**
NMR [4)]	**3.7**	**s-wave (large anisotropy)**
STS [5)]	**3.3-6.6**	**s-wave**
Specific Heat [6)]	**-**	line node (largest anisotropy)



1) T. Ekino *et al*., Phys. Rev. B 56(1997)7851
2) K. Matsuda *et al*., J. Phys. Soc. Jpn. 64(1995)2750
3) M. Hedo *et al*., J. Phys. Soc. Jpn. 67(1998)272
4) H. Mukuda *et al*., J. Phys. Soc. Jpn. 67(1998)2101
5) J.Mag.Mag.Mater 47-48(1985)542.　Phys. Rev.B56(1997)7851.　J. Phys. Soc. Jpn 69(2000)1970.　Low Temp.Phys.27(2001)613.
6) J. G. Sereni *et al*., Mod. Phys.Lett. 3(1989)1225

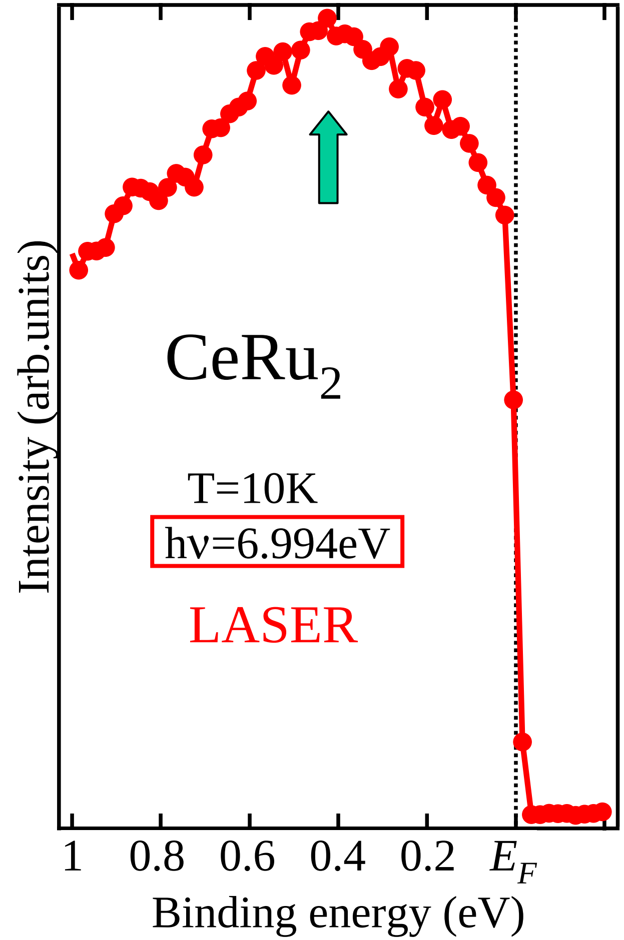
Valence band spectrum of $CeRu_2$



Bulk

Surface

A. Sekiyama *et al.*, Solid State Commun. 121(2002)561



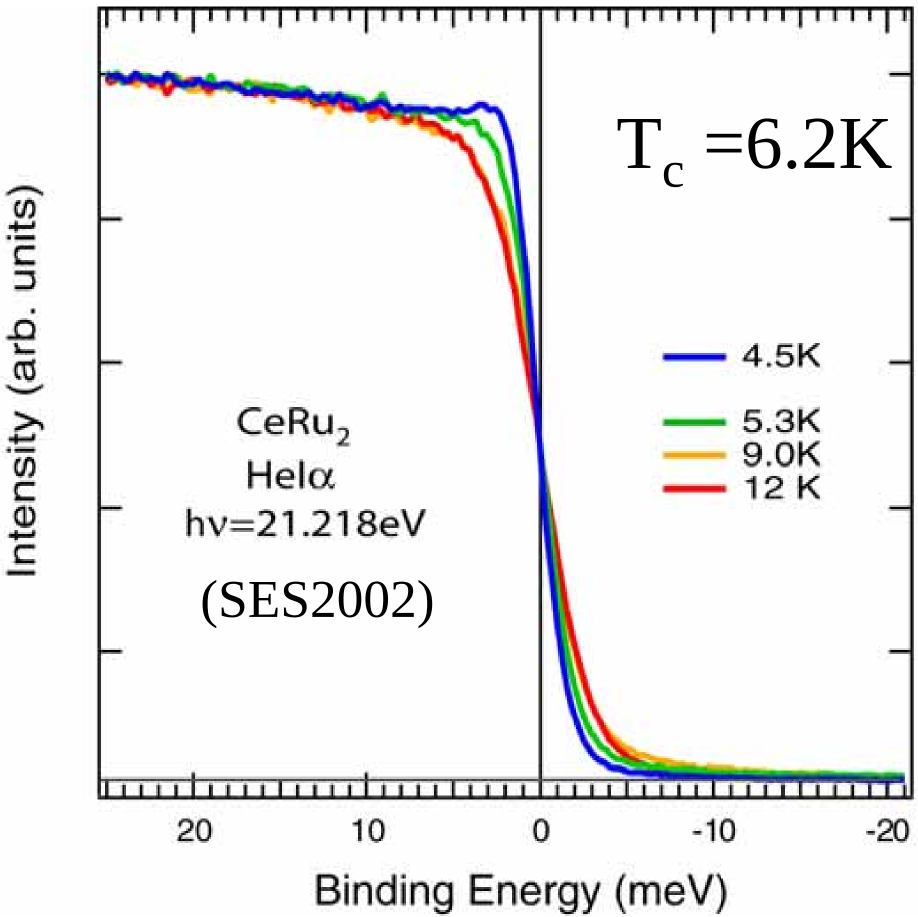
0.4eV peak

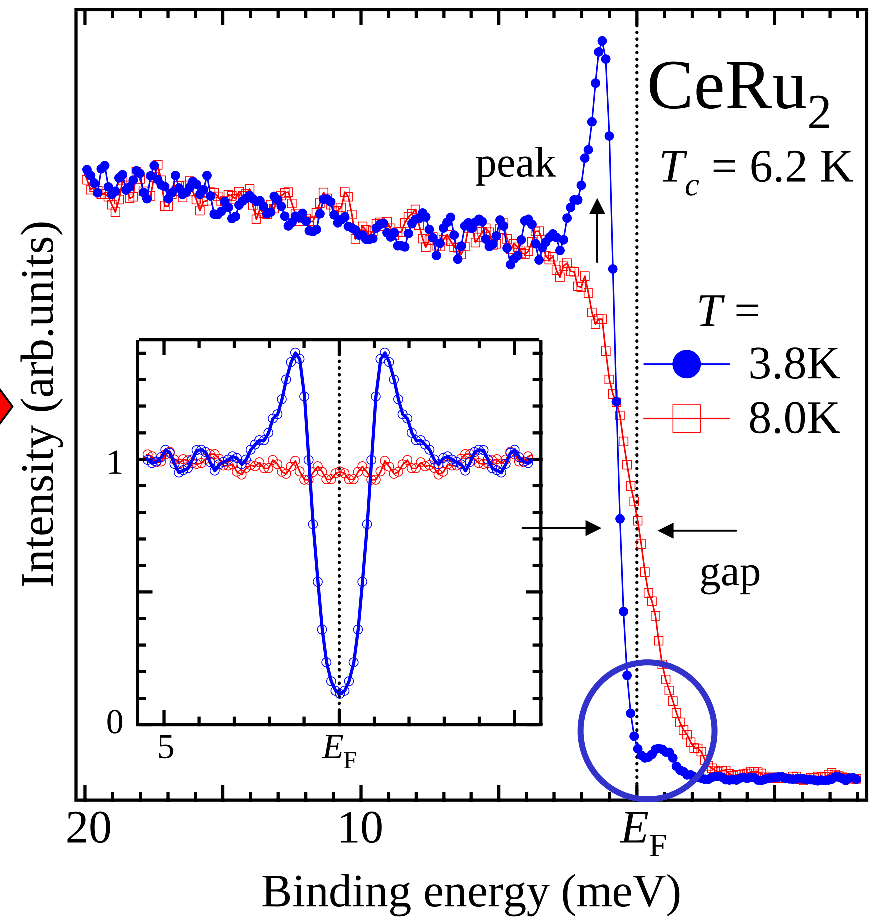
Bulk sensitive

Bulk sensitive in **high** energy region

Bulk sensitive in **low** energy region

Superconducting gap of $CeRu_2$

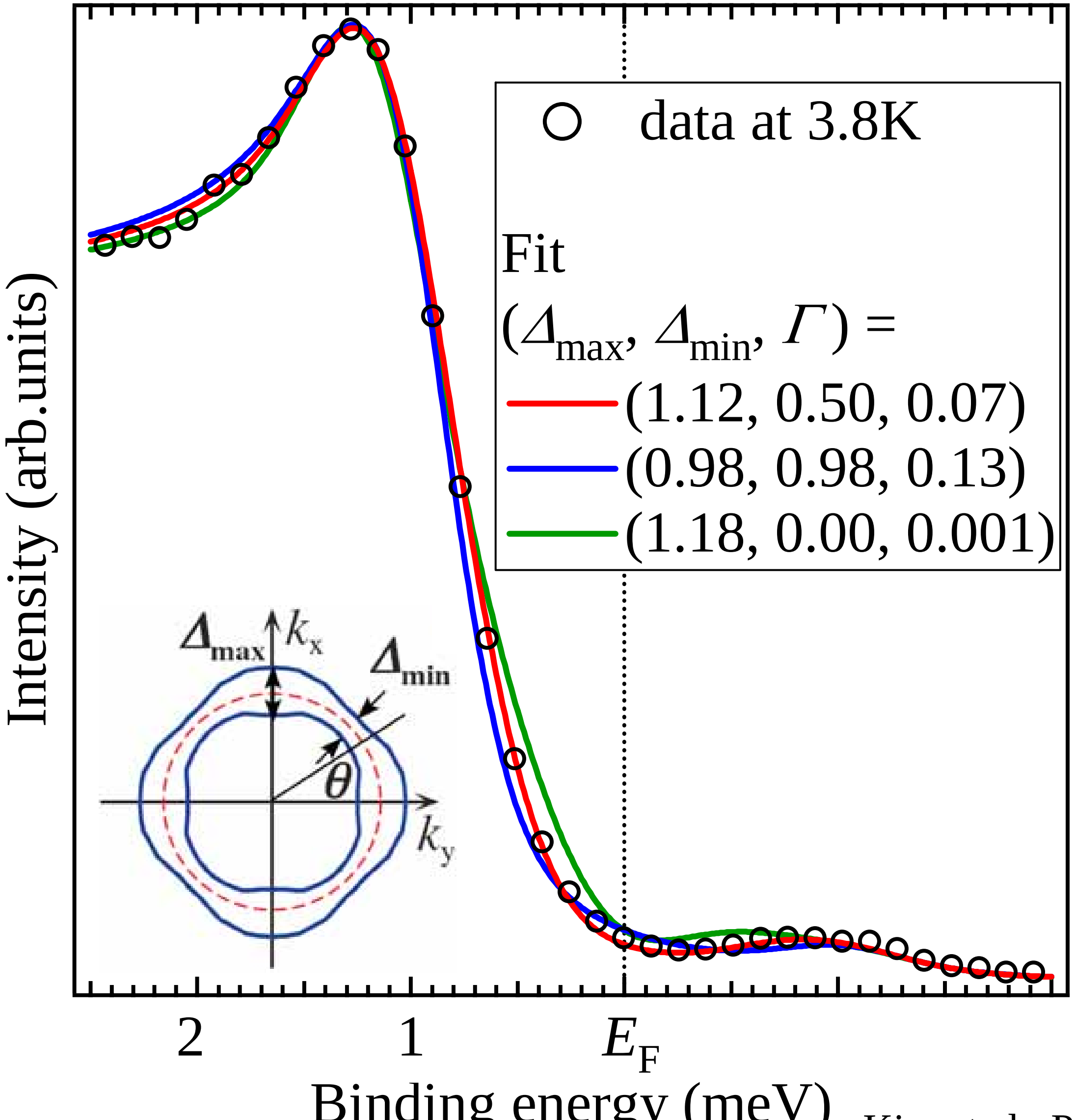




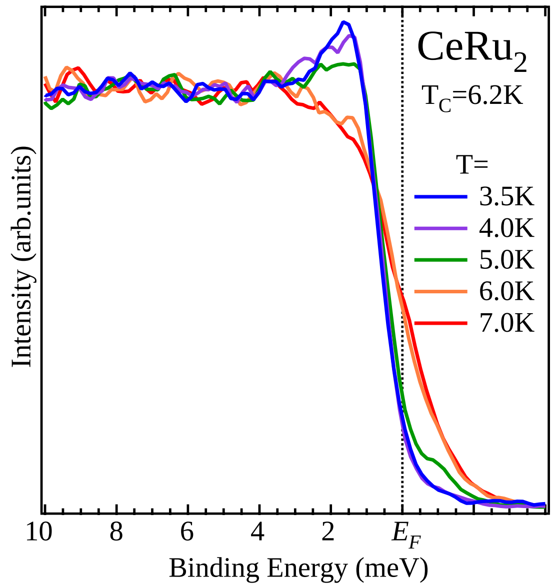
Superconducting gap was clearly observed by laser-PES

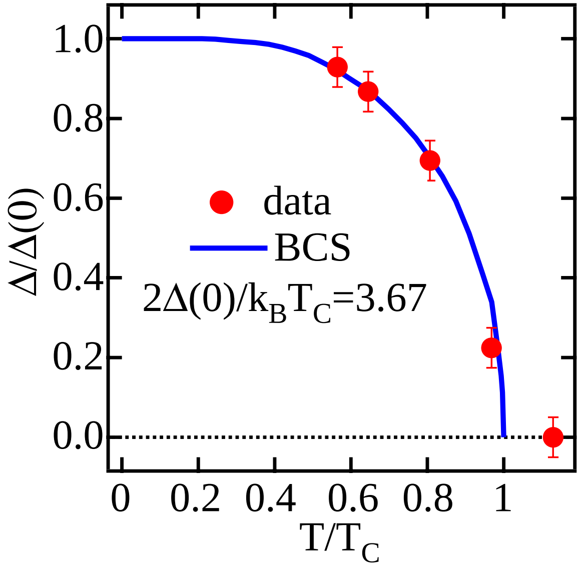
Kiss et al., PRL **94** (2005)57001

Anisotropic Superconducting gap of $CeRu_2$



Temperature dependence





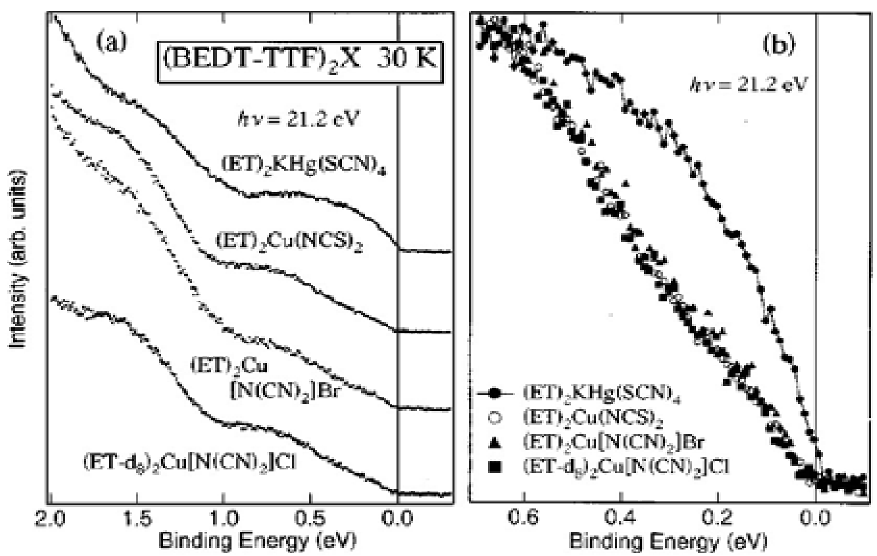
Kiss et al., PRL **94** (2005)57001

Superconducting gap of organic superconductor κ-$(ET)_2Cu[N(CN)_2]Br$, κ-$(ET)_2Cu(SCN)_2$ d-wave superconductor?

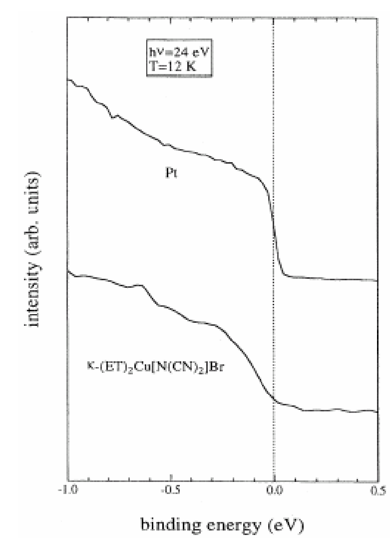
Previous works

- No intensity a E_F; They look like insulators
- All the ET-materials have the similar spectra and are inconsistent with the band calculation

Why ? → Surface effect ?
or
Electron correlation effect ?

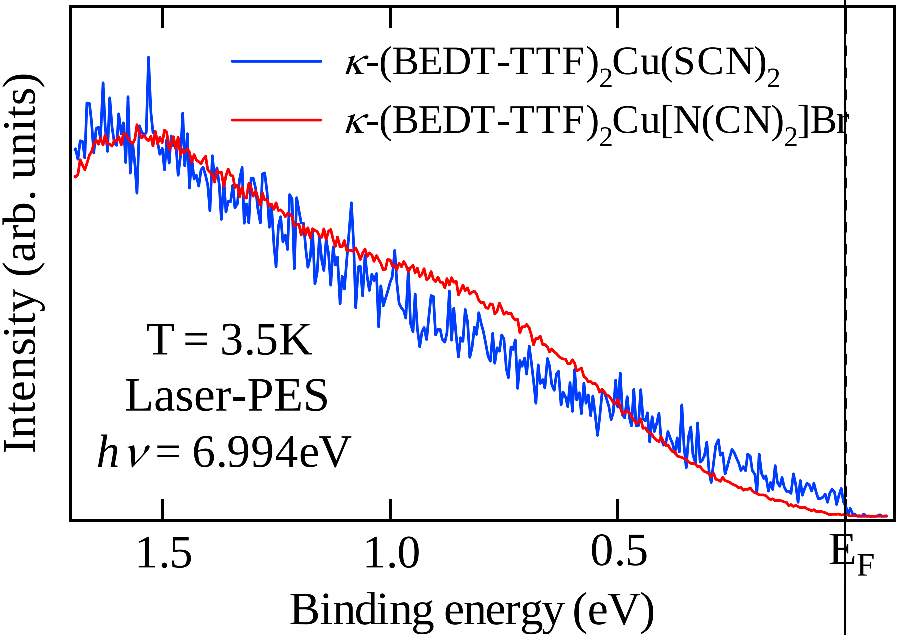


Sekiyama et al., Phys. Rev. B **56** (1997) 9082.

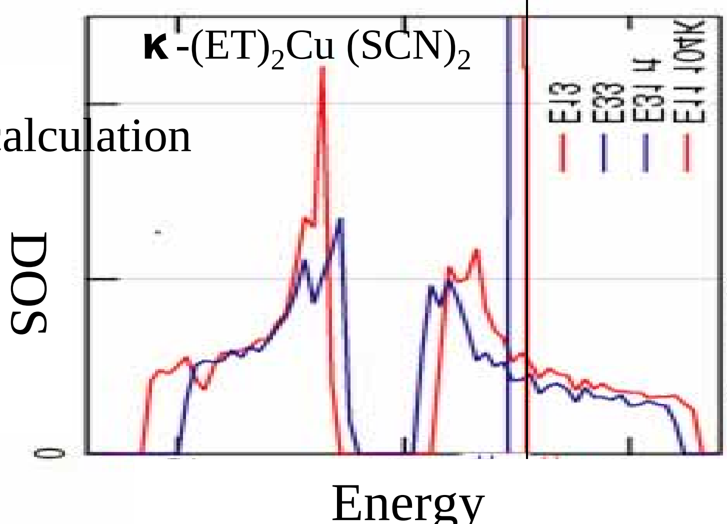


R. Liu et al., Phys. Rev. B **51** (1995)

Comparison with the band calculation
κ-$(ET)_2Cu[N(CN)_2]Br$, κ-$(ET)_2Cu(SCN)_2$



Band calculation



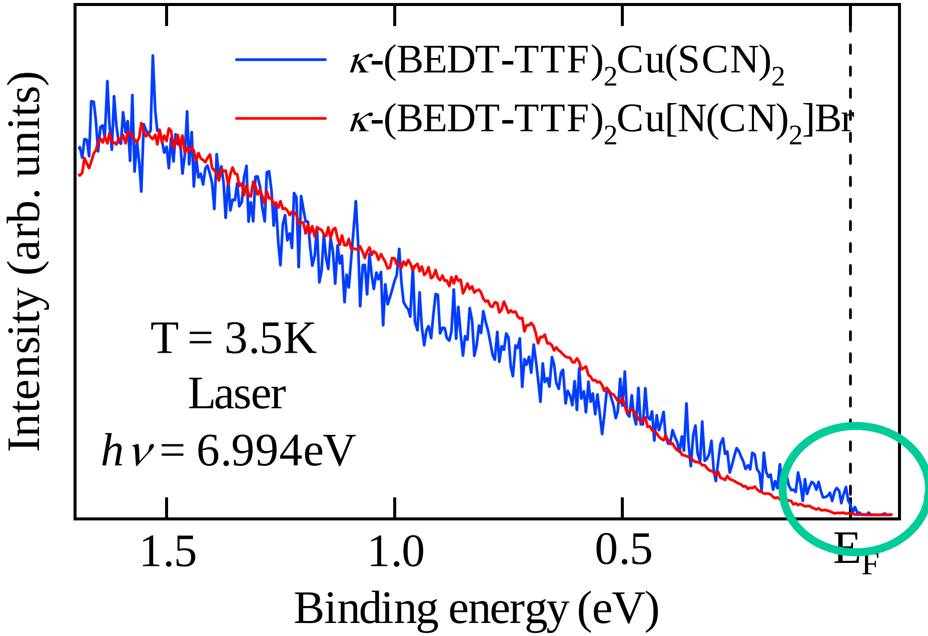
H.Mori, Private communication

1. The band calculation has been quite important for the materials design of new organic materials
2. But bulk-sensitive PES spectra are inconsistent with the band calculation in
   - DOS
   - Fermi edge

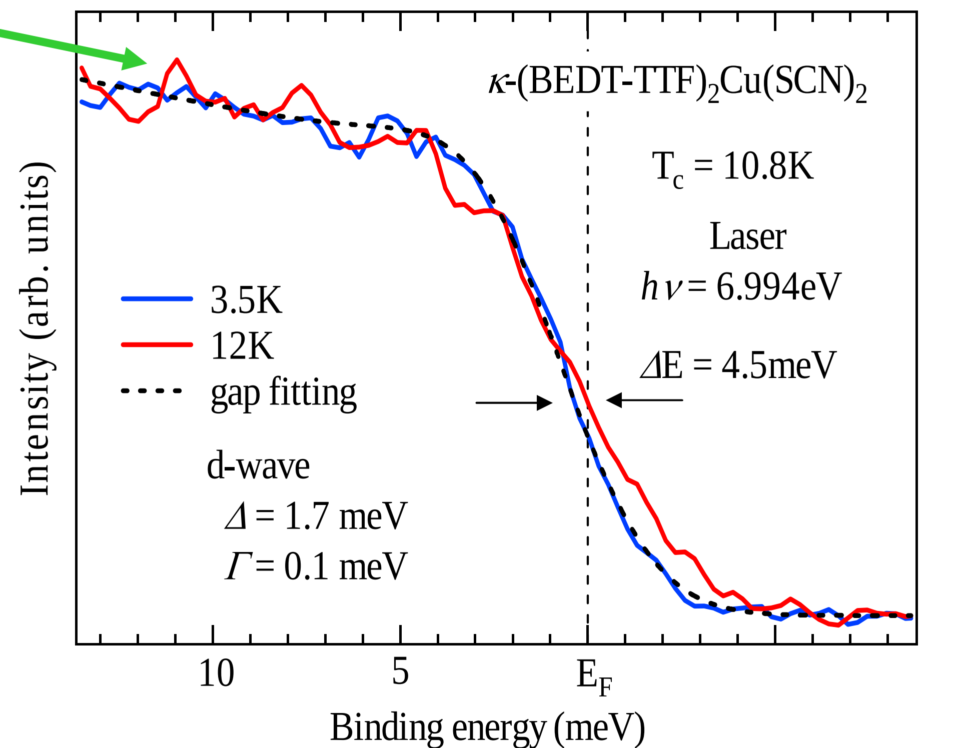
⇩

Electron correlation effect is essential, rather than the Surface effect ?

Fermi edge and Superconducting gap
κ-$(ET)_2Cu[N(CN)_2]Br$, κ-$(ET)_2Cu(SCN)_2$



1. High resolution
2. High intensity
3. Bulk sensitivity
4. Low damage by UV light



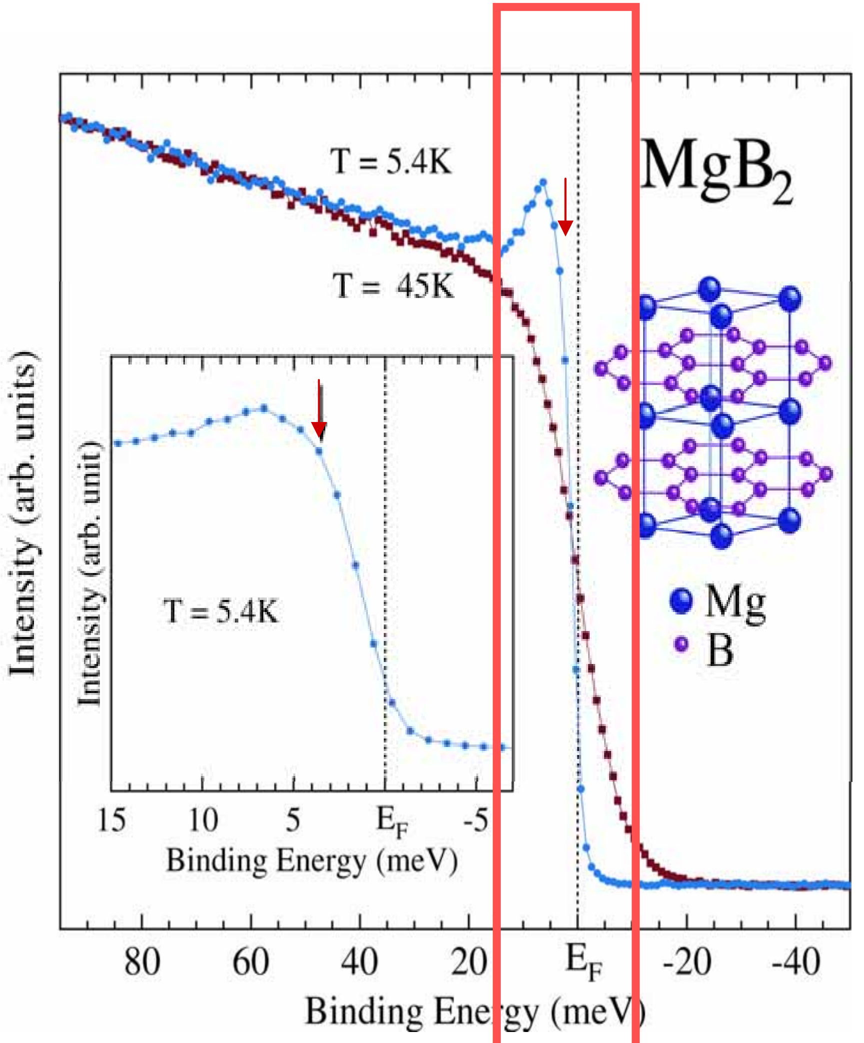
By using strong laser light

- First observation of Fermi edge and superconducting gap.
- Inconsistent with band calculation

⇓

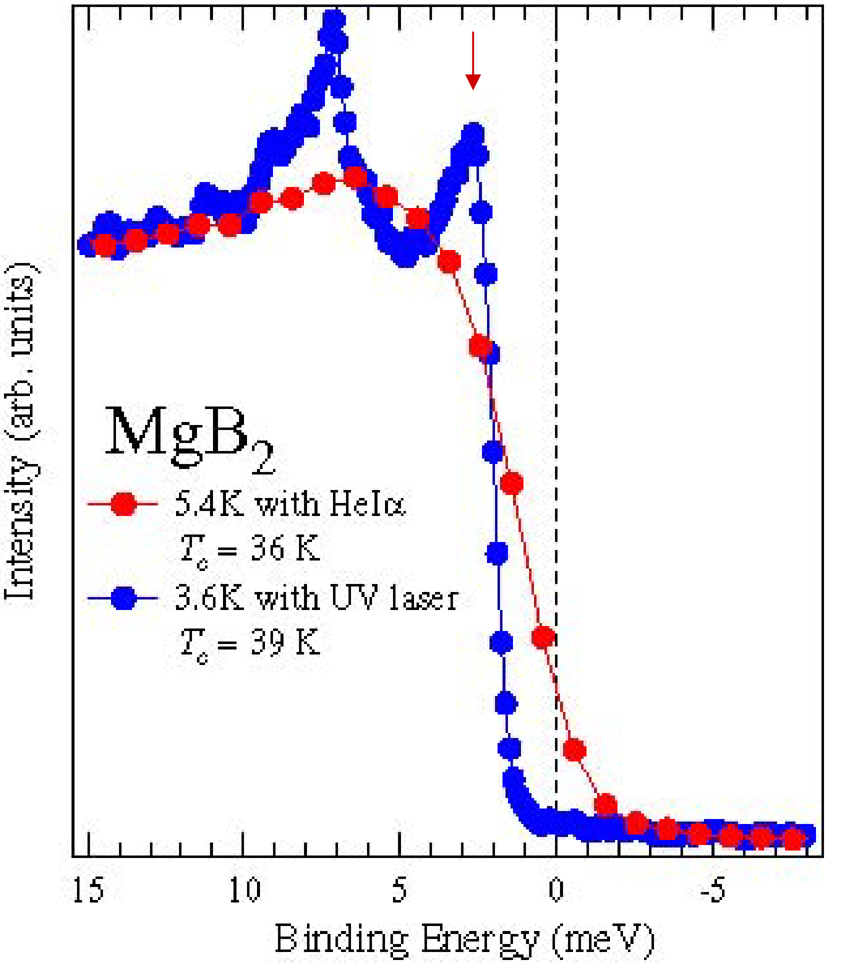
1. Low carrier density
2. Strong correlation effect

“sub meV” resolution spectra on MgB_2 by laser-PES



The spectrum was measured by He lamp and show the two gap structure for the first time

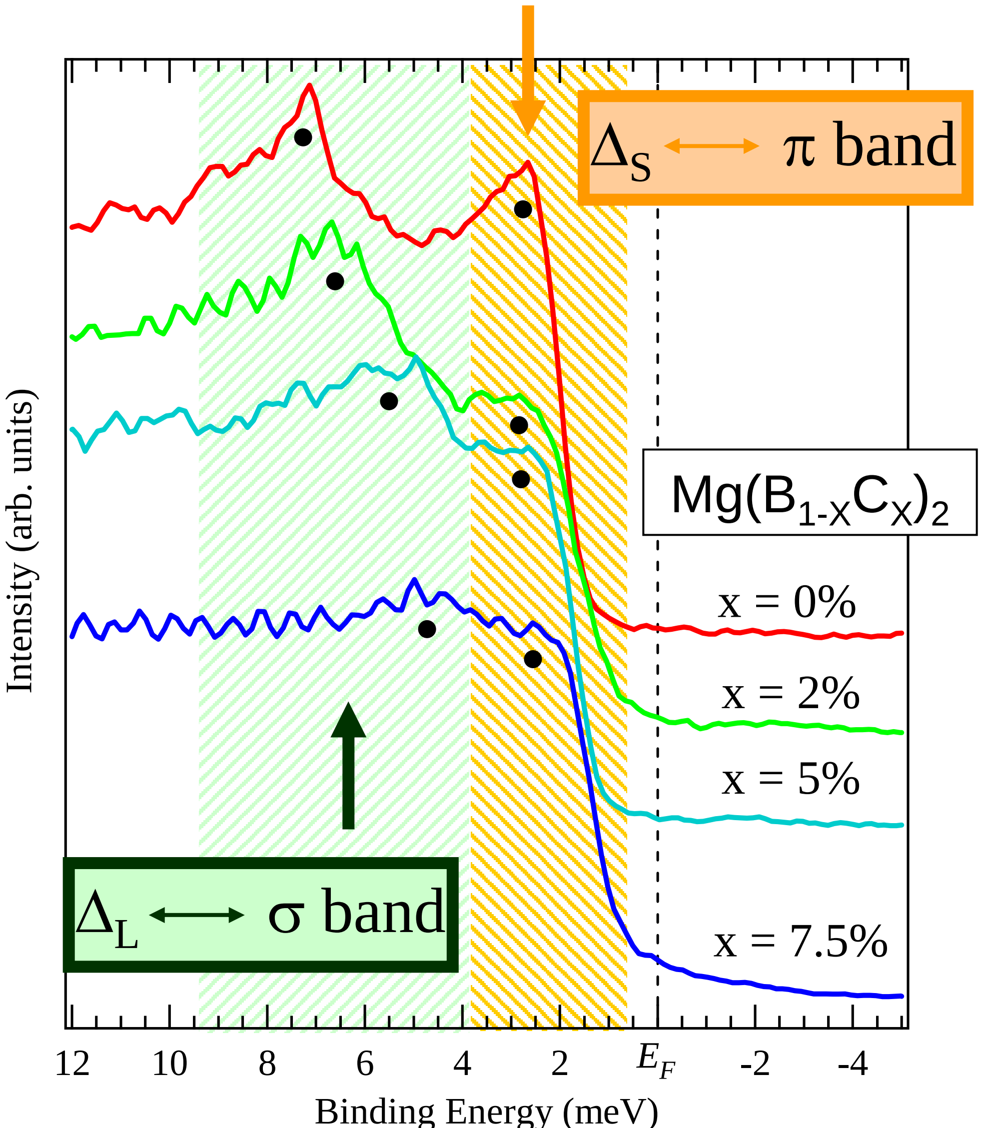
Tsuda et al., PRL**87**,17006(2001)



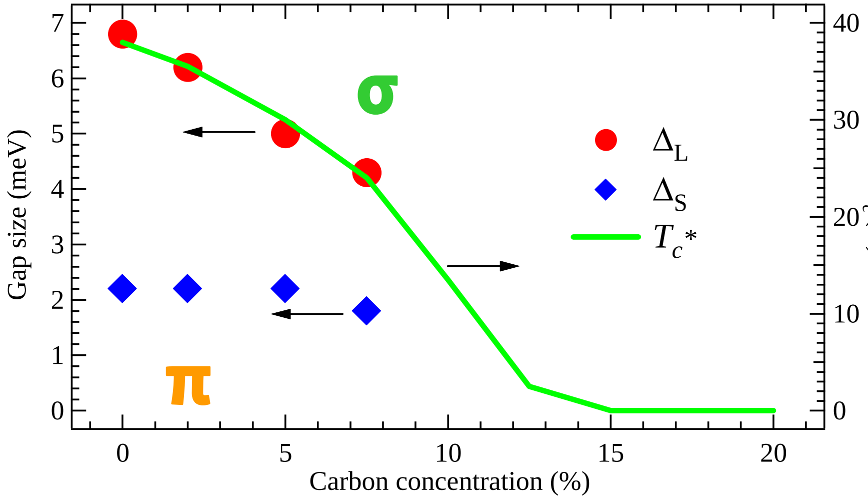
Laser-PES

Tsuda et al.

“sub meV” resolution spectra on MgB_2 by laser-PES
Carbon substitution effect



- Carbon concentration
  strong increase of the interband coupling
- $2\Delta_L/k_BT_c \sim 4.1$
- Tc increase by the interband coupling



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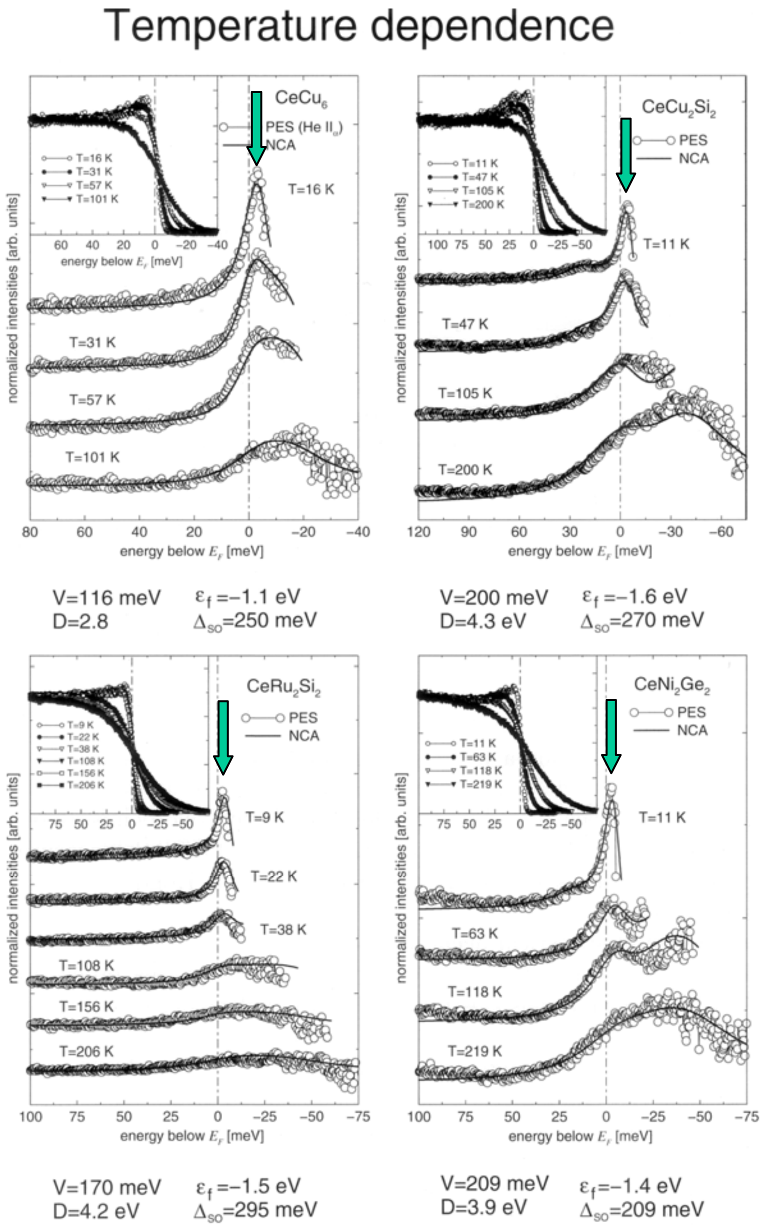
Ultra-high resolution photoemission; Kondo peak



Hüfner, "Photoelectron spectroscopy"

The case of Ce compounds

The Kondo peak becomes sharper and stronger as the temperature decreases

Bulk sensitive spectra using VUV laser ;

Typical coherent and incoherent peaks have been observed in $Sr_xCa_{1-x}VO_3$ PES spectra

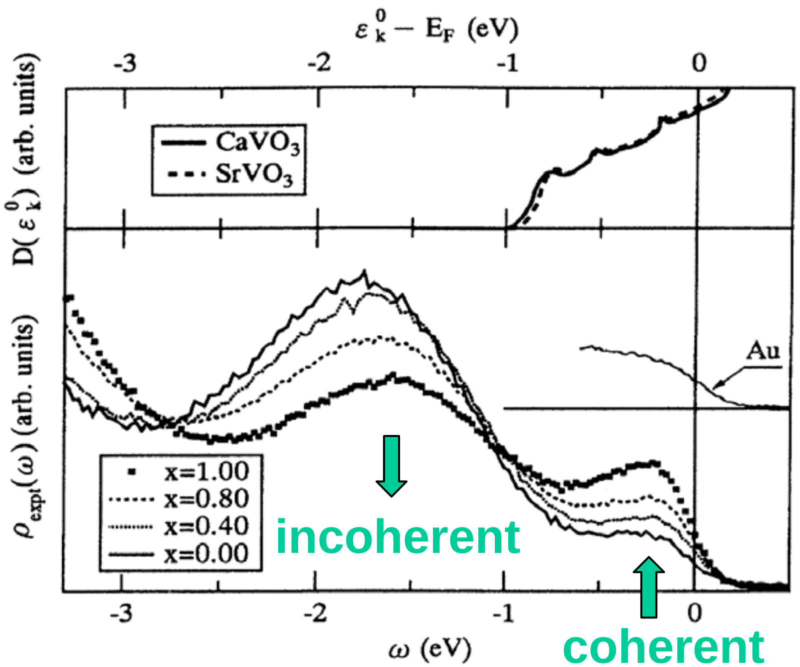
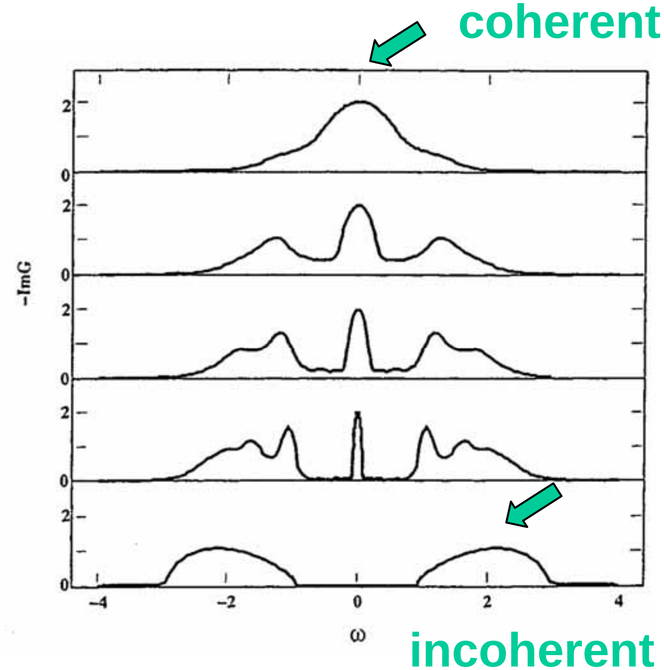


FIG. 1. Top: DOS $D(\varepsilon_k^0)$ of $CaVO_3$ and $SrVO_3$ calculated by the APW method. Bottom: measured photoemission spectra $\rho_{expt}(\omega)$ of $Ca_{1-x}Sr_xVO_3$ taken with $h\nu = 50$ eV. A spectrum of Au is also shown as a reference to E_F and the instrumental resolution.

Inoue et al., PRL74,2539(1995)

DMF theory



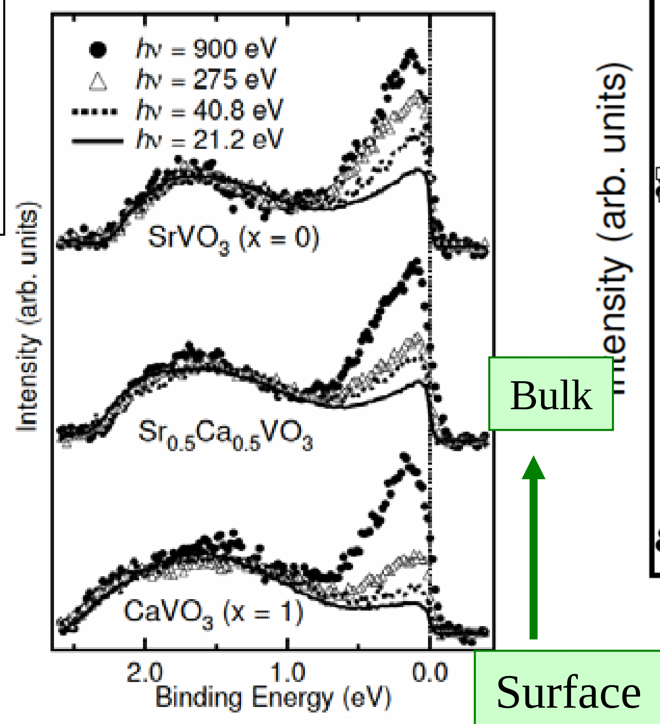
Density of states $\rho(\varpi)$=-Im G by the $d=\infty$ approach at T=0 for the half-filled Hubbard model at U/t^*=1, 2, 2.5, 3, and 4 from top to bottom. The calculation is done by iterative perturbation in terms of U. The bottom one (U/t^*=4) is an insulator.
Georges, Kotliar, Krauth, and Rozenberg, PRL1996.

Bulk sensitive spectra using SX PES

- Incoherent peak becomes weak in SX PES
- $CaVO_3$ and $SrVO_3$ spectra are similar in SX PES

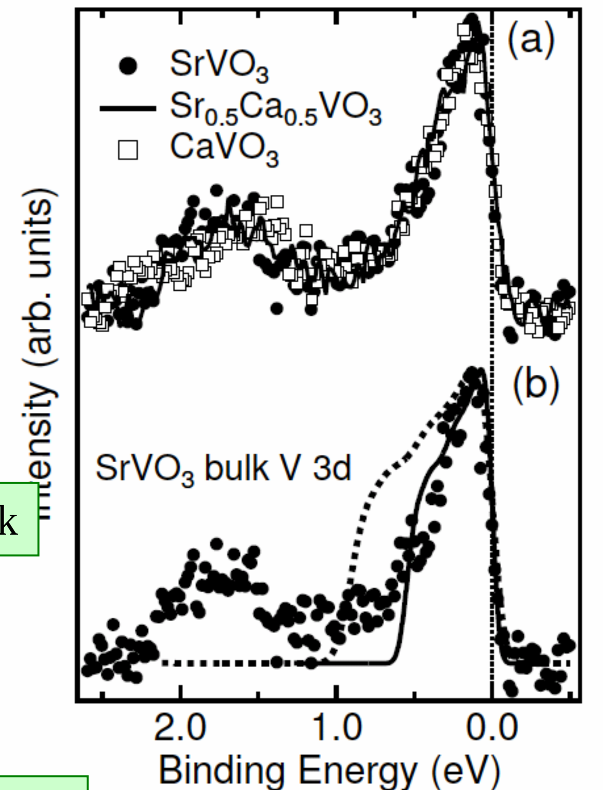
What happens in the bulk sensitive spectra using VUV laser ?

- More bulk sensitive
- much higher resolution



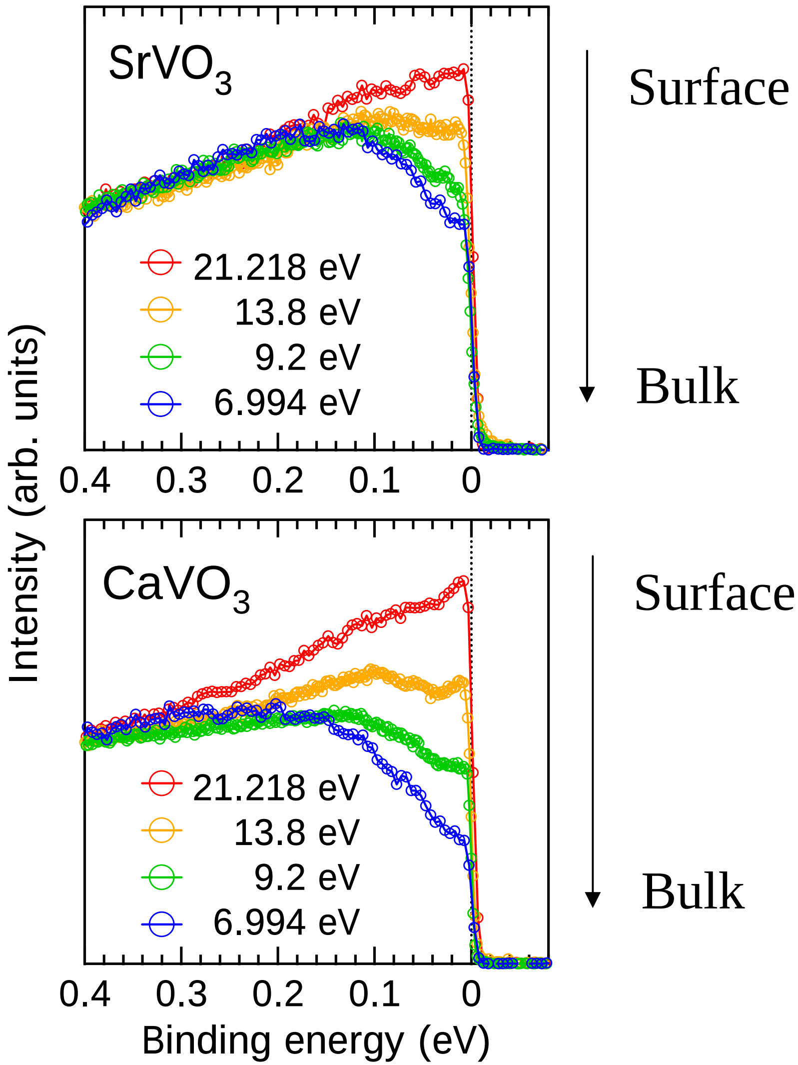
Photon-energy dependence of the V 3d spectral weights for $Sr_{1-x}Ca_xVO_3$. The V 3d spectra are normalized by the integrated intensities of the incoherent part ranging from 0.8 to 2.6 eV.

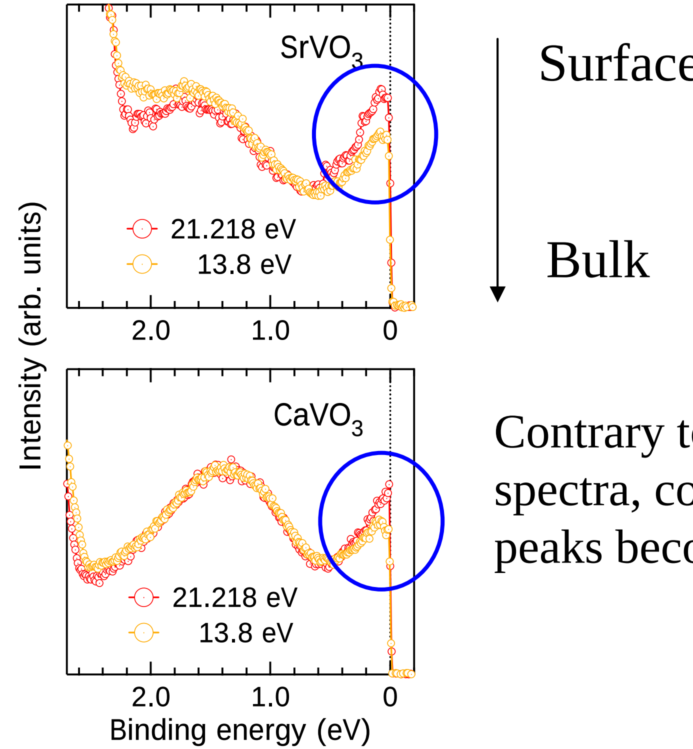
Sekiyama, PRL(2004)



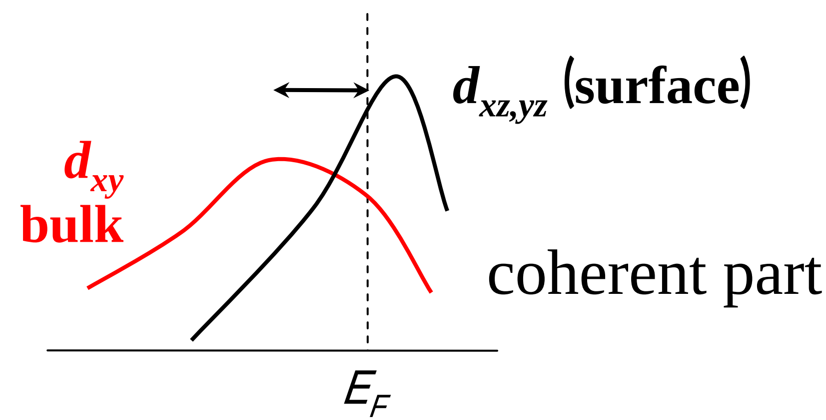
(a) Bulk V 3d spectral functions of $SrVO_3$ (closed circles), $Sr_{0.5}Ca_{0.5}VO_3$ (solid line) and $CaVO_3$ (open squares).
(b) Comparison of the experimentally obtained bulk V 3d spectral function of $SrVO_3$ (closed circles) to the V 3d partial density of states for $SrVO_3$ (dashed curve) obtained from the band-structure calculation, which has been broadened by the experimental resolution of 140 meV. The solid curve shows the same V 3d partial density of states but the energy is scaled down by a factor of 0.6.

Excitation energy dependence of coherence peak

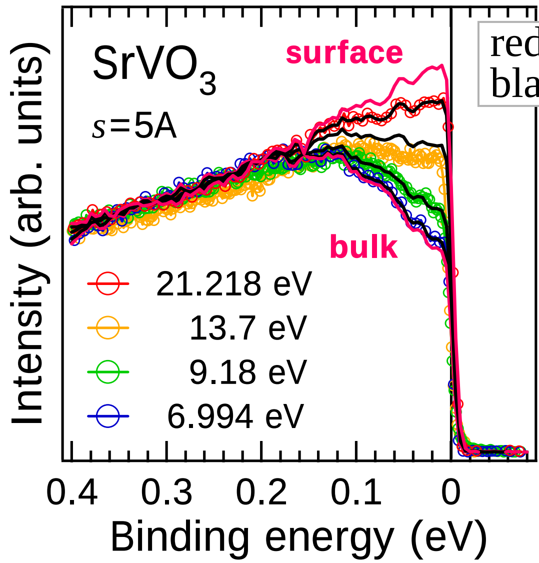




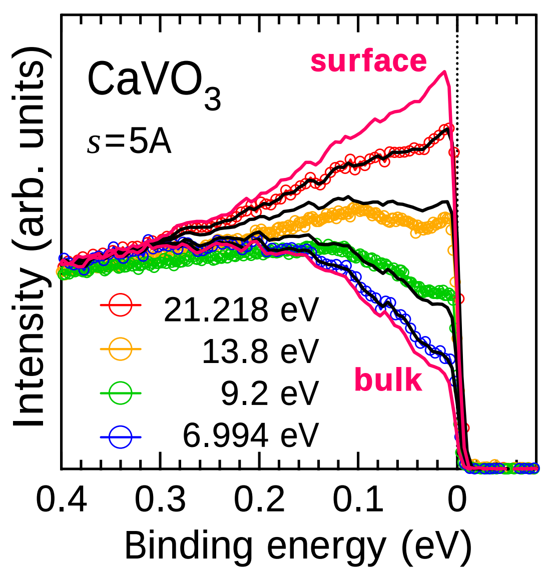
Contrary to the SX spectra, coherent peaks become weaker



The temperature dependence is well reproduced by the Surface and Bulk spectra



red line: $I_{surface}(E)$ and $I_{bulk}(E)$
black line: $I(E)$



$$I(E)=exp(-s/\lambda)\ I_{bulk}(E)+[1-exp(-s/\lambda)]\ I_{surface}(E)$$

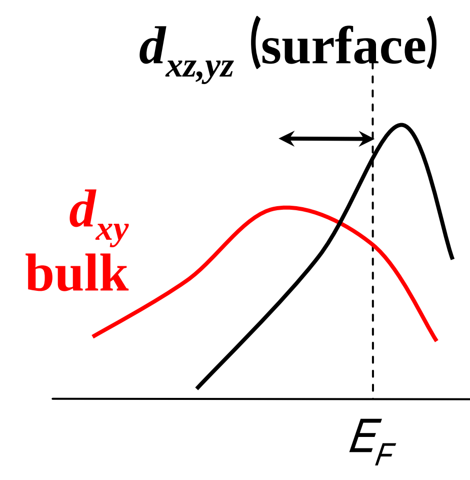
s : thickness of surface layer

λ : mean free path

parameters

s : 5 Å

E(eV)	λ(Å)
21.218	3
13.8	6
9.2	20
6.994	100



coherent part

Summary

- Development of Quasi CW VUV-laser ;

  Highest hν of 7eV with 0.26meV line width

- Development of analyzer with high energy resolution at low temperature ;

  Highest energy resolution of 0.36 meV, Lowest temperature of 2.7K

- Laser PES is powerful for the study on low Tc superconductors and strongly correlated materials

  High resolution PES and Bulk sensitive Fermiology

  - $CeRu_2$; 4f electron system, gap anisotropy
  - MgB_2 ; multigap, interband interactions
  - κ-$(ET)_2Cu(SCN)_2$; d-wave superconductor
  - LiV_2O_4 ; Kondo state in TMO
  - $SrVO_3$; comparison with DMFT(Surface and bulk)

Laser-PES has solved the weaknesses of PES

Low resolution	→	High resolution
Surface sensitive	→	Bulk sensitive
High temperature	→	Low temperature
Large spot size	→	Small spot size