高分解能広立体角2次元光電子顕微分光器（DELMA)の開発

松田博之[a], László Tóth[b], 後藤謙太郎[a], 松井文彦[a], 松下智裕[c] , 橋本美絵[a], 酒井智香子[a], 北川　哲[a]、森田　誠[a]、大門　寛[a]

[a]奈良先端科学技術大学院大学 (NAIST), [b]Debrecen大学, [c]JASRI-SPring-8

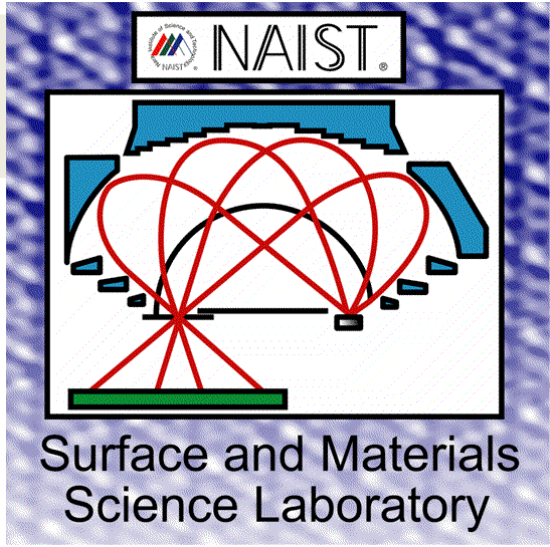
松田巌、山本達（ISSP)

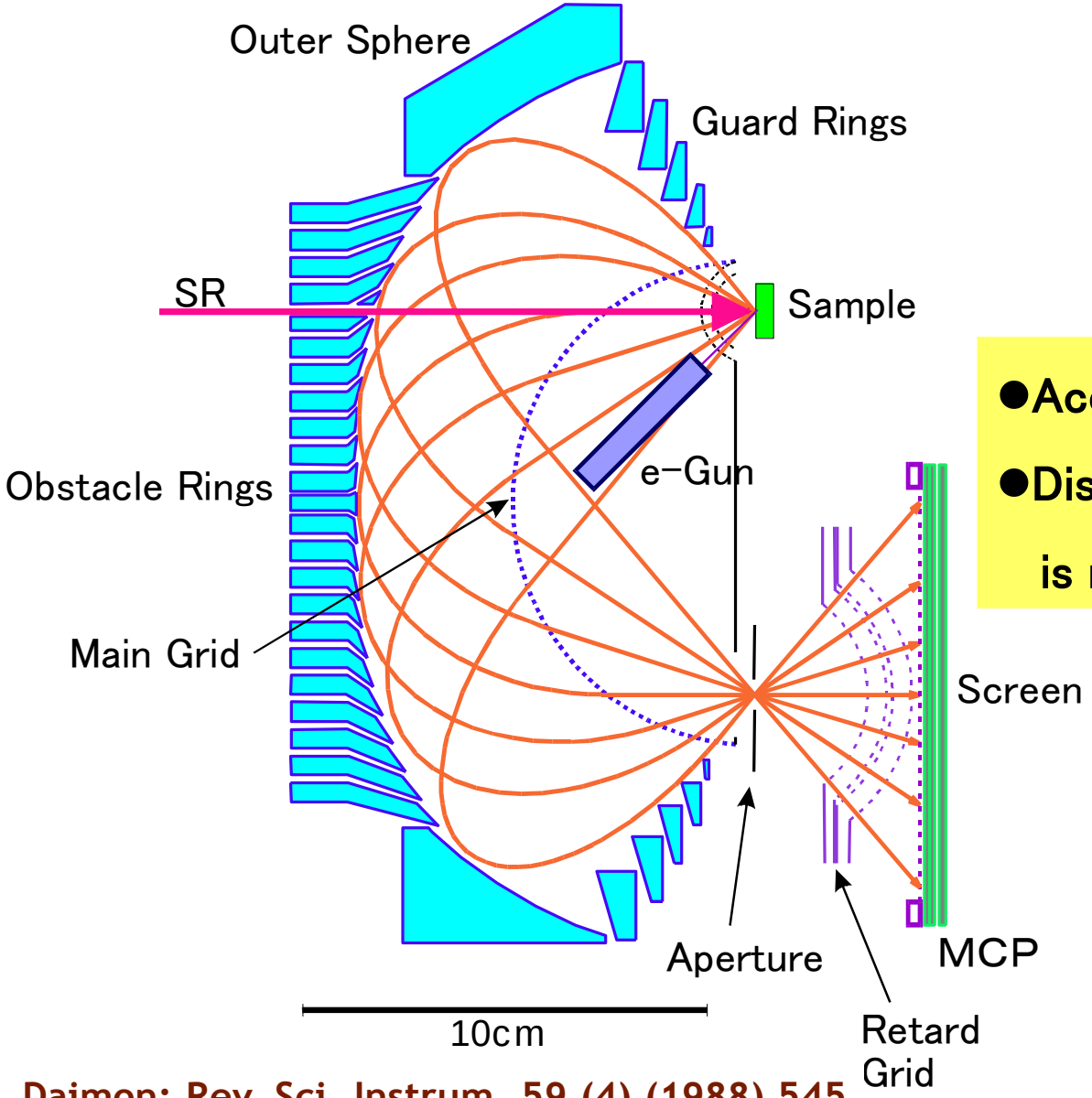
Outline

- Conventional Two-dimensional photoelectron spectrometer (DIANA)
- New Photoelectron Emission Microscope for Stereophotography (DELMA)
- Application to graphene

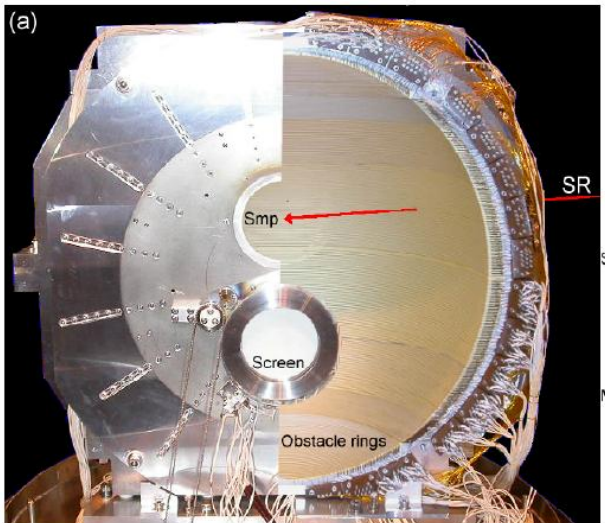
科研費　基盤(S) (20224007）

Two-dimensional photoelectron spectrometer
Display-type Spherical Mirror Analyzer (DIANA)





- Acceptance angle ±60°
- Displayed angular distribution is not distorted



H. Daimon: Rev. Sci. Instrum. 59 (4) (1988) 545.

Rits BL-7: LP-VUV 2D-ARPES Beam line

BL-7(Rits-SR center):

35 ~ 130 eV

Linearly Polarized light

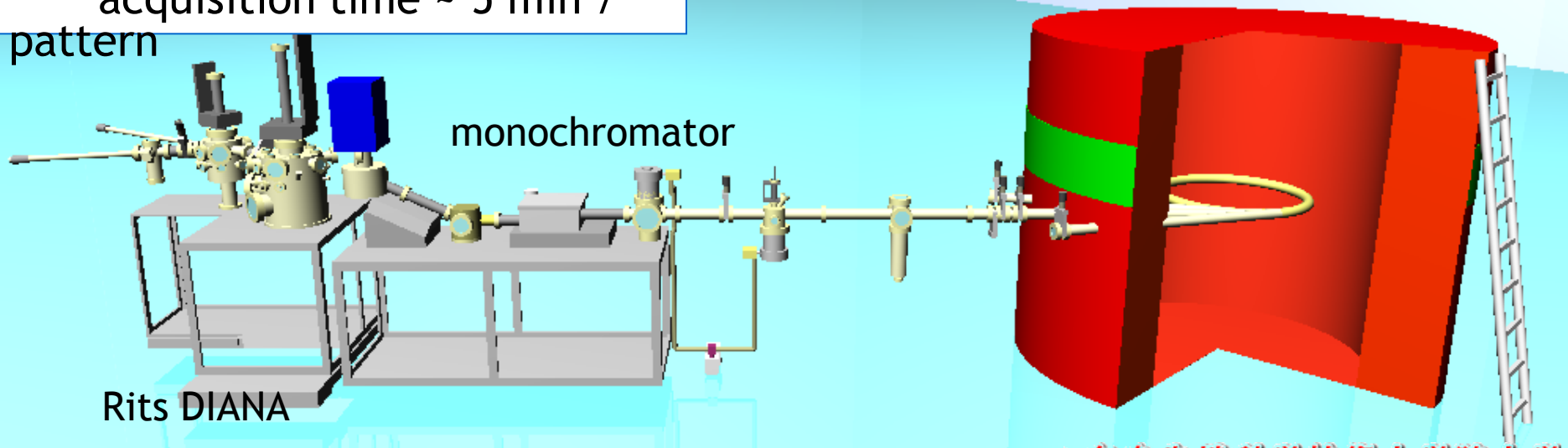
Analyzer:

total resolution ΔE / E ~ 1%

angular resolution ±0.5°

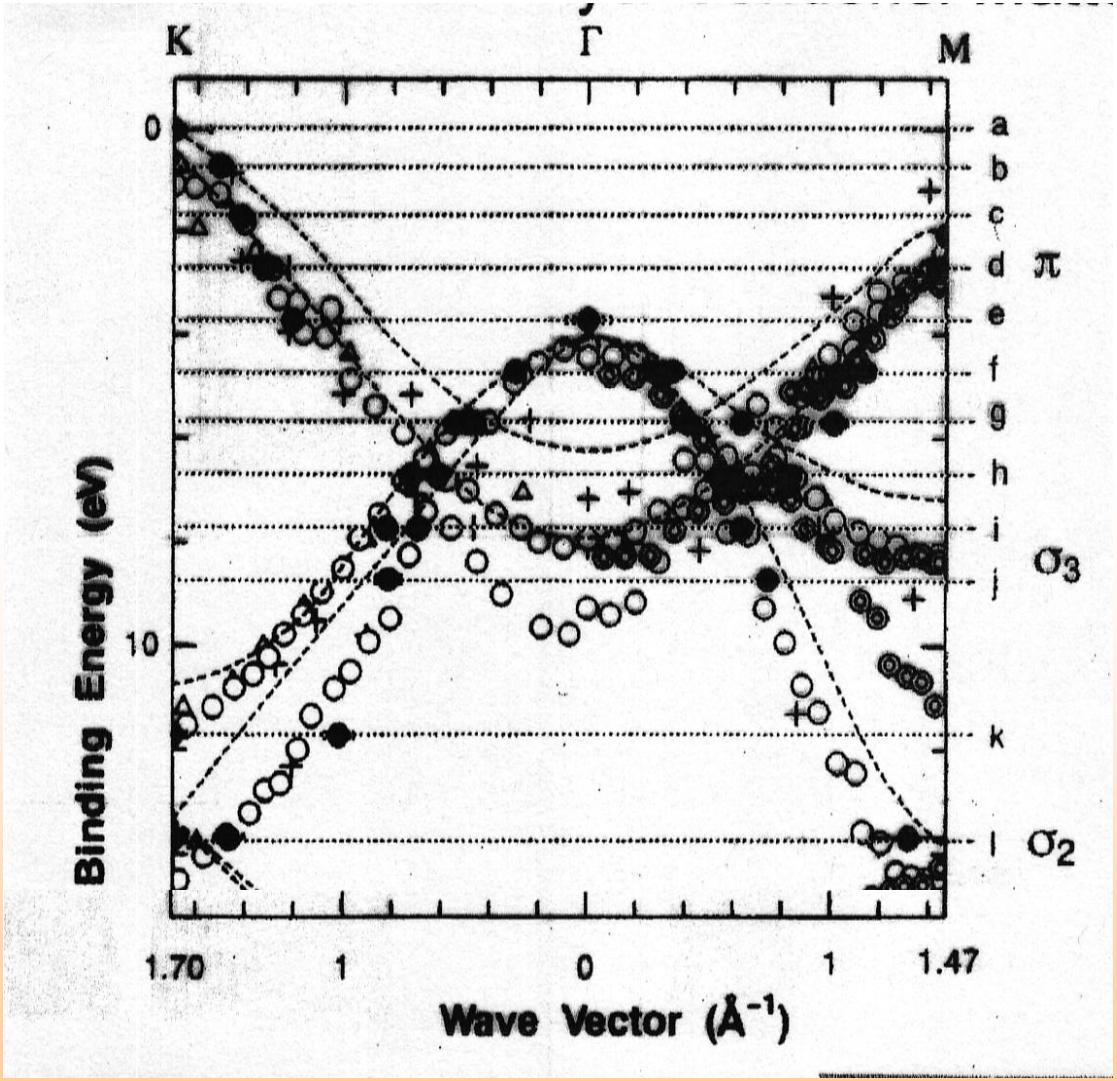
acceptance angle ±50°

acquisition time ~ 5 min / pattern

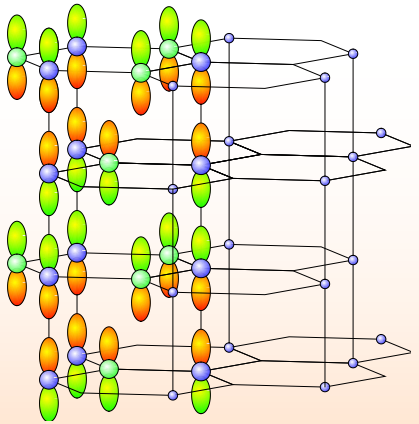


奈良先端科学技術大学院大学

π band dispersion of graphite in k_z direction

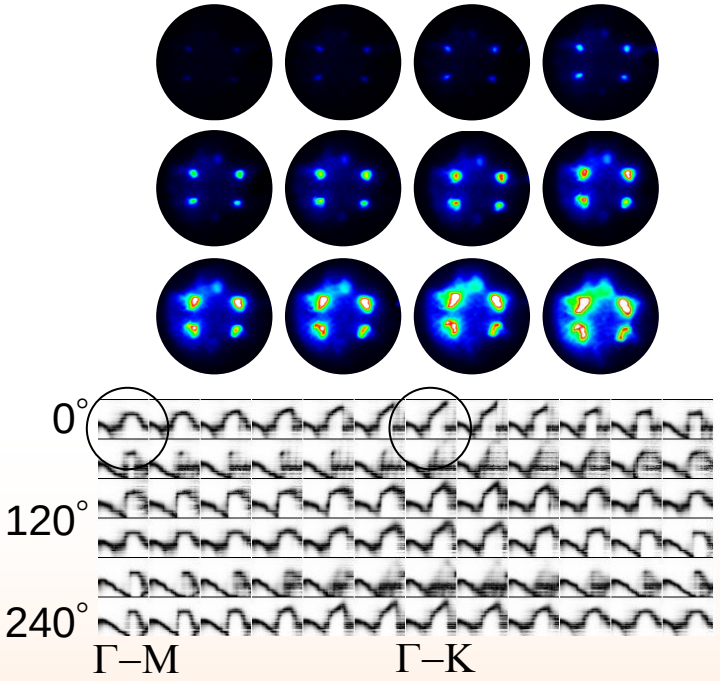


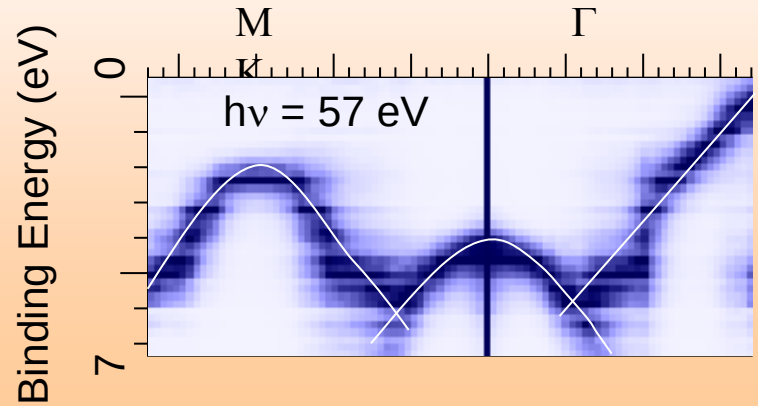
π bonding band

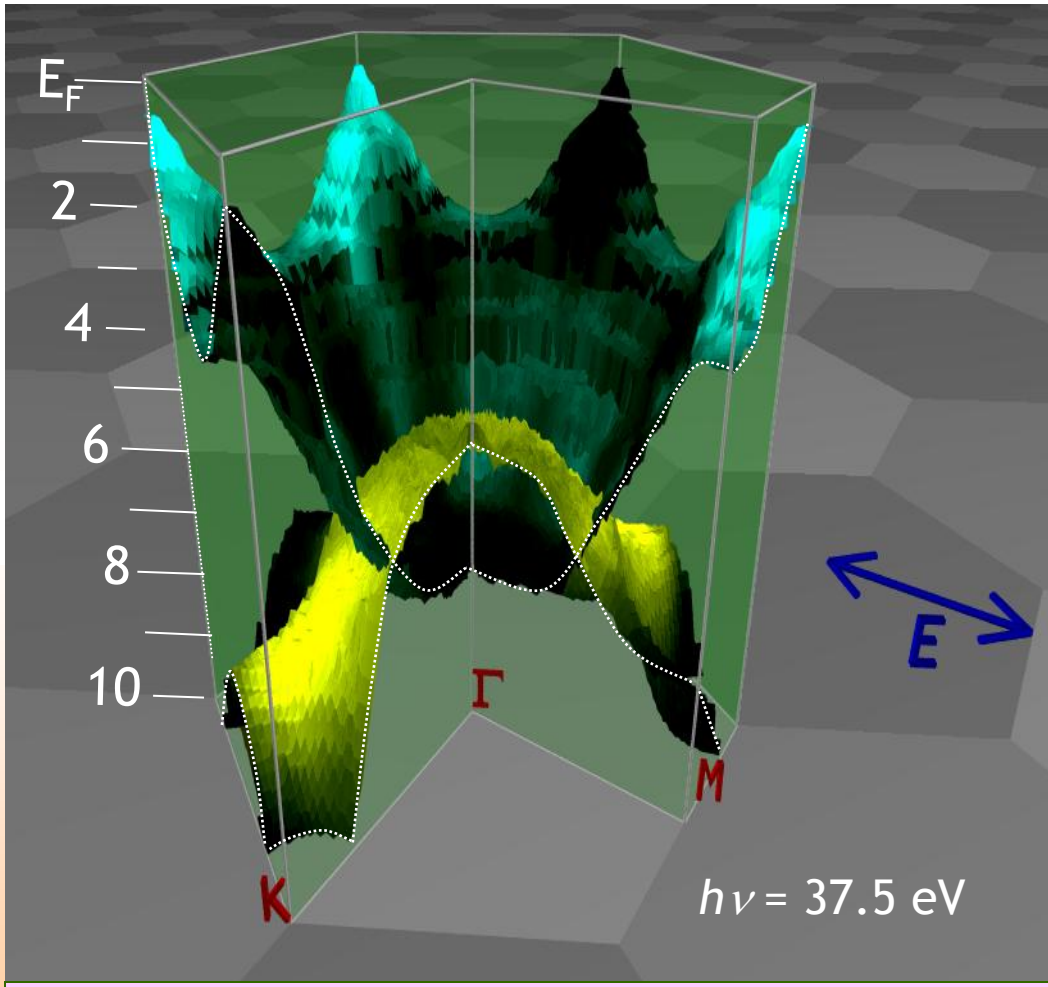


Three Dimensional Valence Band Dispersion

F. Matsui, Y. Hori, H. Miyatake, N. Suganuma, H. Daimon, H. Totsuka, K. Ogawa, T. Furukubo, H. Namba: Appl. Phys. Lett. **81** (2002) 2556.

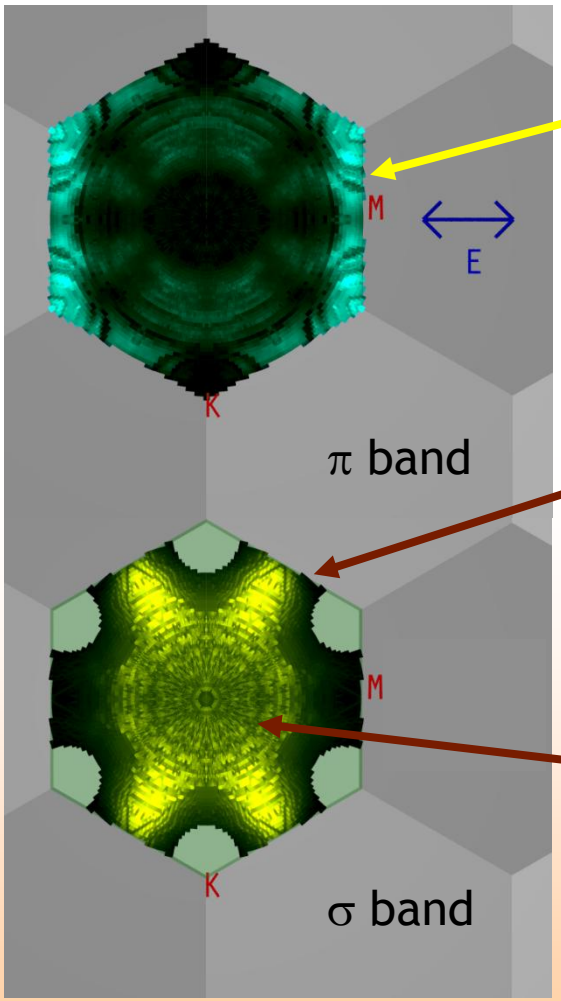


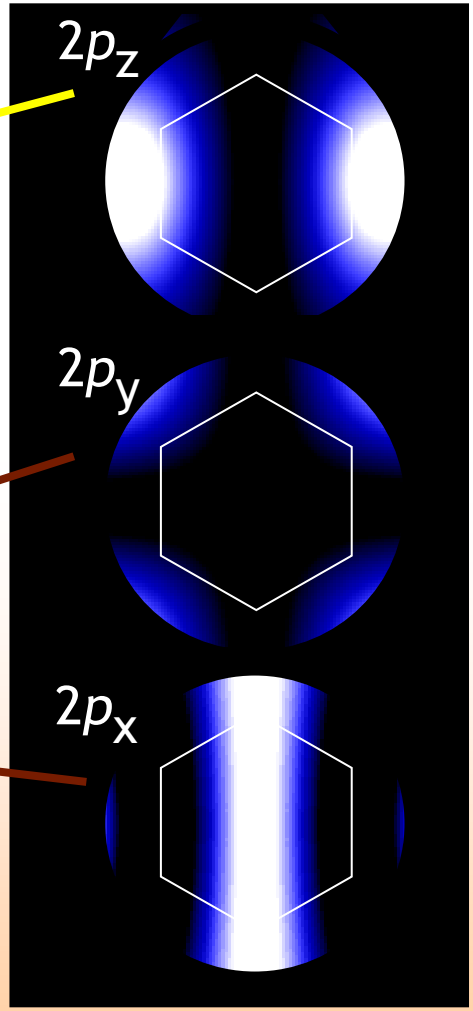




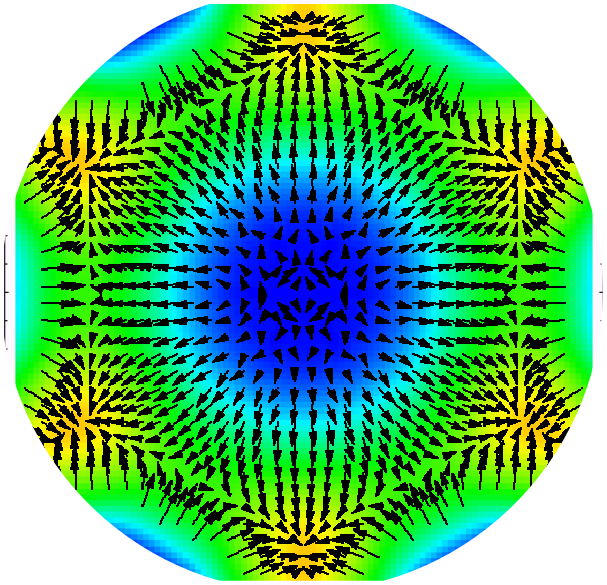
3D structure of graphite valence band

Valence band dispersion of graphite

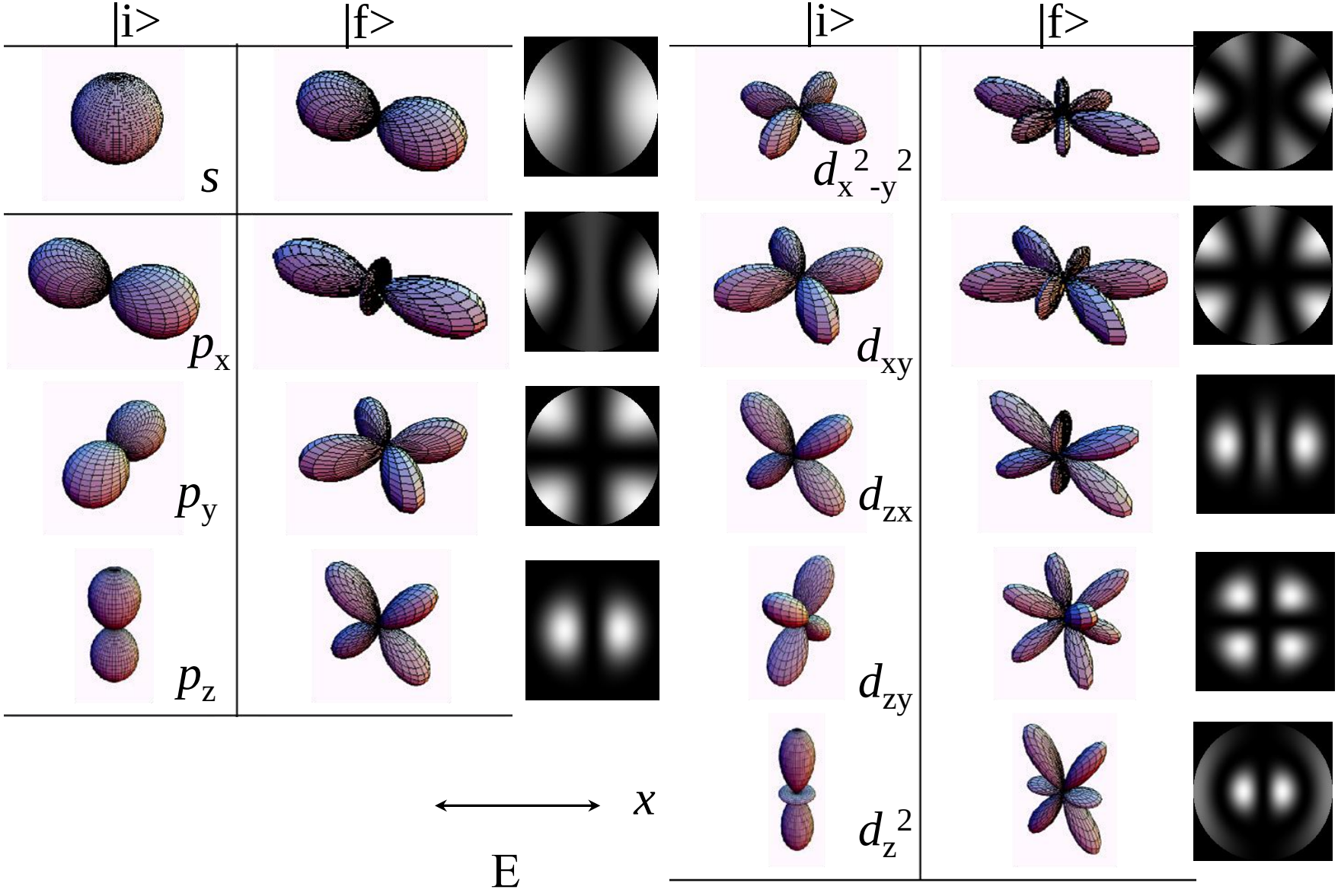




F. Matsui, et al., Appl. Phys. Lett. **81** (2002) 2556.

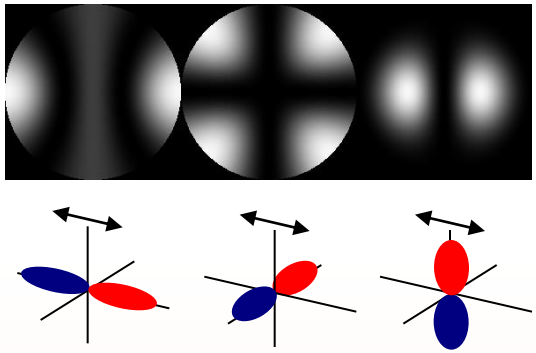


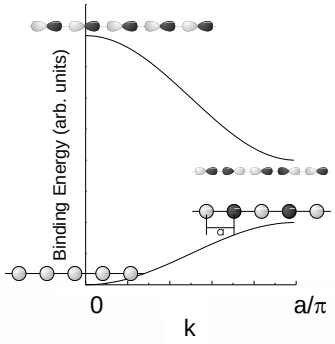
Differential of π band:
Group velocity of electrons
∝differential of band dispersion
$\mathbf{v}_g = 1/\hbar(d\varepsilon(\mathbf{k})/d\mathbf{k})$

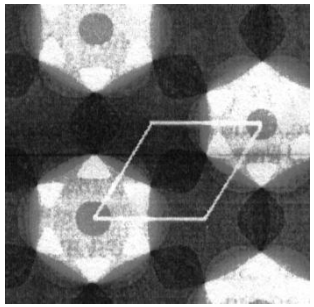

|i>
|f>
|i>
|f>
s
p_x
p_y
p_z
$d_{x^2-y^2}$
d_{xy}
d_{zx}
d_{zy}
d_{z^2}
x
E

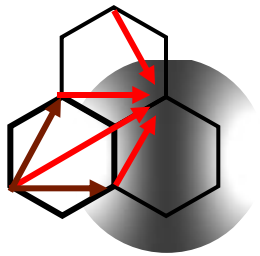
4 factors in photoelectron angular distribution

Angular distribution from atomic orbital · Dispersion of valence bands · Structure factor of photoelectron · Final state effects

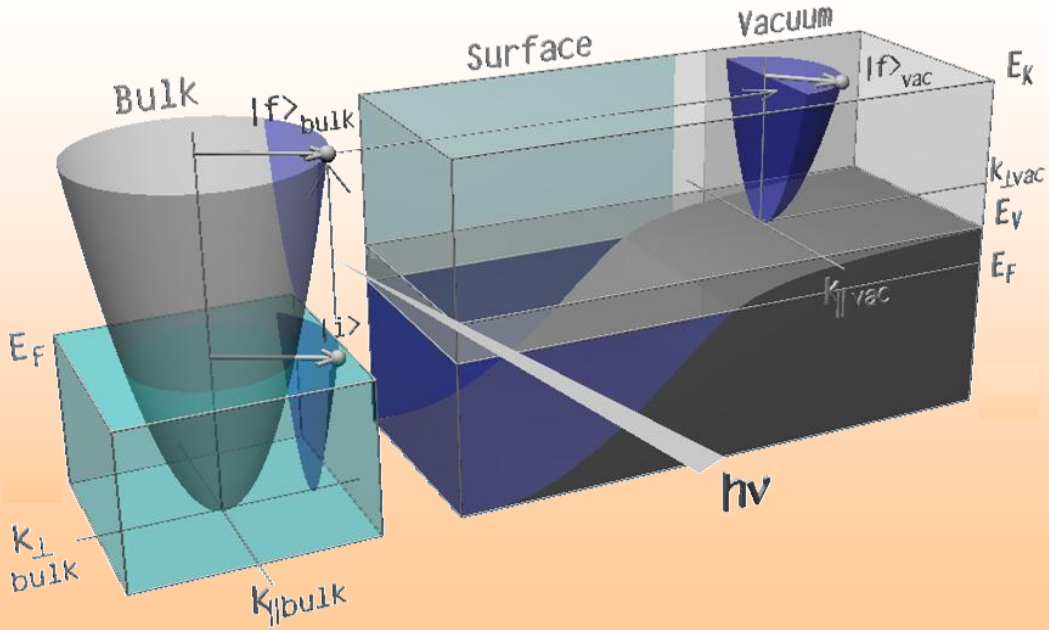








Umklapp process etc.



Binding energy scan:

valence band dispersion

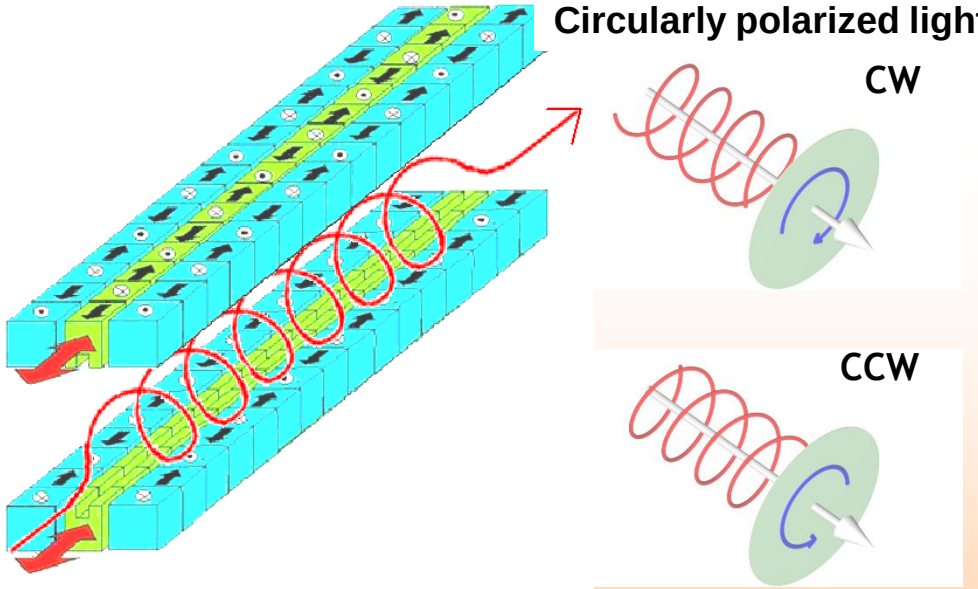
Photon energy scan:

k_z variation of isoenergy surface

circularly polarized light synchrotron radiation

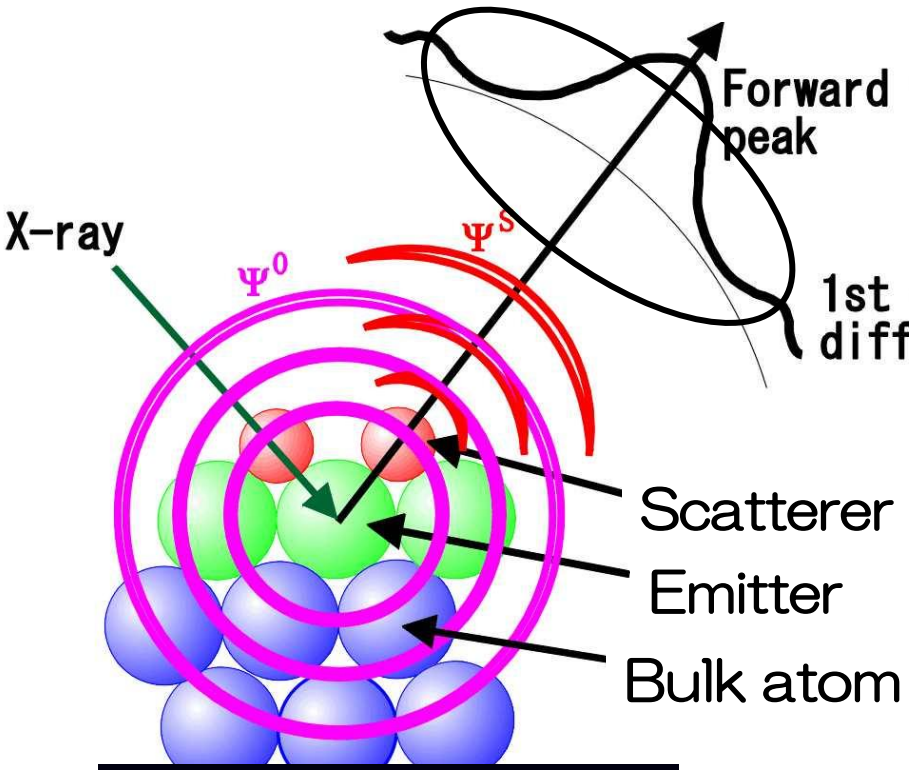
BL25SU (SPring-8) with Twin helical undulators

(hν = 500 -1500 eV)



SPring-8: the largest SR ring in the world.

Forward focusing peak (FFP) in Photoelectron Intensity Angular Distribution (PIAD)



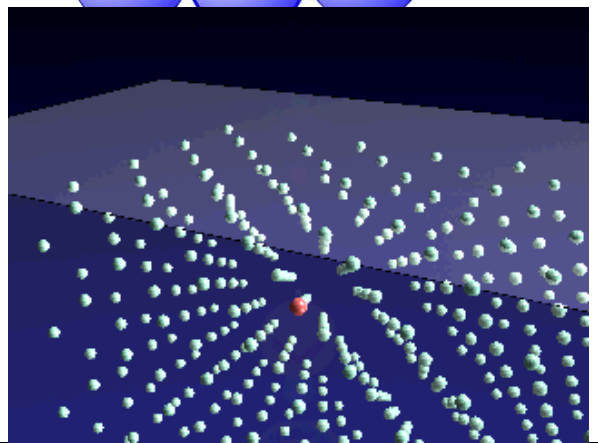
Real space, direct, direction

projected atomic image around emitter

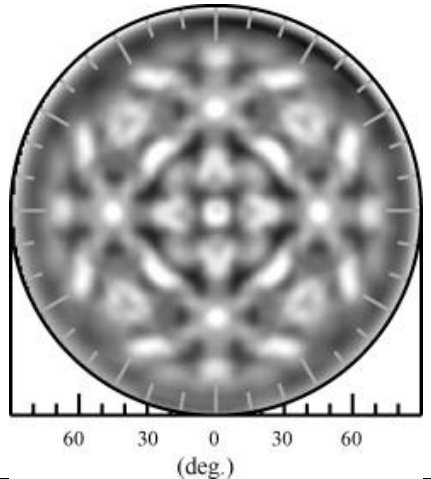
Circularly polarized light ⇒ distance
Stereo-photograph

Photoelectron Diffraction
Photoelectron Holography

Reciprocal space, calculate 3D structure
indirect



Example of 2D-PIAD
Si2p from Si(001)



Initial *m* increases or decreases by 1 for ccw or cw light excited photoelectrons

ccw
$m = m_i + 1$

cw
$m = m_i - 1$

s : $l = 0$

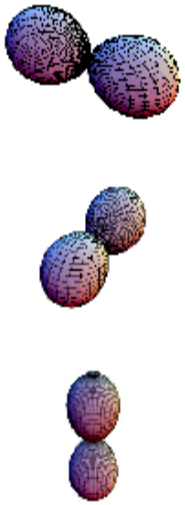
Initial

$m_i = 0$

photoelectrons

ccw
$m = 1$

p : $l = 1$



Initial

$m_i = \pm 1$

$m_i = 0$

photoelectrons

ccw
$m = 2$

$m = 1$

$m = 0$

d : $l = 2$

Initial

$m_i = \pm 2$

$m_i = \pm 1$

$m_i = 0$

photoelectrons

ccw
$m = 3$

$m = 2$

$m = 1$

$m = 0$

$m = -1$

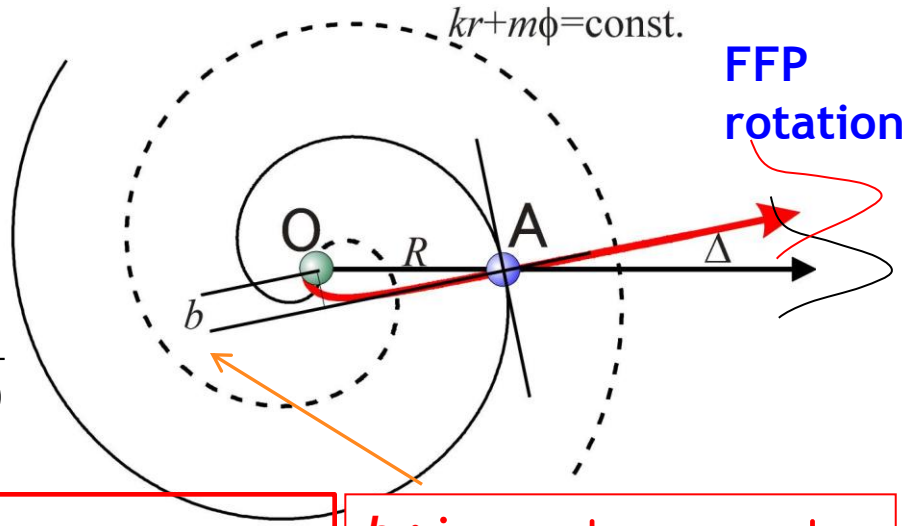
Rotation of the forward direction of the wave by orbital angular momentum

$$\Psi \propto \frac{e^{ikr}}{r} Y_{lm}(\theta,\phi)$$

$$\propto \frac{e^{ikr}}{r} \Theta_{lm}(\theta) e^{im\phi}$$

Phase $\exp[i(kr+m\,\phi)]$

Rotation $\Delta = \tan^{-1}\dfrac{m}{kR\sin^2\theta} \cong \dfrac{m}{kR\sin^2\theta}$



Classical Angular momentum

(angular momentum)=(momentum)×(impact parameter)

***b* : impact parameter is not zero if *m* >< 0**

$$m\hbar = \hbar k \times b$$

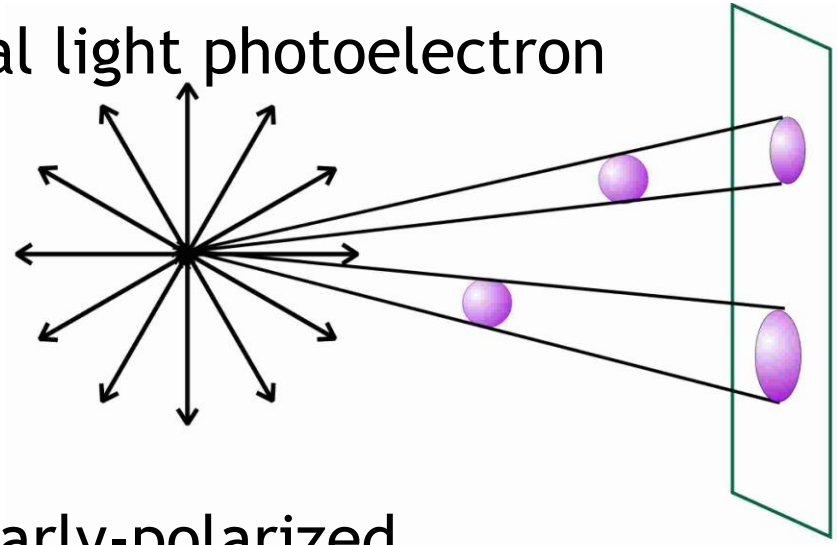
$$\Delta_{//} = \sin^{-1}\frac{b}{R_{//}} = \sin^{-1}\frac{m}{kR_{//}}$$

$$\rightarrow \quad \Delta = \sin^{-1}\frac{m}{kR\sin^2\theta}$$

H. Daimon, T. Nakatani, S. Imada, S. Suga, Y. Kagoshima, and T. Miyahara, Jpn. J. Appl. Phys. **32** Part 2, (1993) L1480.

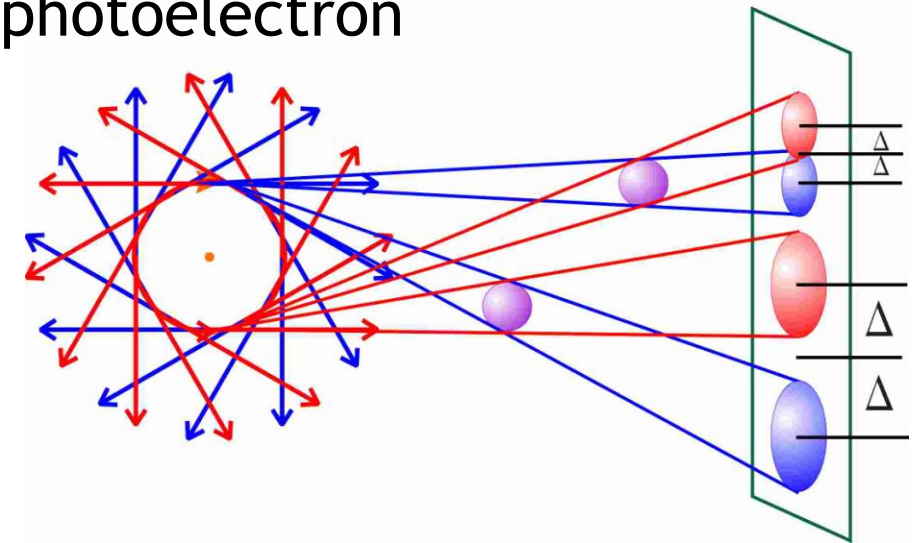
Direction of FFP for normal and circularly-polarized light

Normal light photoelectron



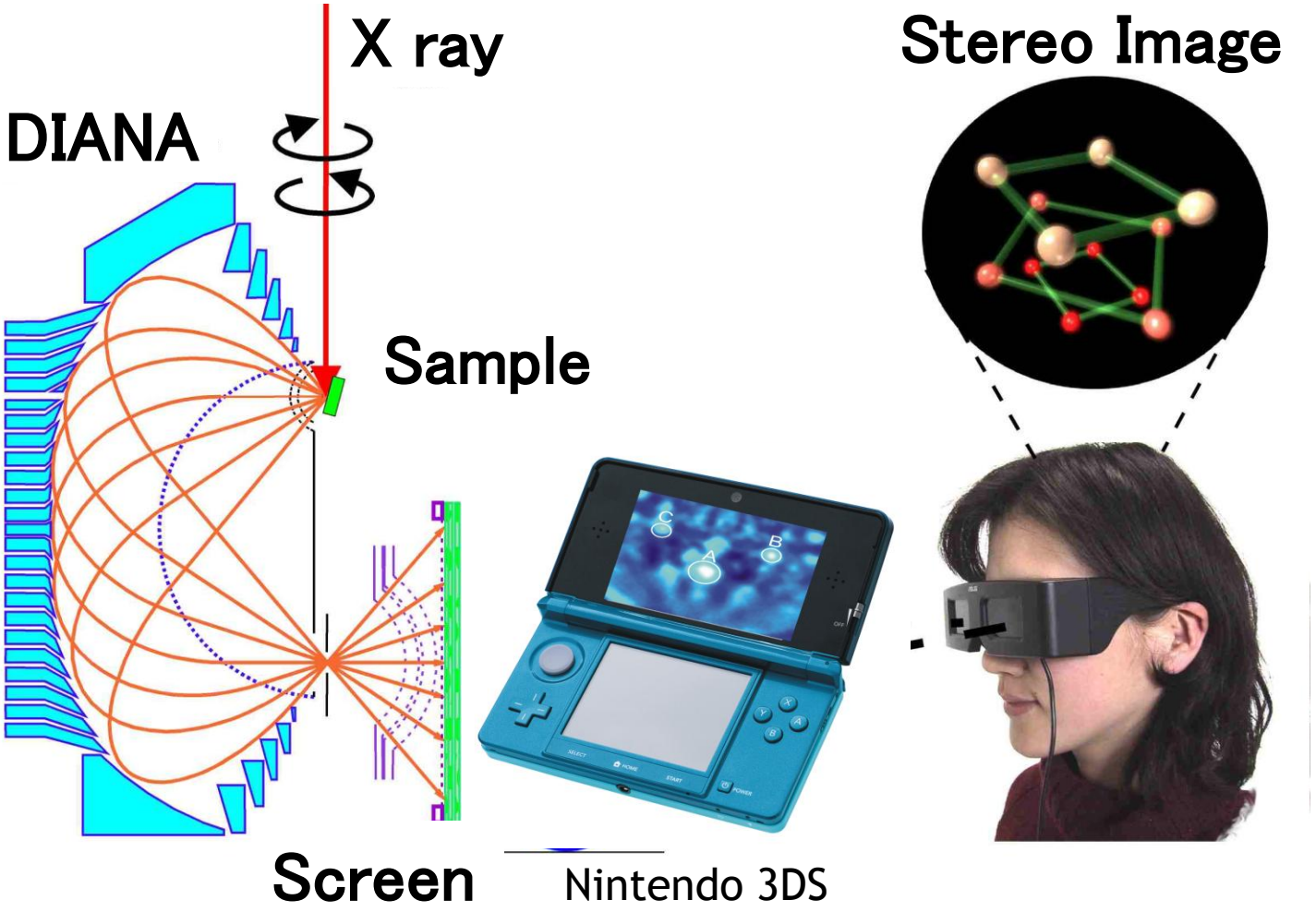
Projected atomic image on the screen

Circularly-polarized light photoelectron



Parallax of Atomic image on the screen

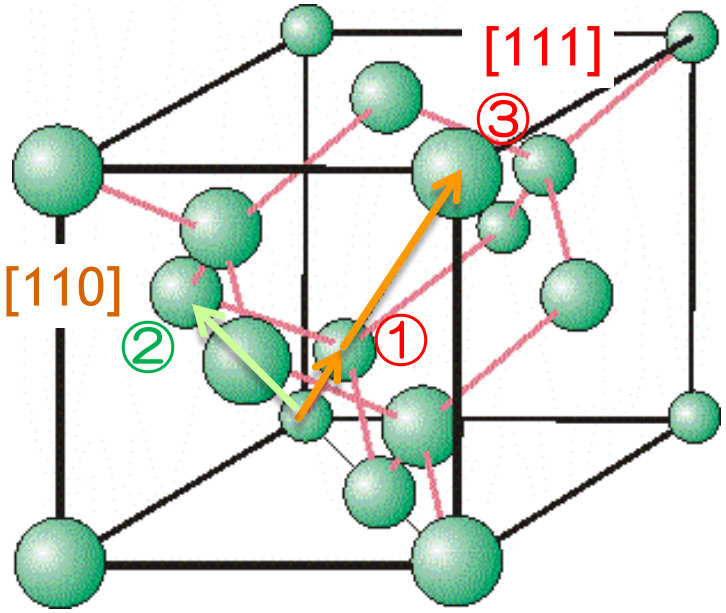
Real-time direct observation of non-periodic 3D atomic arrangement

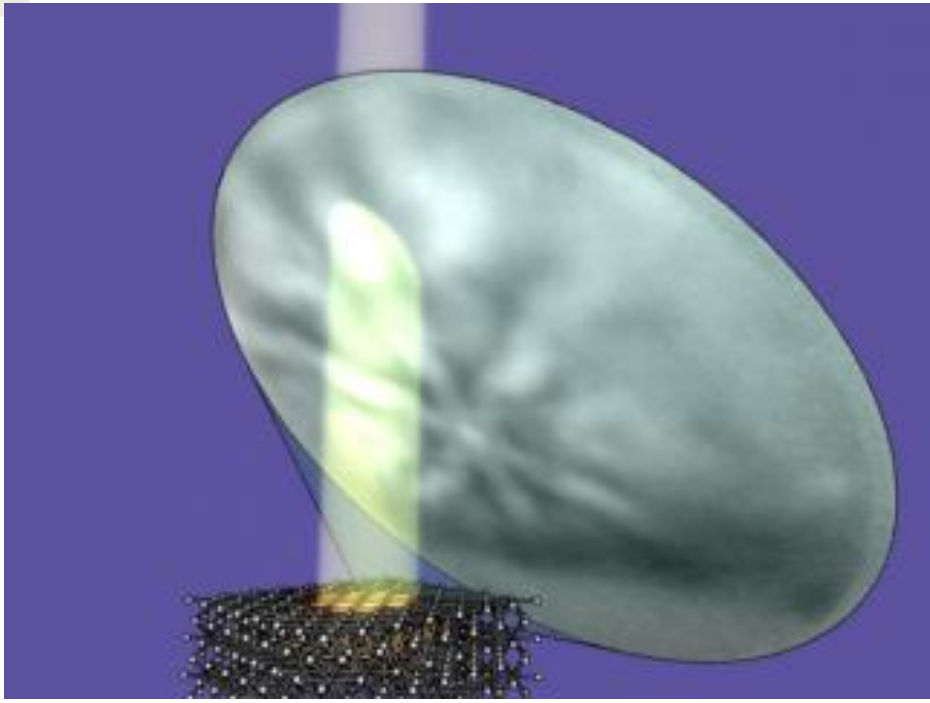


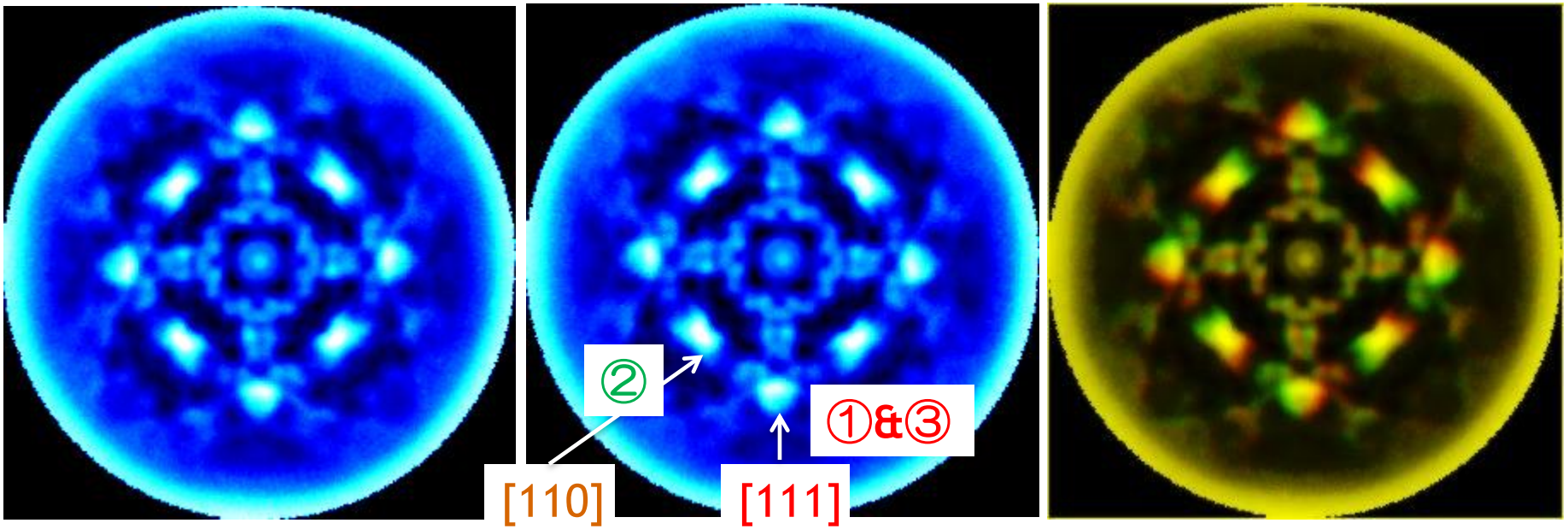
H. Daimon, Phys. Rev. Lett. 86, (2001) 2034

2π-steradian stereo photograph

Si(001)

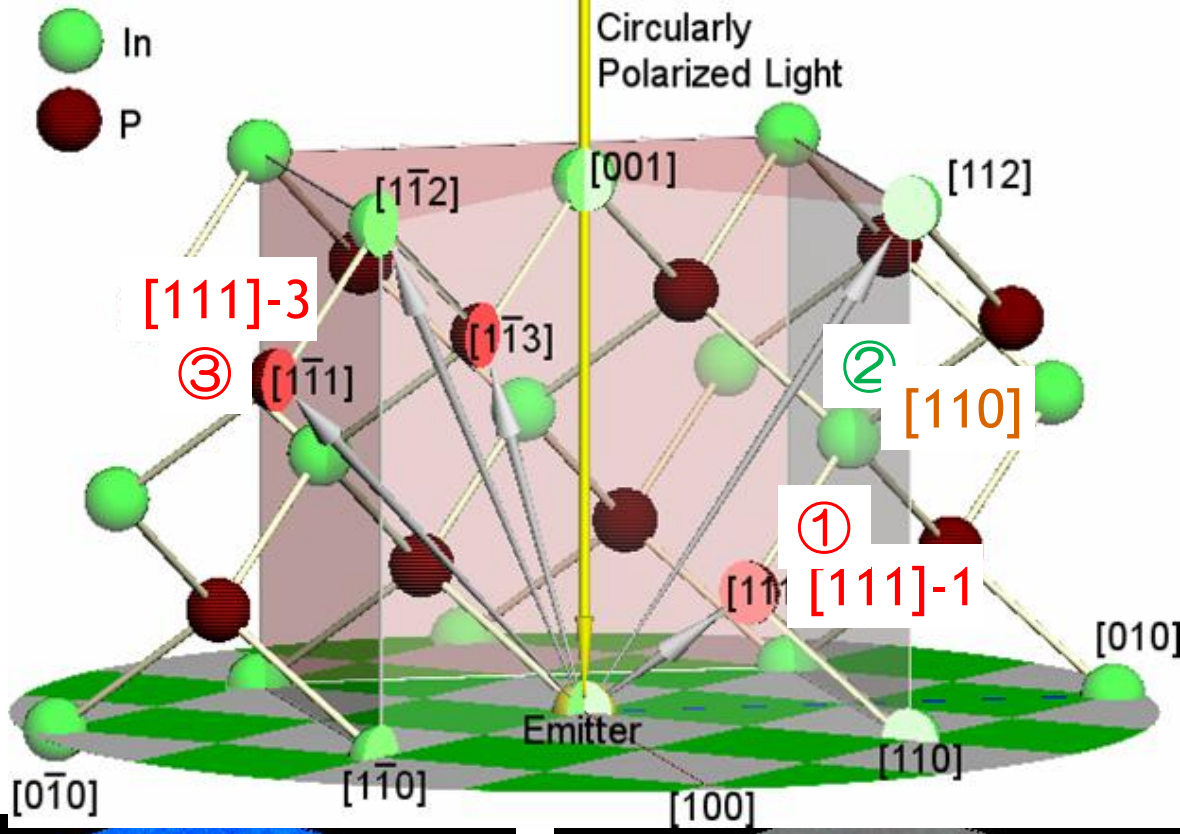


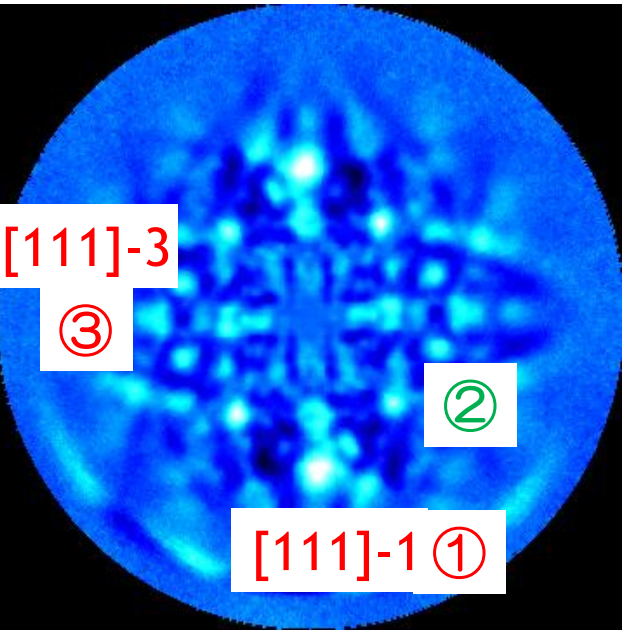


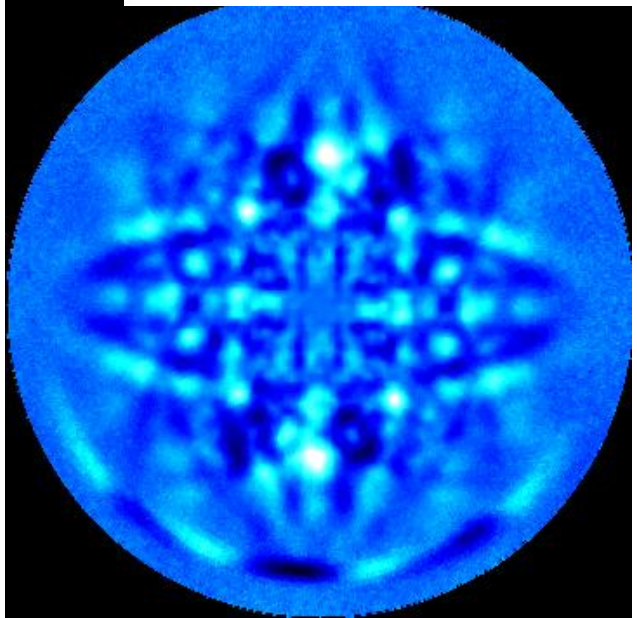


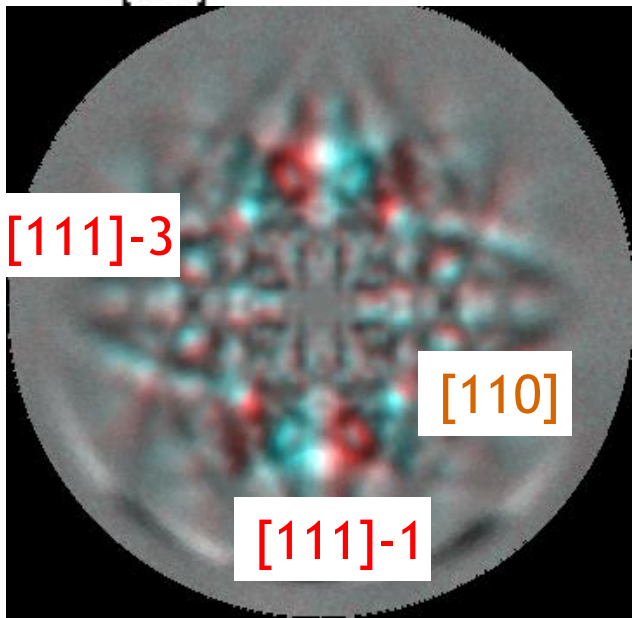
2π-steradian stereo photograph

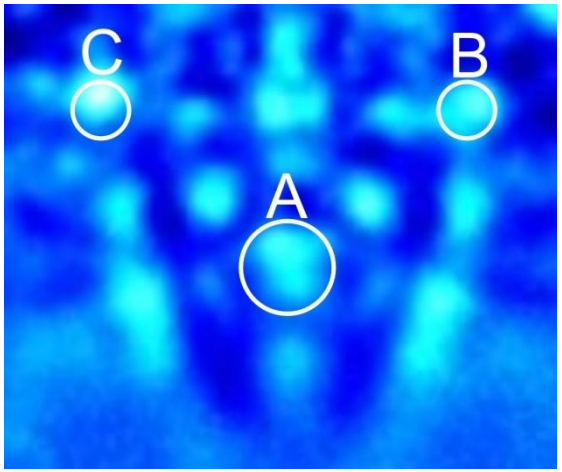
In3d from InP(001)

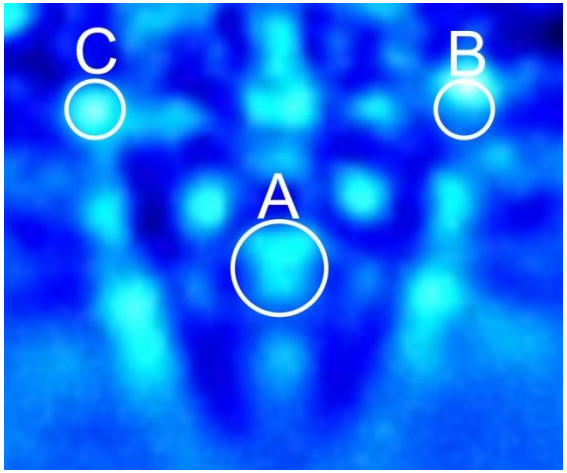




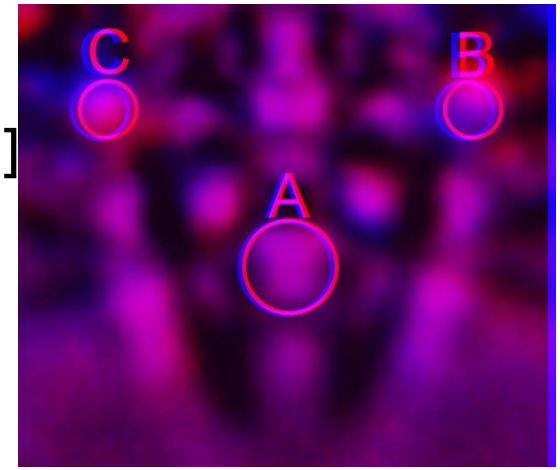


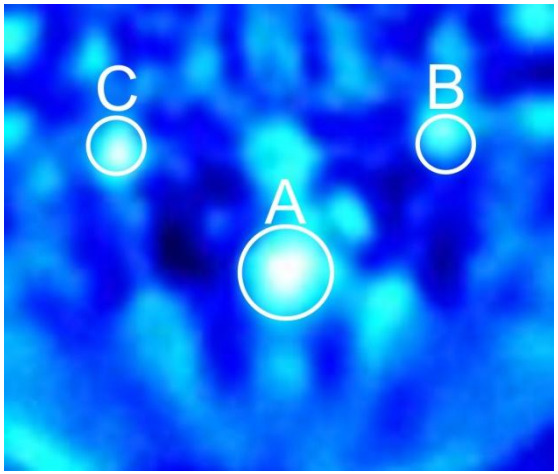


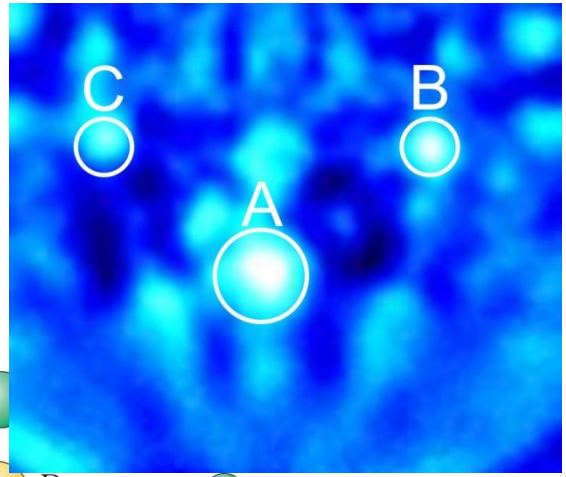




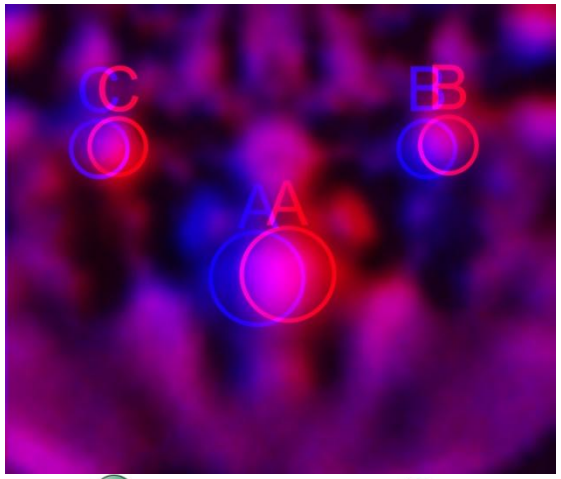
(a)
[1-11]



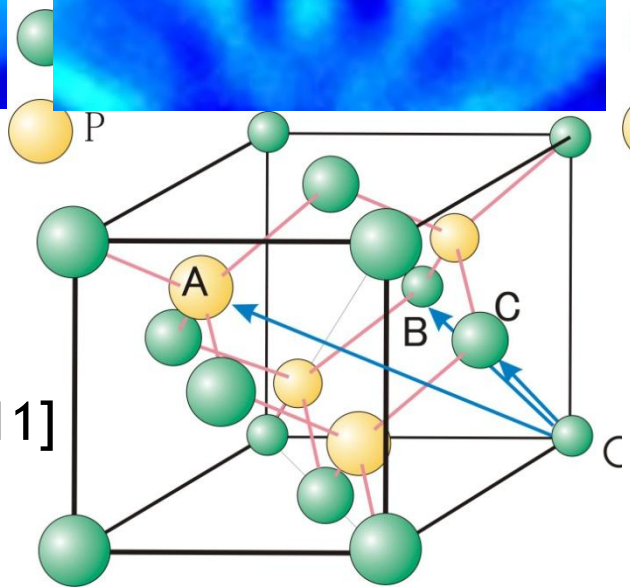




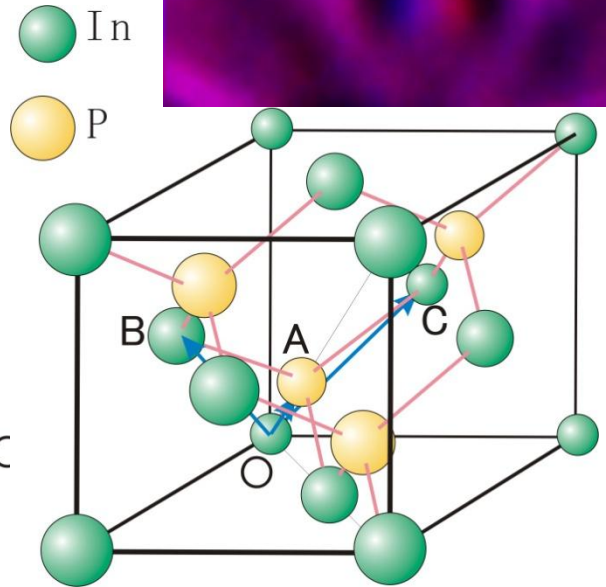
(b)
[111]



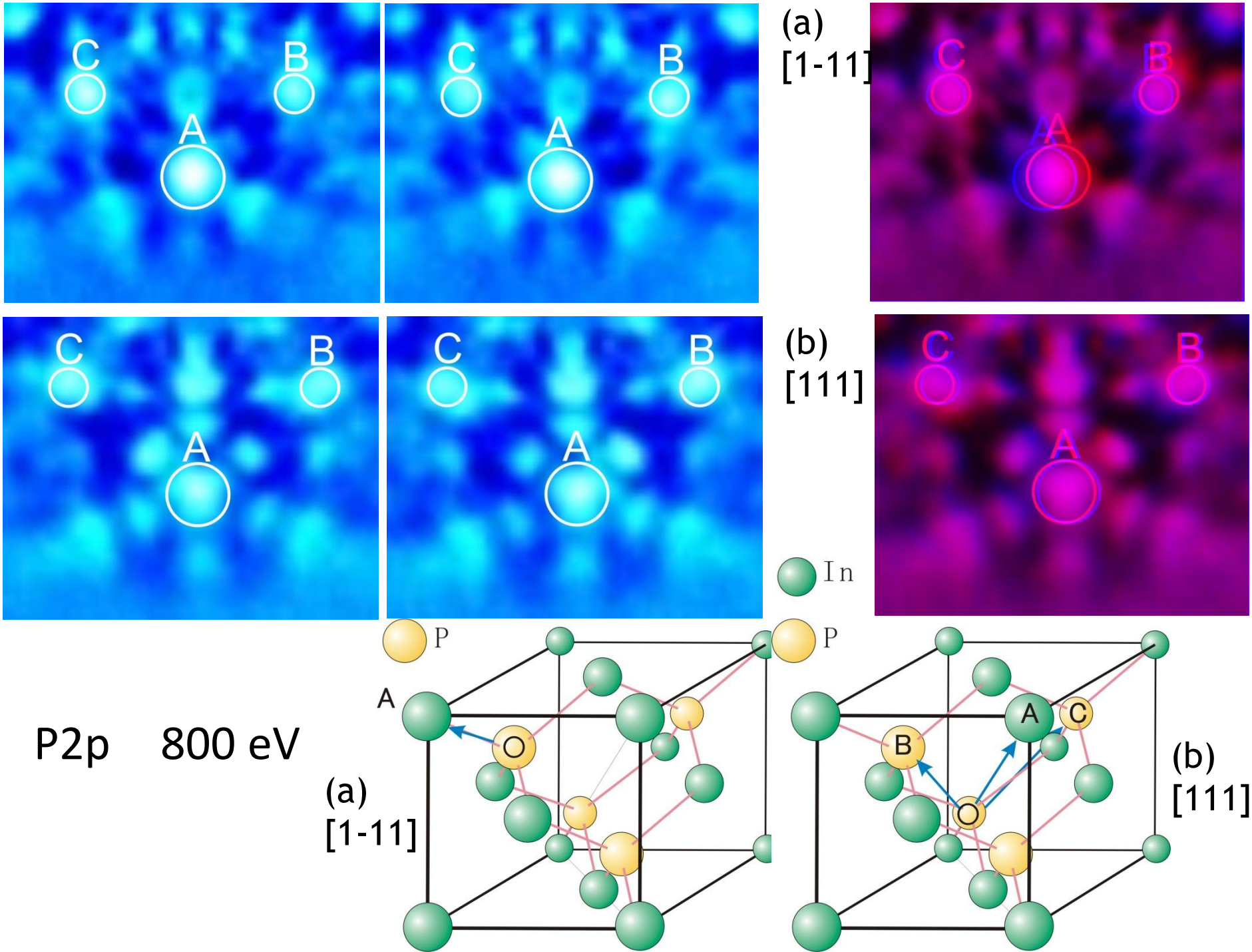
In3d 800 eV



(a)
[1-11]



(b)
[111]



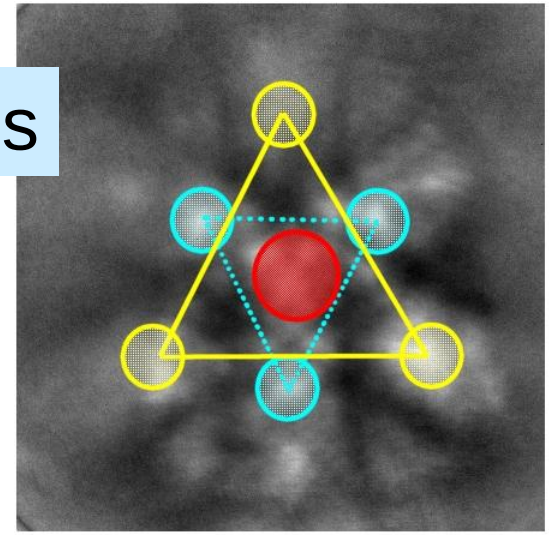
P2p 800 eV

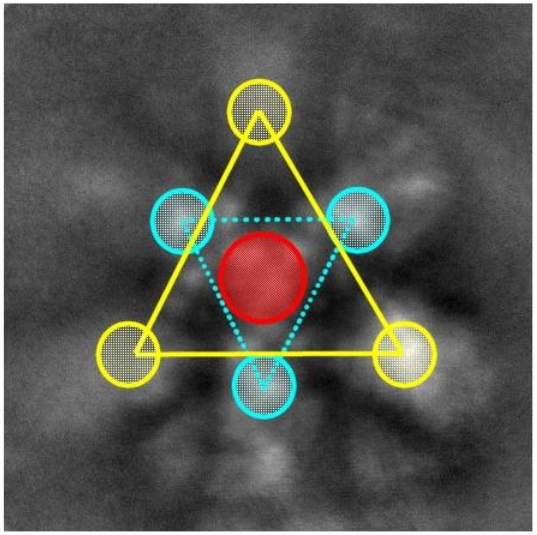
B-dope Diamond(111)

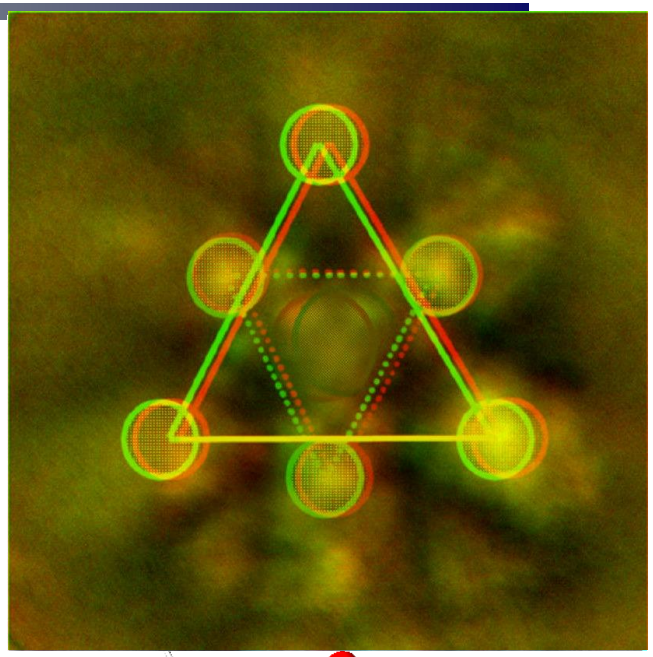
Superconductor

2% E_K : 600 eV

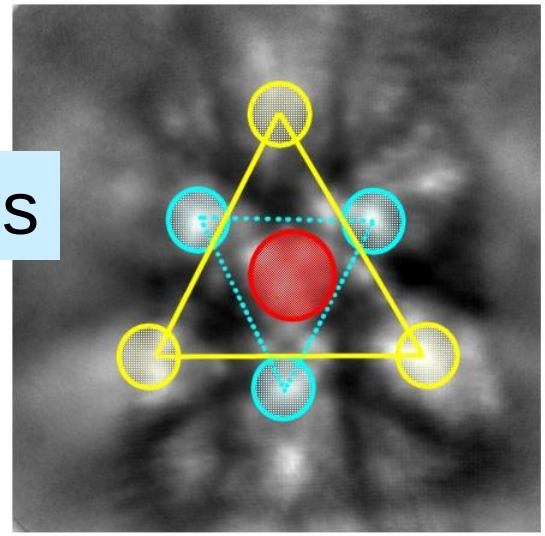
B 1s

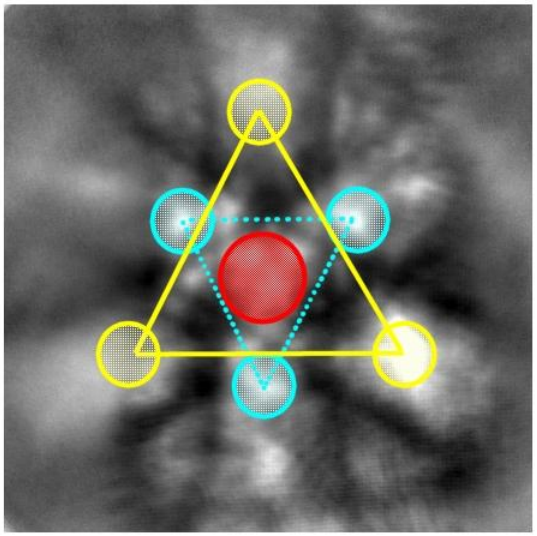


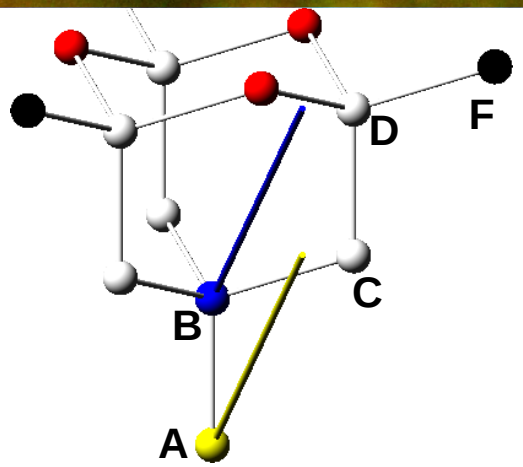




C 1s







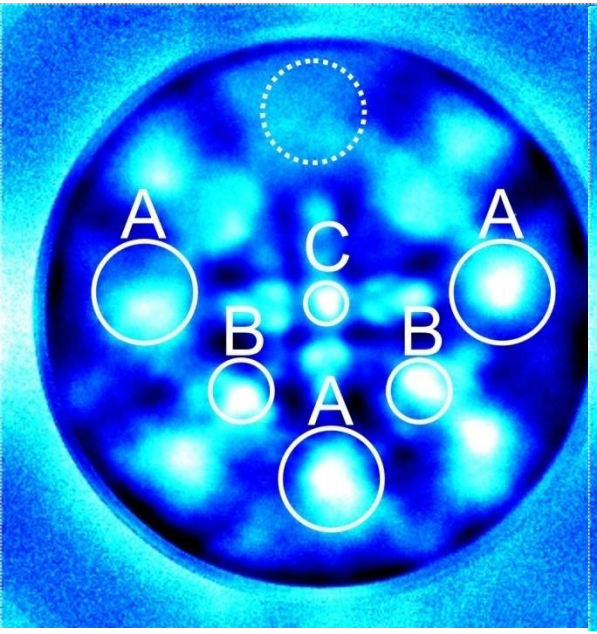
C1s & B1s patterns are ***similar***

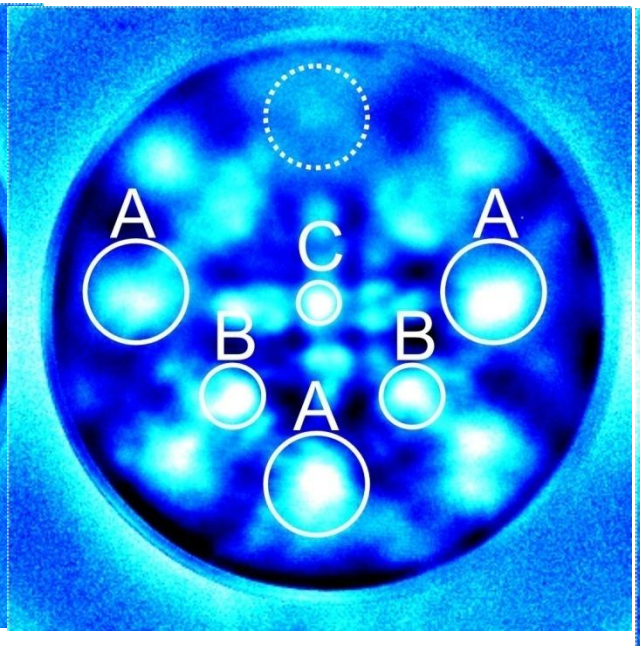
One substitutional site

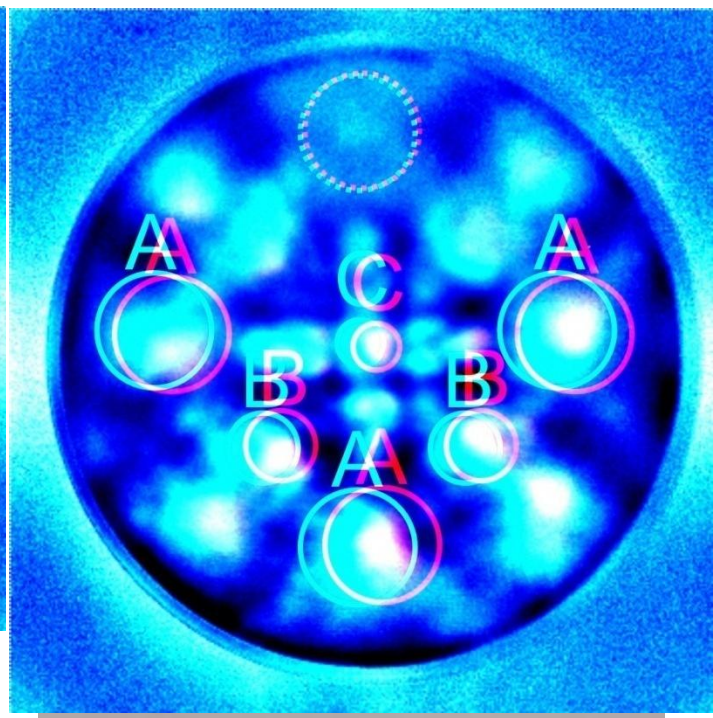
Atomic arrangement around 2% impurity could be clarified

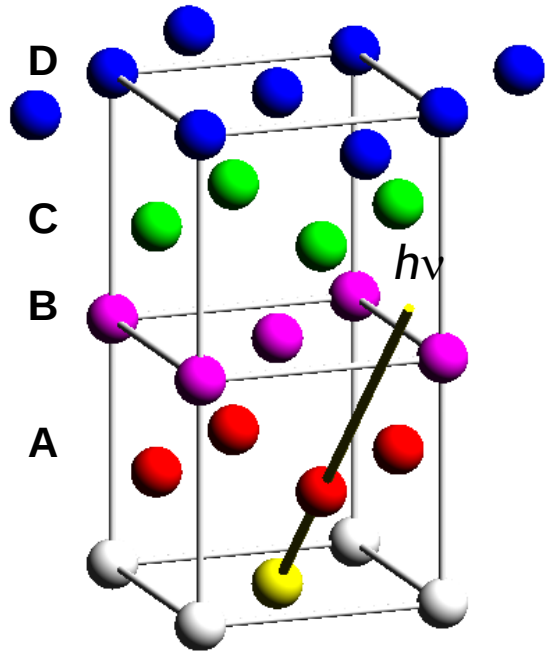
Cu(001)

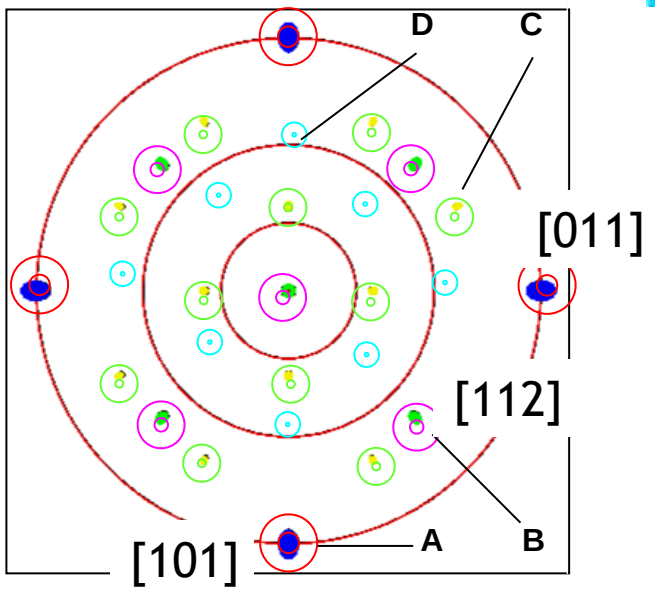
E_k 600 eV $h\nu$ 680.6 eV Cu $3p_{3/2}$





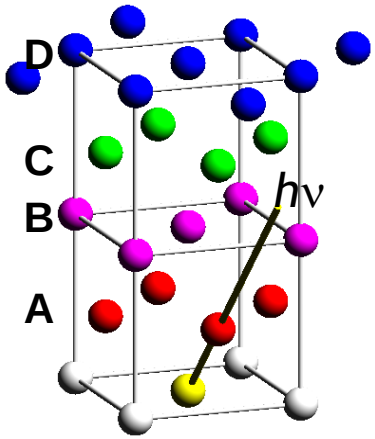


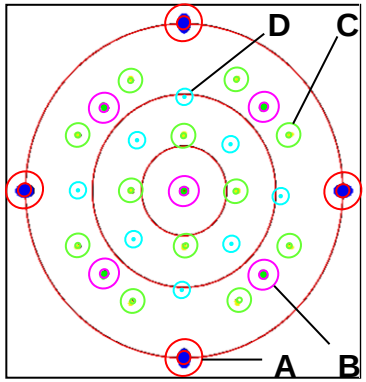


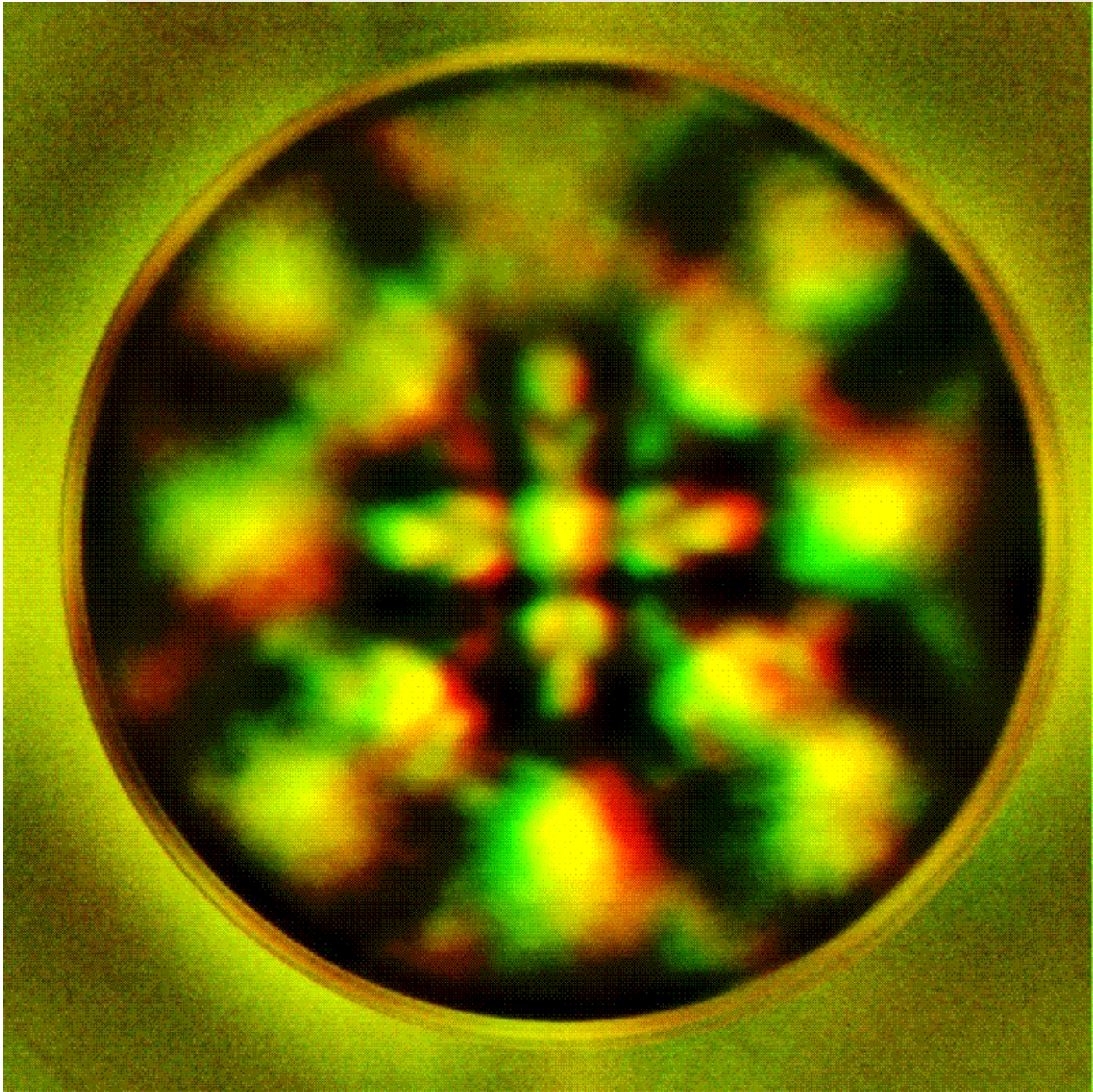


Stereo movie of Cu

fcc structure







Stereo-PEEM

- **PEEM : Δx= μm, ΔE=0.1%**
- **Stereo Atomic Image**

(Circularly polarized-light PEAD)

Δx= 0.2 Å , Θ=±60°

Wide Acceptance Angle (about 1 sr) Electrostatic Lens (WAAEL)

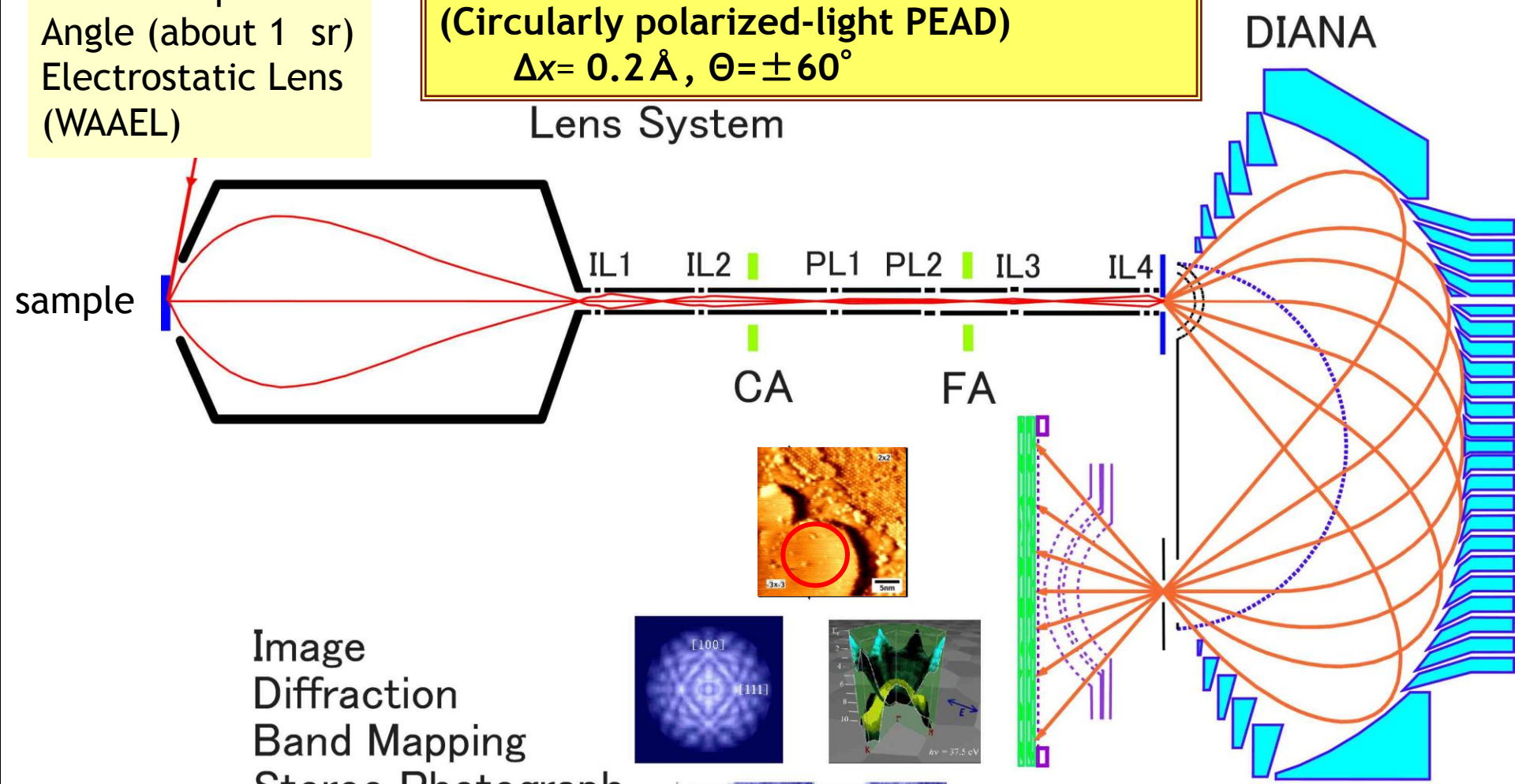
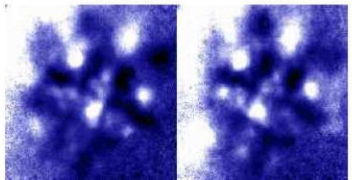


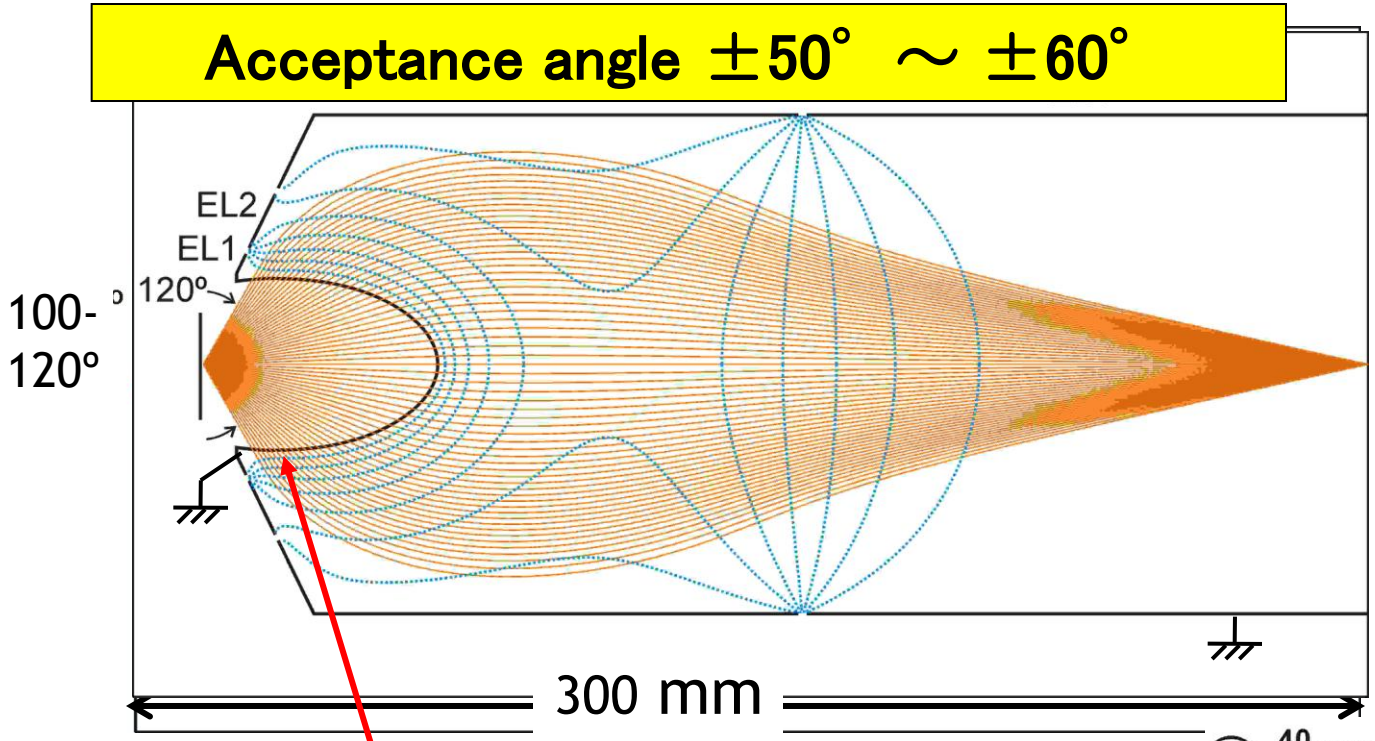
Image
Diffraction
Band Mapping
Stereo Photograph

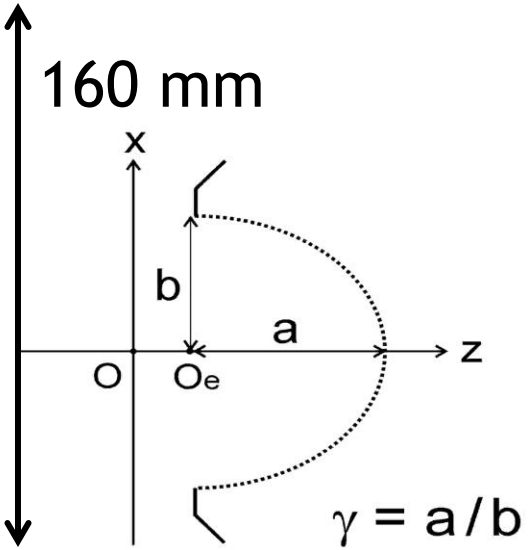


For selected micro-area!

Wide Acceptance Angle Electrostatic Lens (WAAEL)

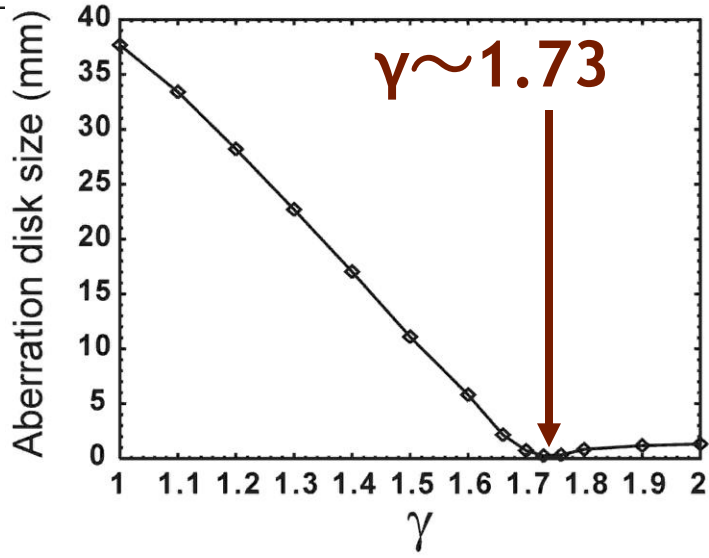
Acceptance angle ±50° ～ ±60°



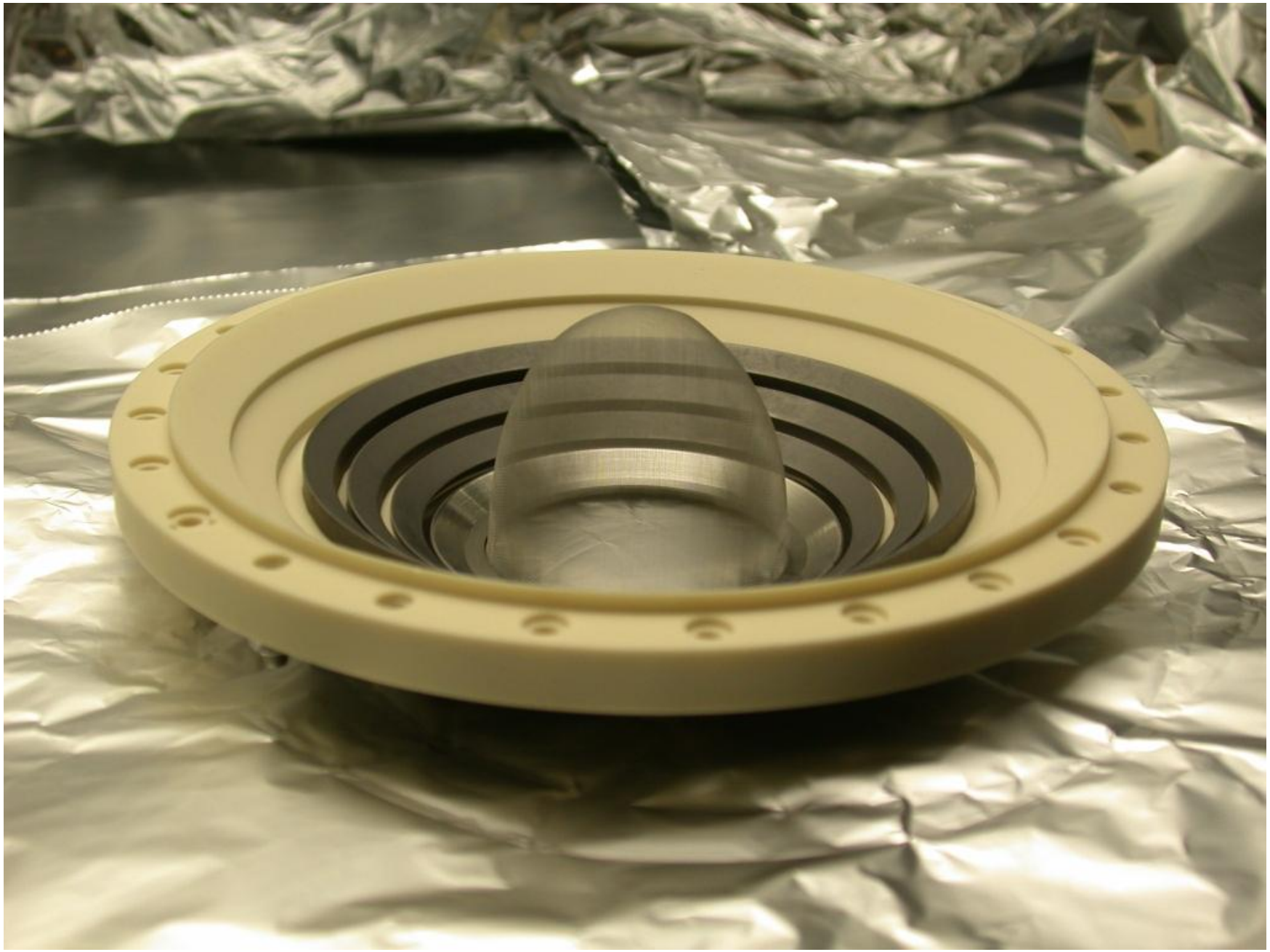


Nearly ellipsoidal shape mesh

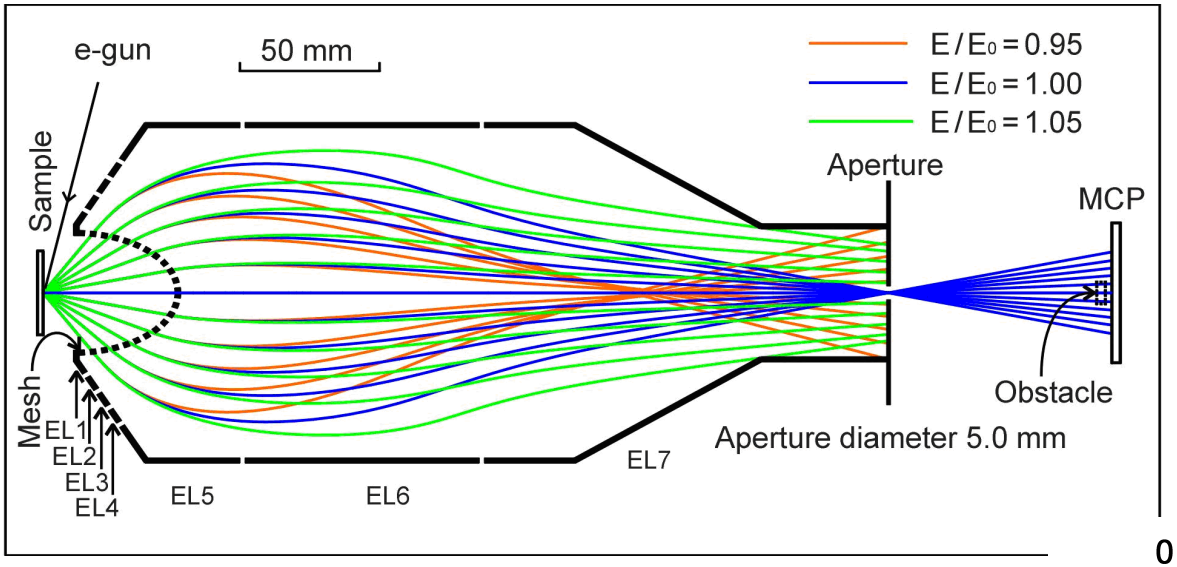
Sample region : field free
Acceptance angle reduces to 1/5

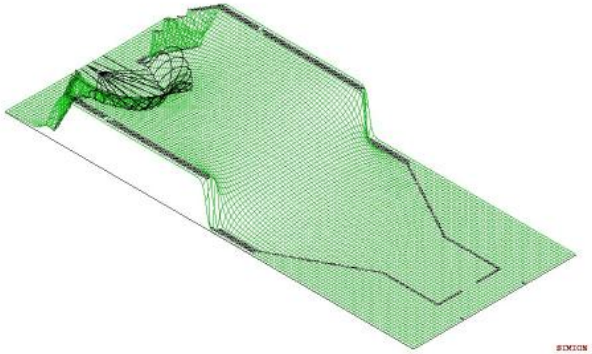


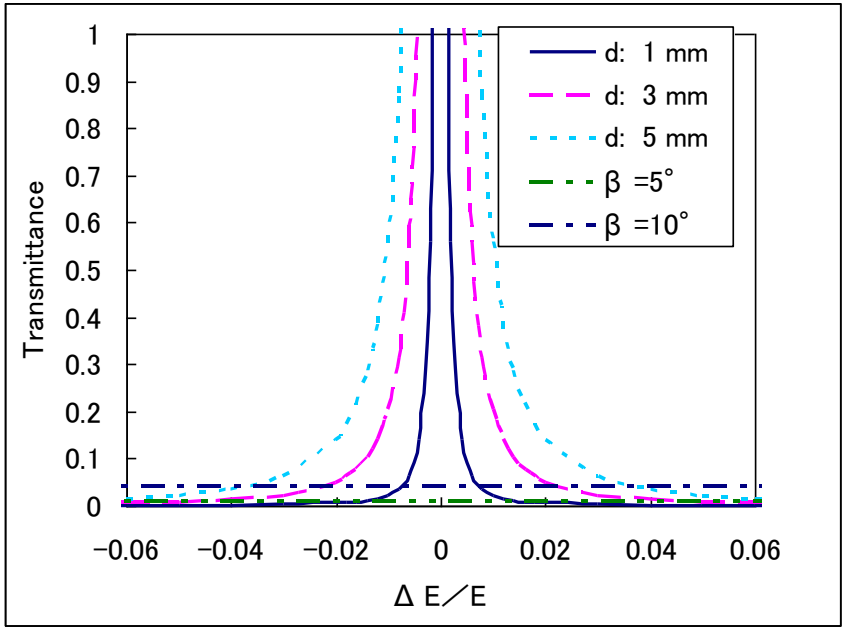
[1] H. Matsuda, H. Daimon, M. Kato and M. Kudo, Phys. Rev. E 71, 066503 (2005)
[2]patent: PCT/jp2004/016602, Japan 2004-208926
[3] L. Tóth, H. Matsuda, T. Shimizu, F. Matsui and H. Daimon, J. Vac. Soc. Jpn. 51, 135 (2008)

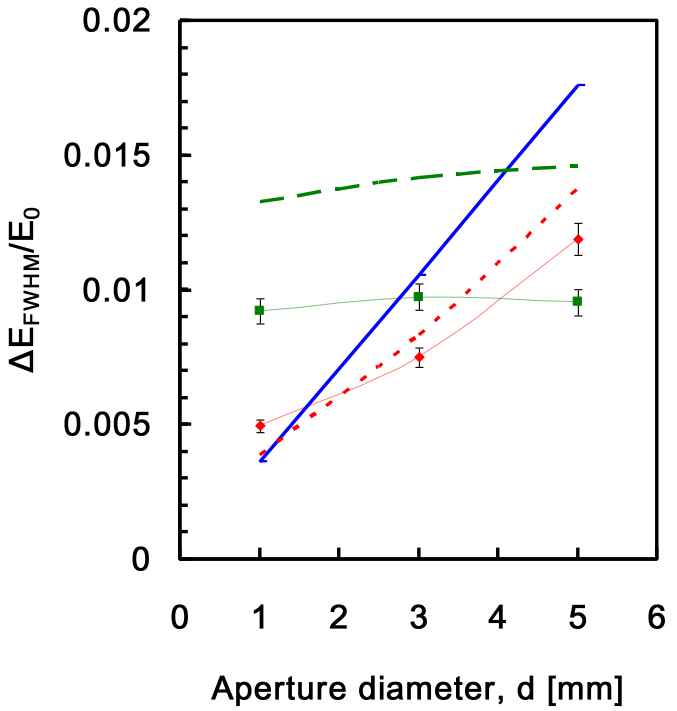


Energy analysis of DELMA by energy aperture

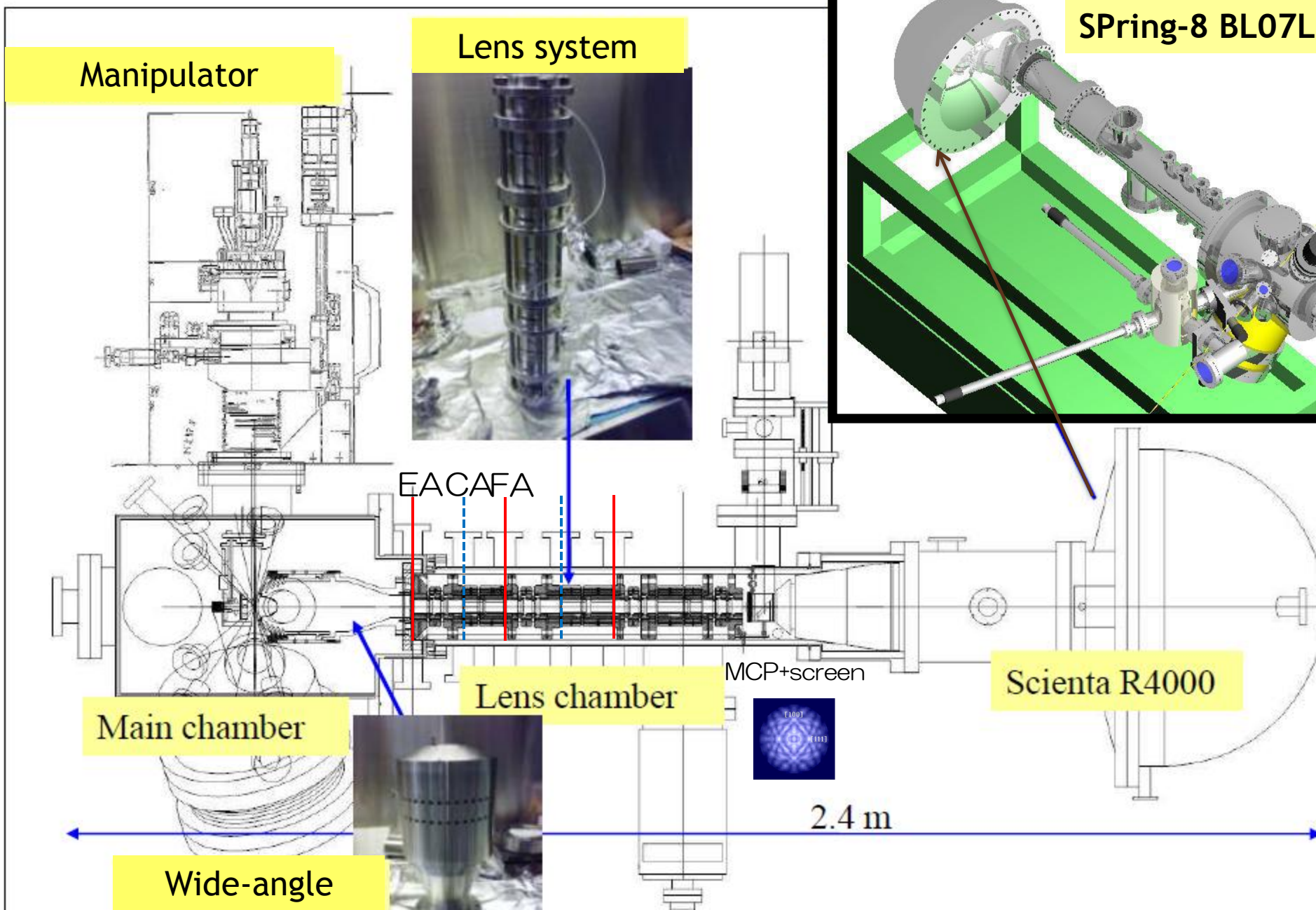




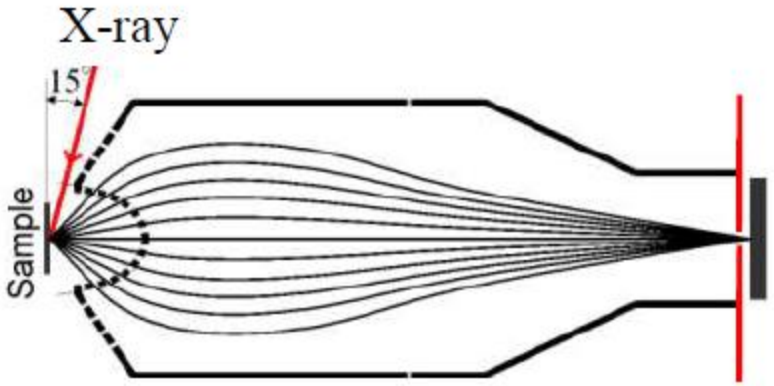




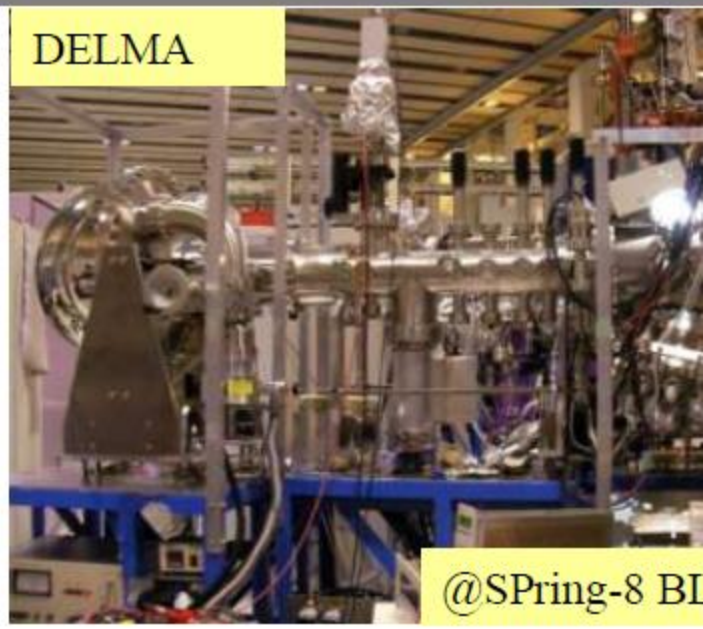
DELMA (Display-type ellipsoidal mesh analyzer)+HSA



Photoelectron Spectrum

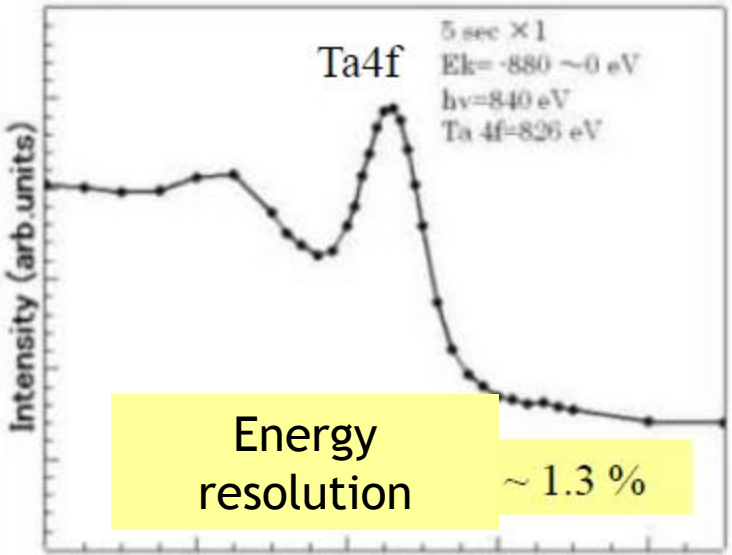


Φ 1.3mm aperture



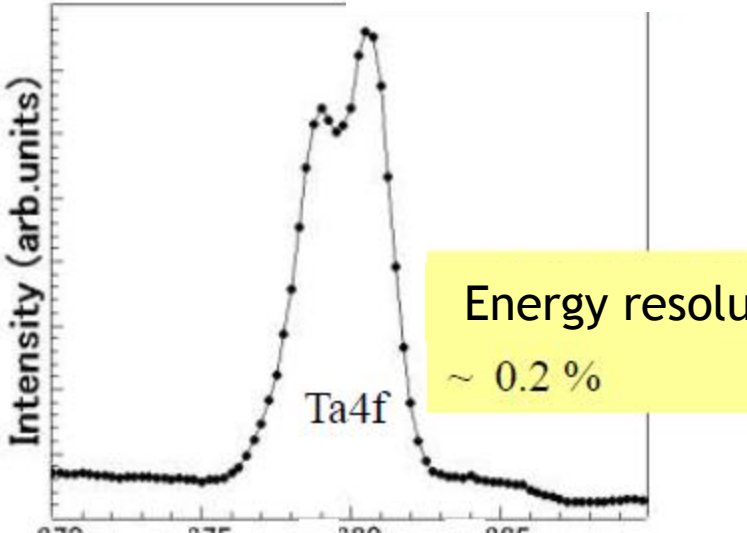
Sample: TaS_2

Spectrum obtained by energy aperture



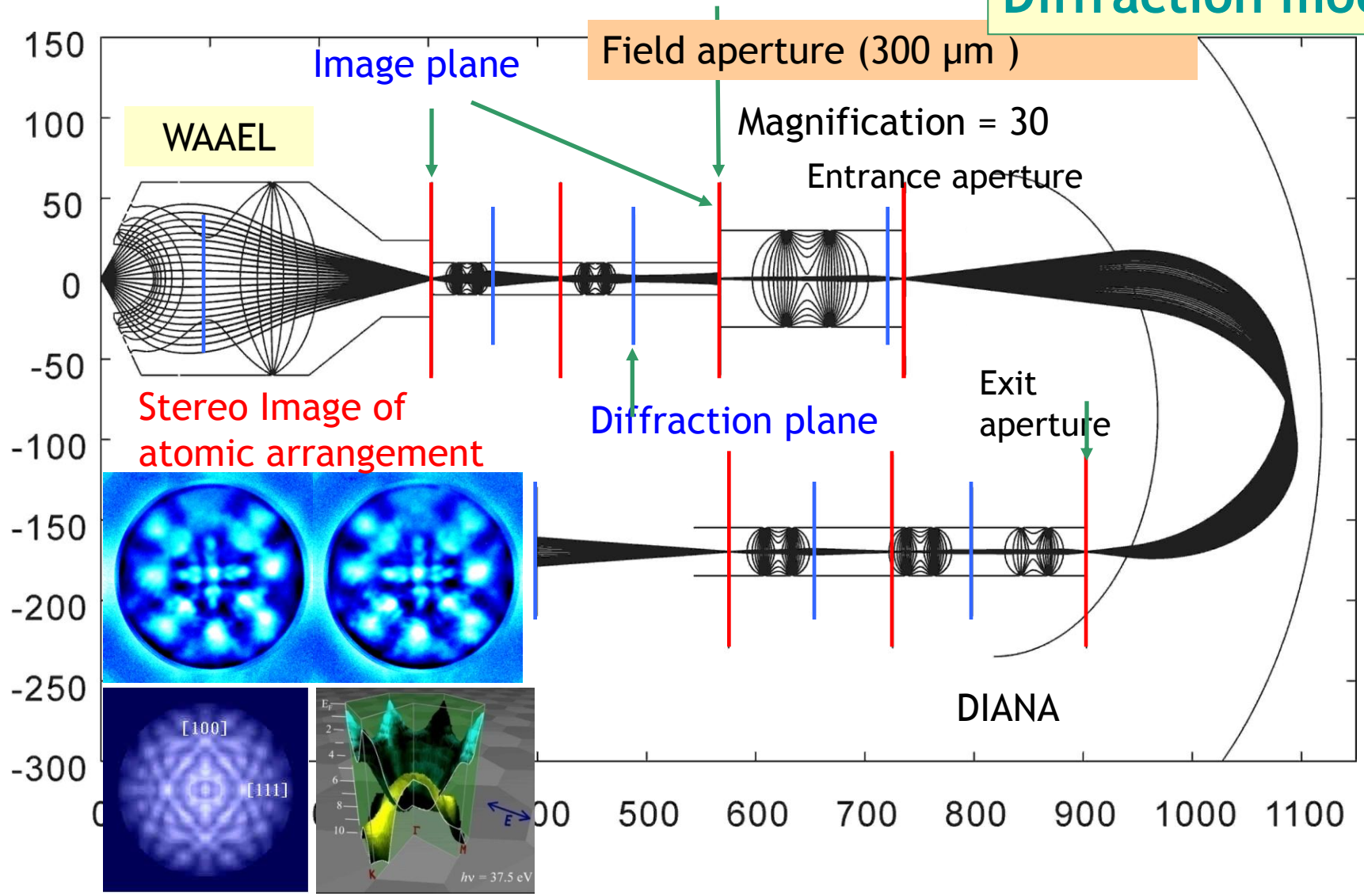
DELMA+HSA

(Slit width is 3 mm



DELMA: Display-type Ellipsoidal Mesh Analyser

Diffraction mode



Diffraction

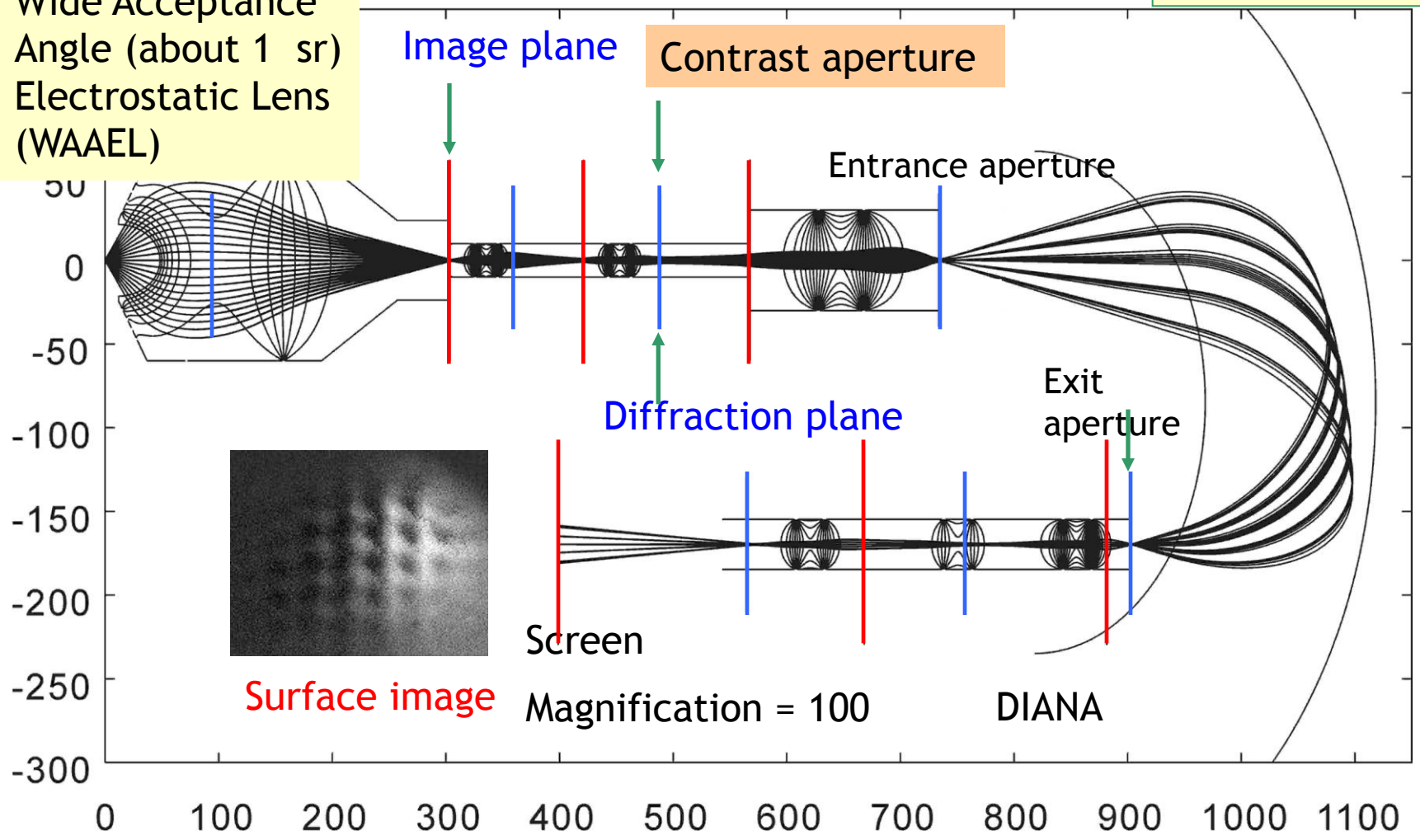
Band
structure

H. Daimon, H. Matsuda, L. Toth, F. Matsui, Surface Science, 601, 20, 4748

DELMA: Display-type Ellipsoidal Mesh Analyser

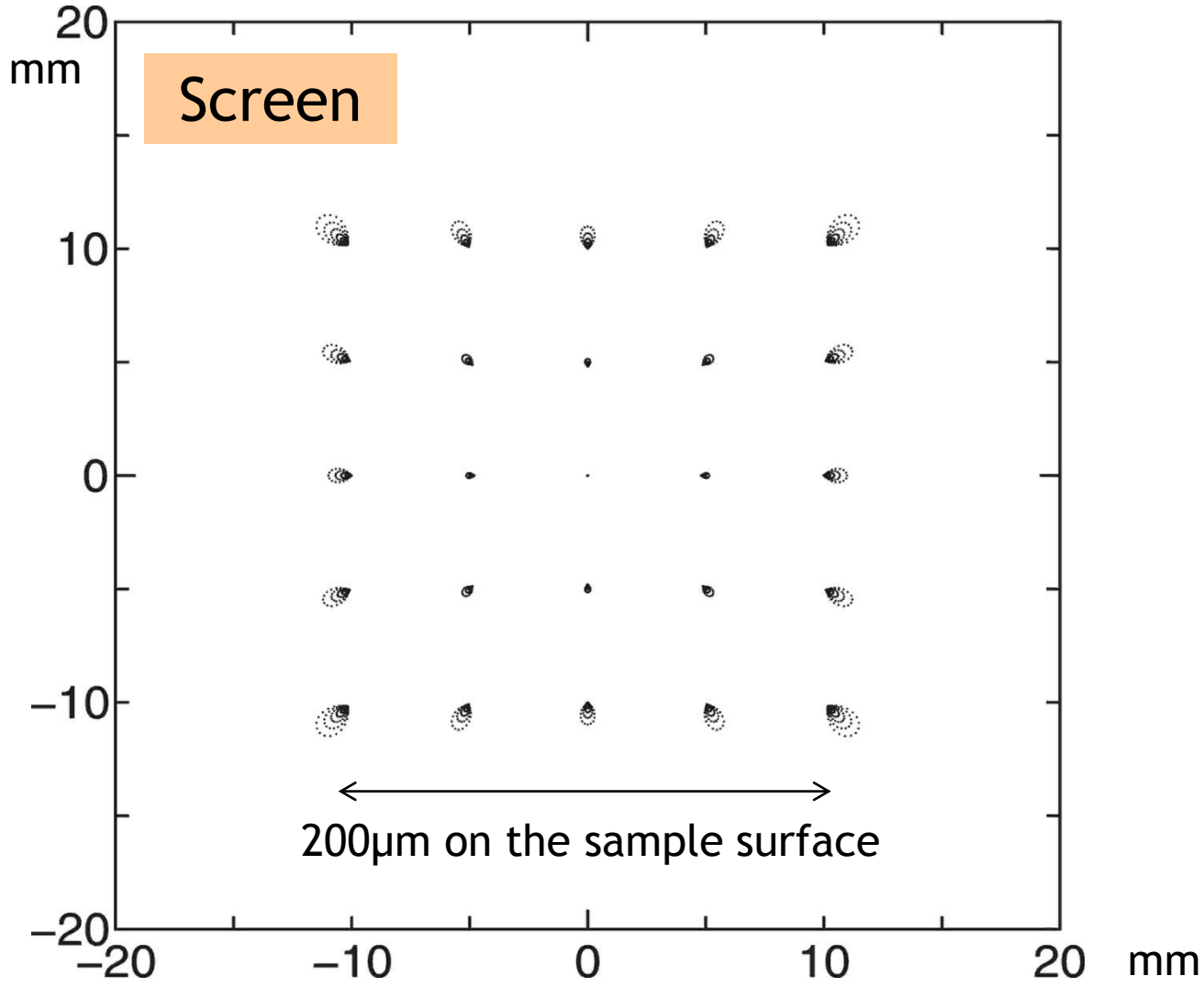
Imaging mode

Wide Acceptance Angle (about 1 sr) Electrostatic Lens (WAAEL)

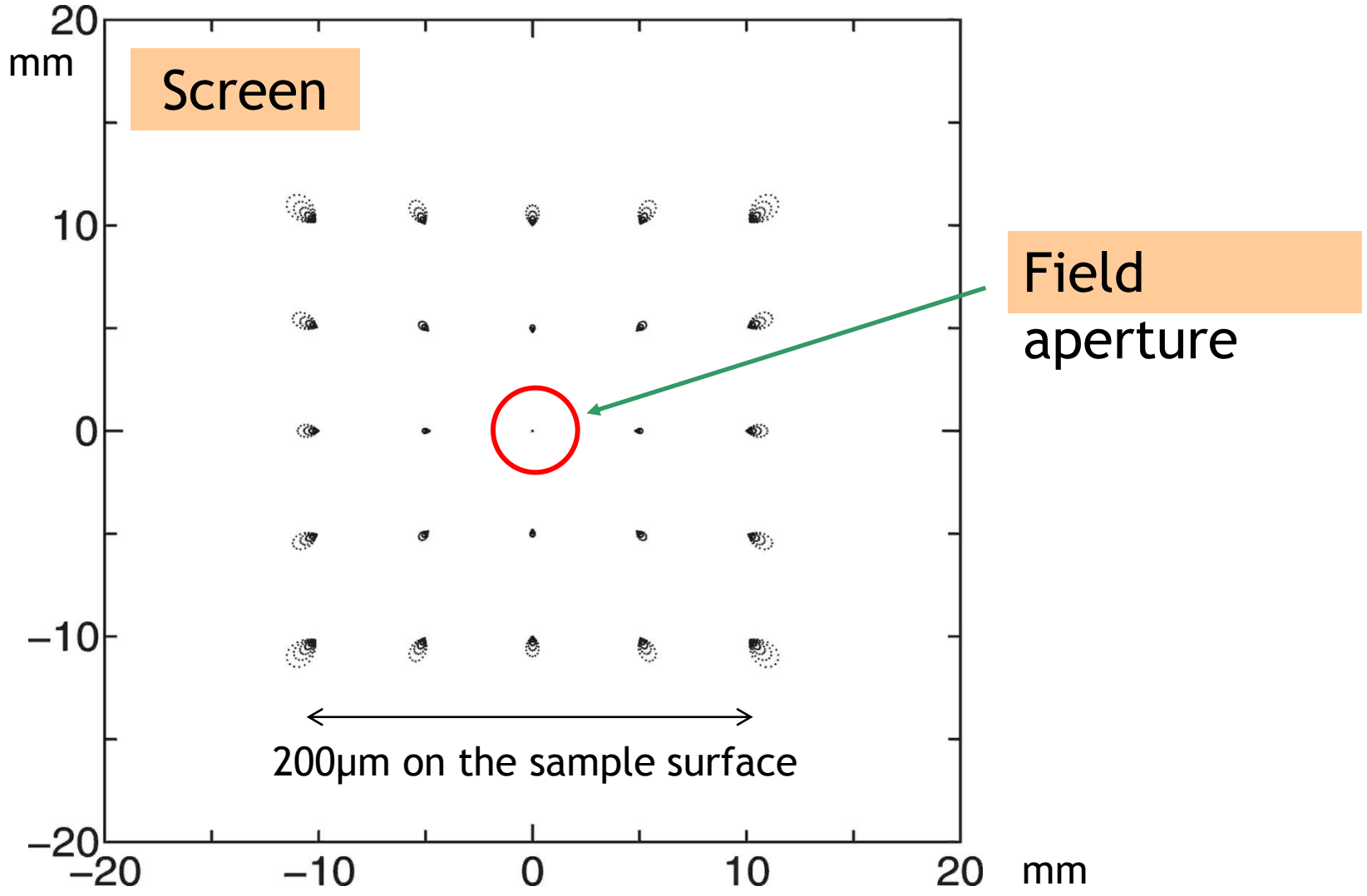


H. Daimon, H. Matsuda, L. Toth, F. Matsui, Surface Science, 601, 20, 4748, (2007).

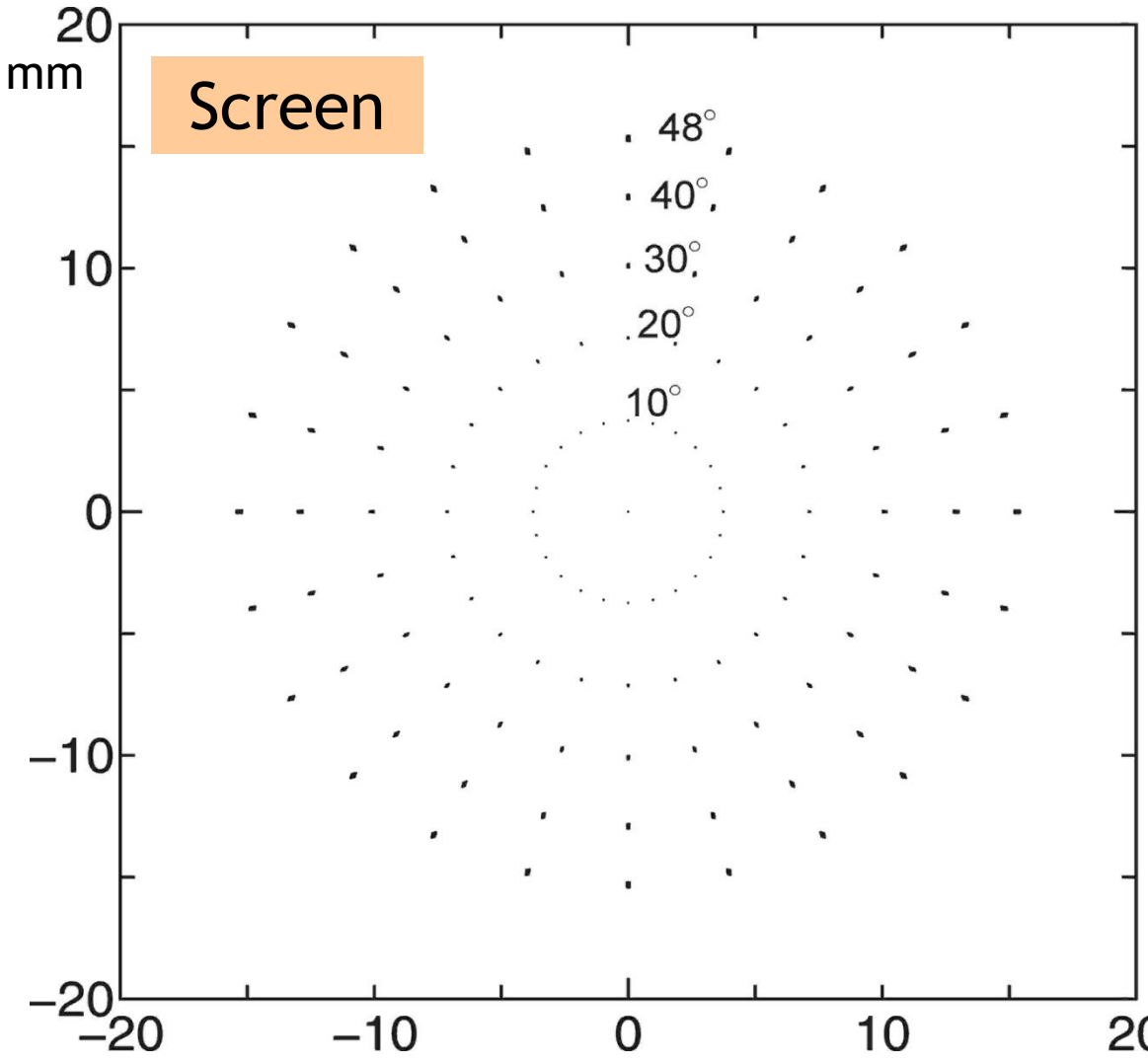
Off-Axes Point-source image



Point-source image



Point-source image



300 μm field aperture

10 μm selected area

30 μm field aperture

→1 μm selected area

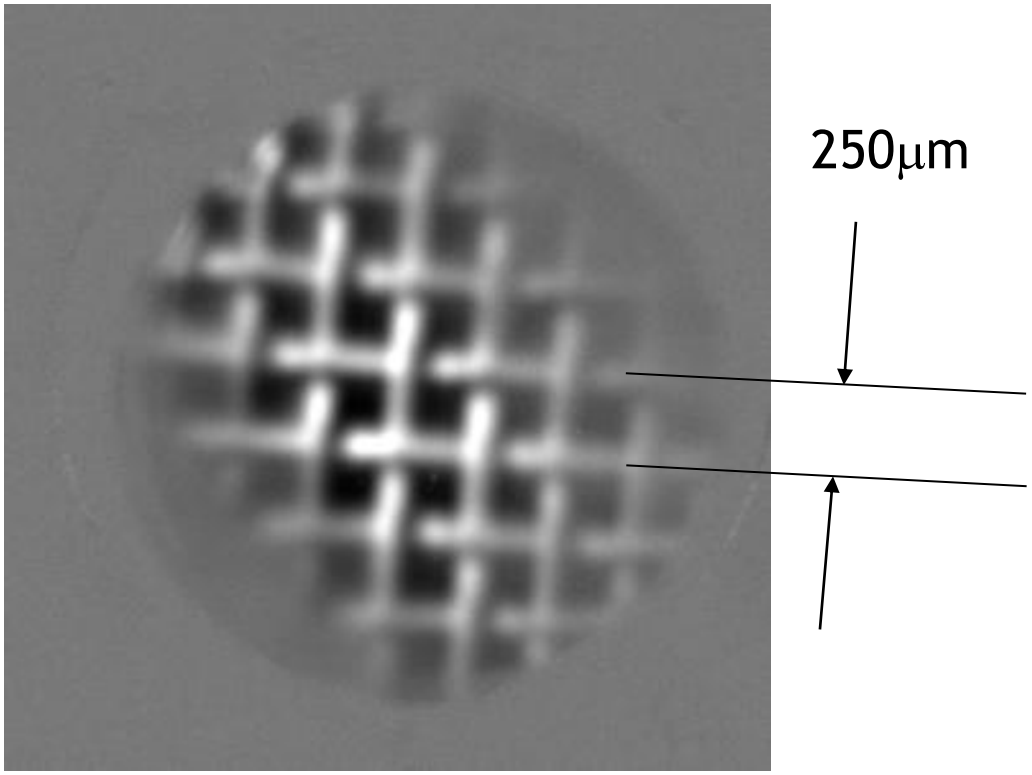
Test experiments using an electron gun

Imaging mode

Sample: SUS316 woven mesh (#100, φ_wire=50 μm)

Ek=1000eV

Magnification ~10



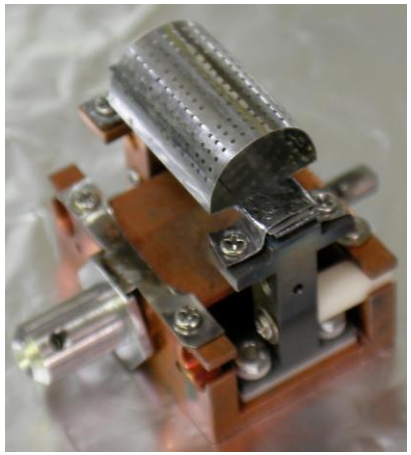
Contrast aperture : the largest one (6mm)

Spatial resolution ~ 20 μm

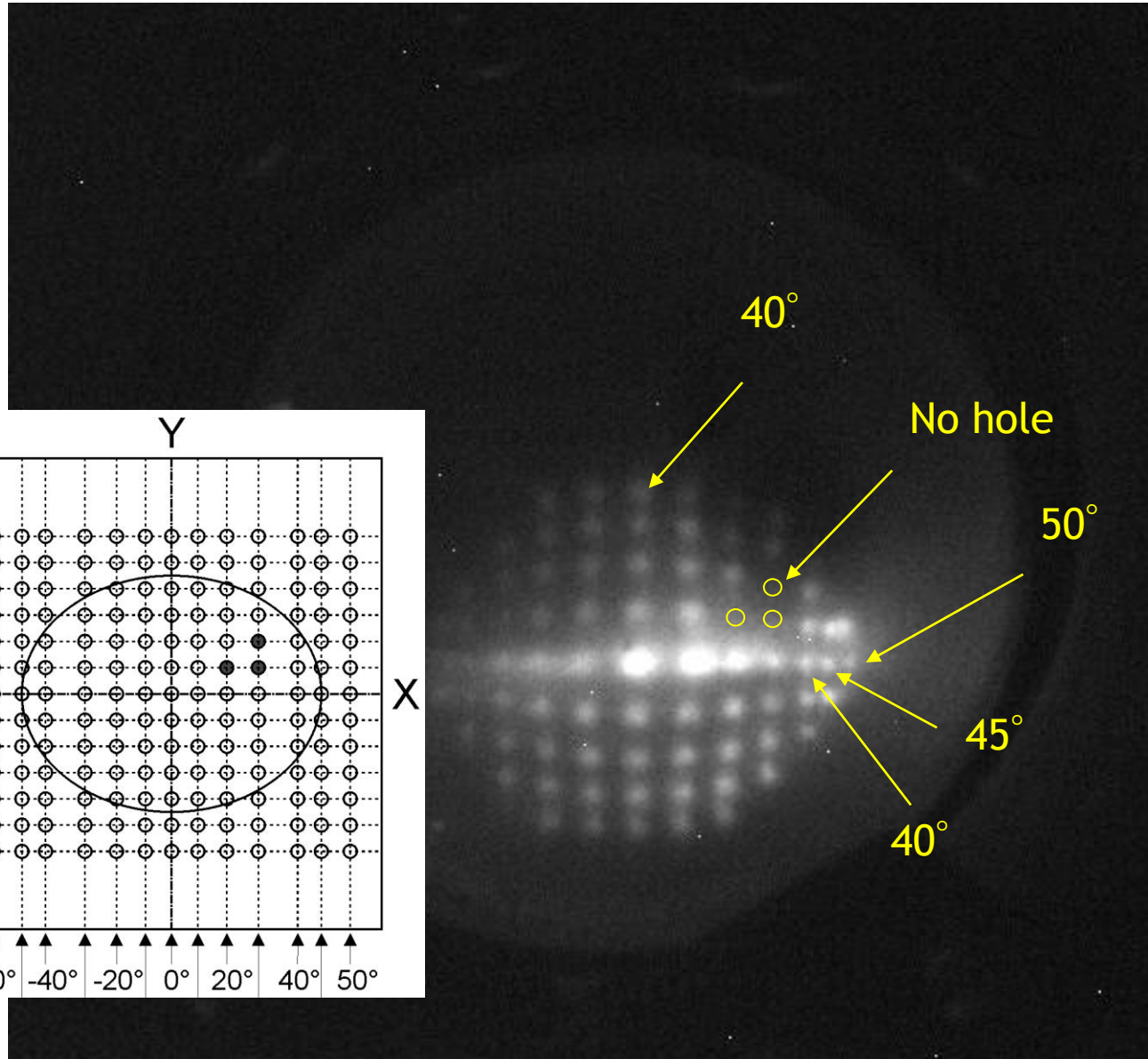
Test experiments using an electron gun

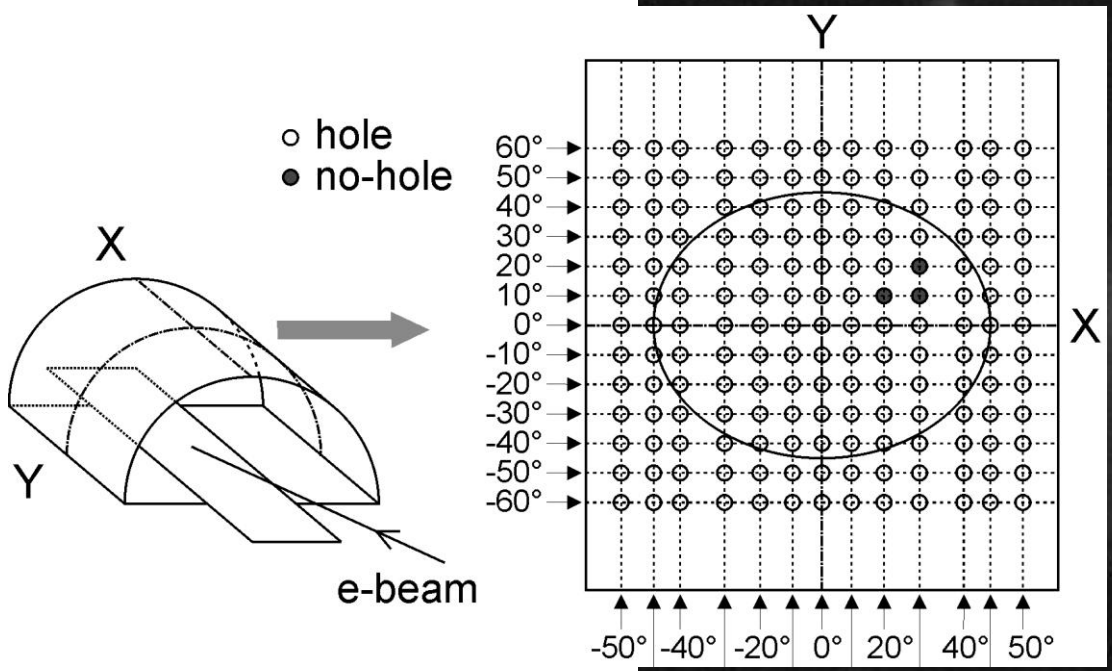
Angular mode

Angle test device



Ek=1000eV

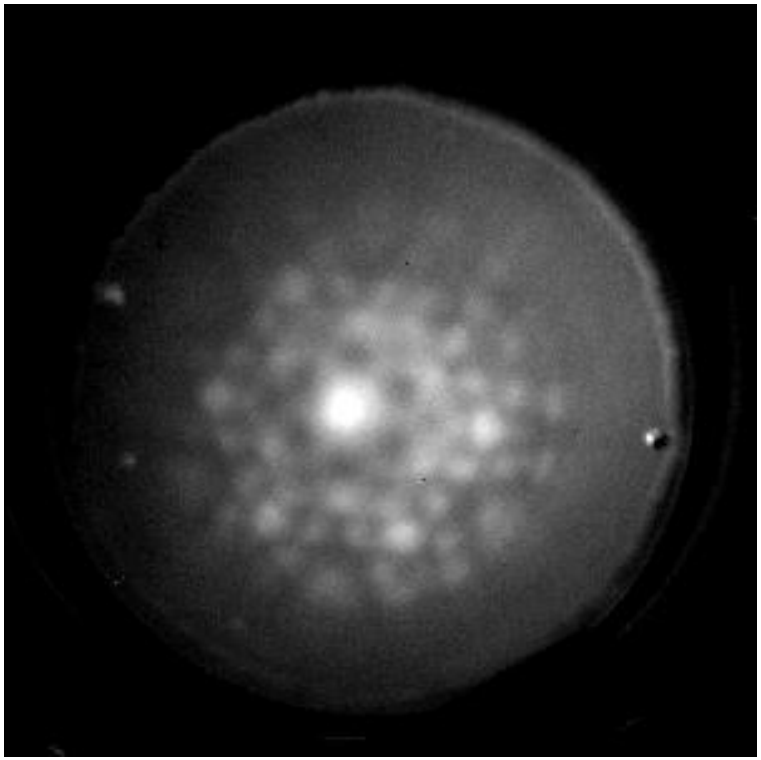


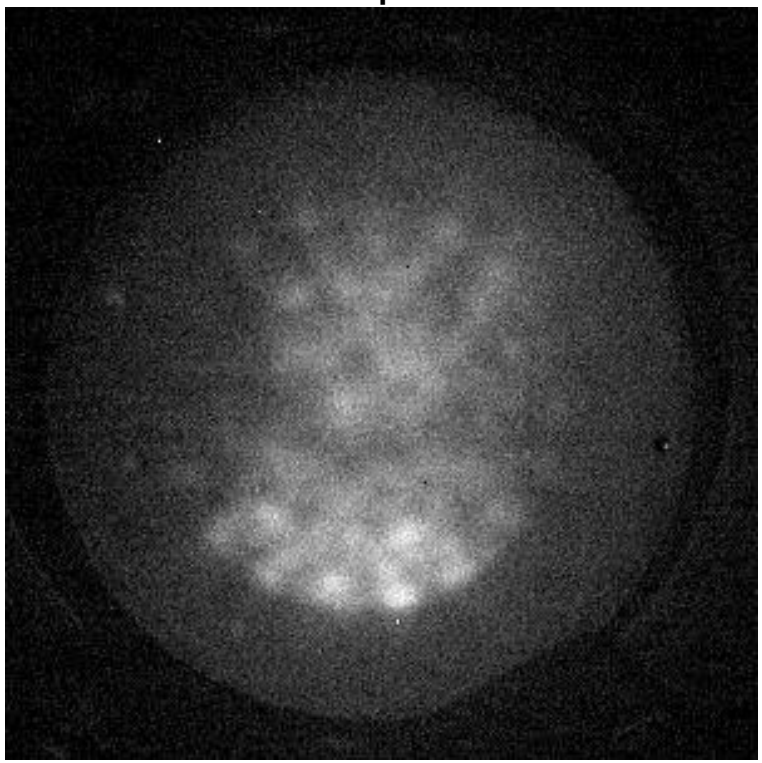


Sample: single crystalline graphite

$h\nu$=990 eV Ek=700 eV (BL07LSU)

Horizontal vs. Vertical linear polarization





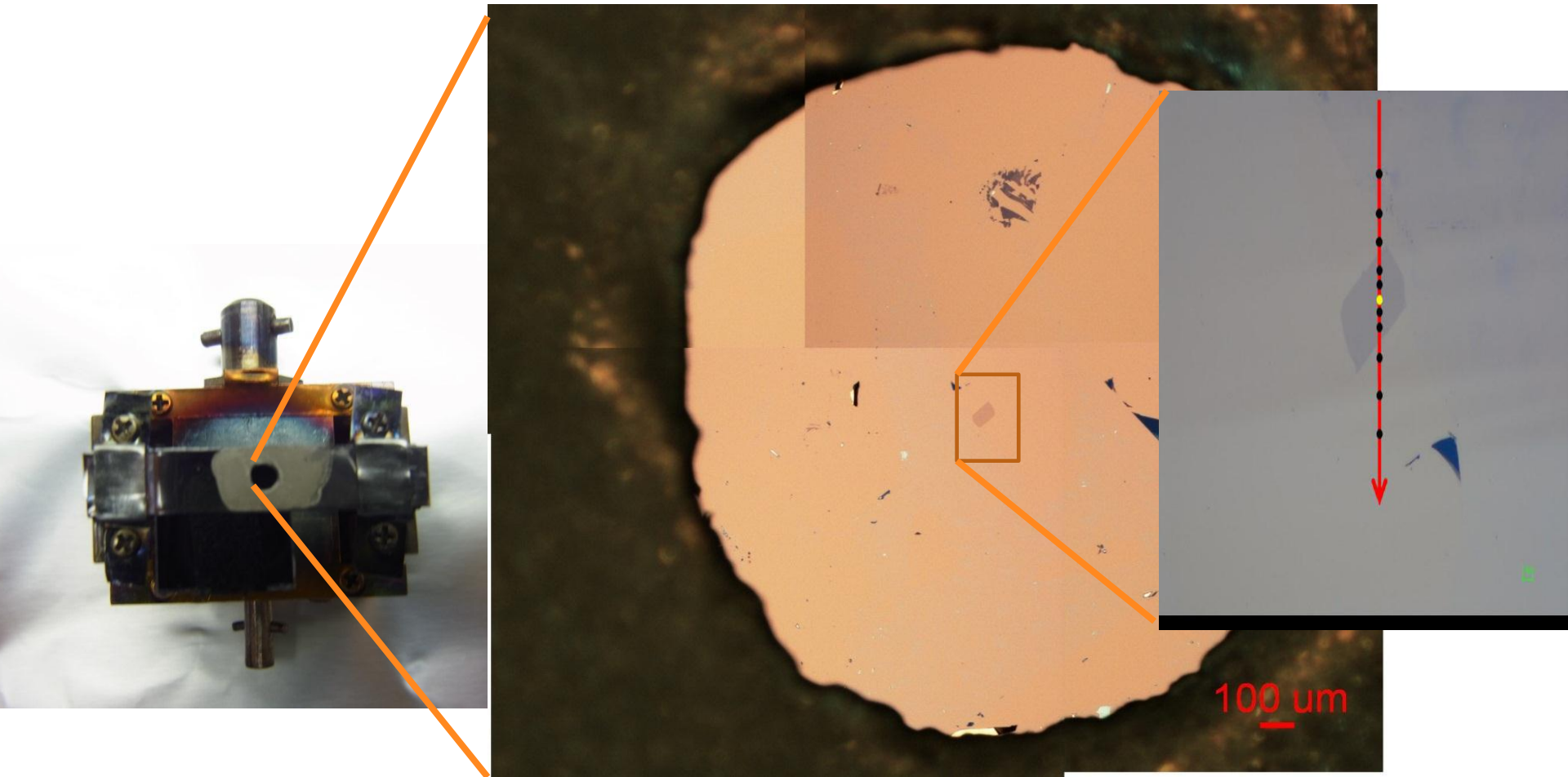
Photoelectron diffraction pattern

NAIST Daimon Group

グラフェンの光電子回折像の測定

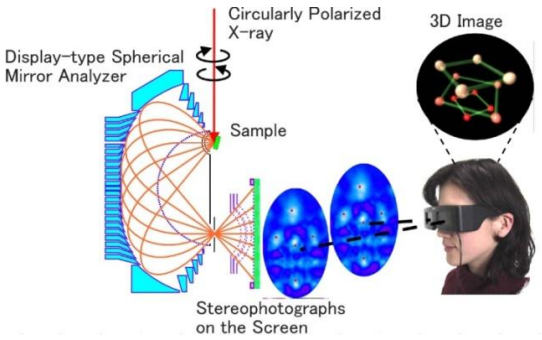
奈良先端大・物質創成、JASRI/SPring-8[A]

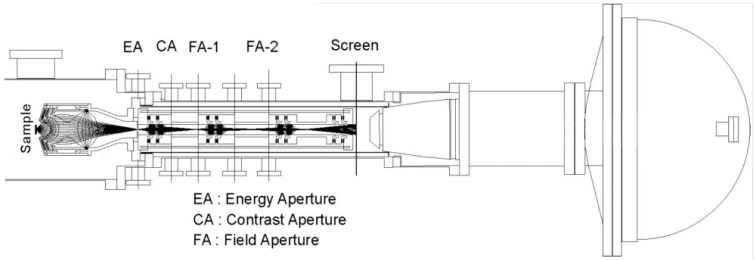
石井良、松井文彦、黄晋二、細川陽一郎、松下智裕[A]、森田誠、
北川哲、橋村詩織、藤田將喜、安田馨、大門寛

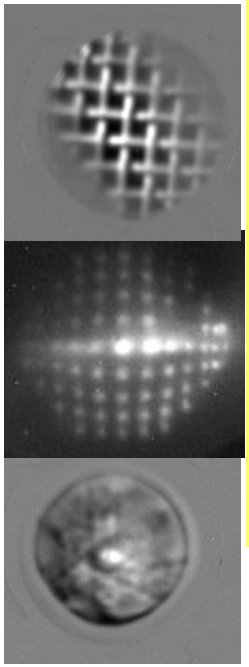


DIANA

- ±60°
- Δ E=0.002E0
- 価電子帯光電子分光
  - 軌道解析（直線偏光）
- 内殻光電子回折
  - 原子立体写真（円偏光）
  - 原子層分解MCD
  - 光電子回折分光







⇒

DELMA

- ±45 ～ 60°
- Δ E=0.0002E0

  R4000で高分解能
- 価電子帯光電子分光

  軌道解析　⇒ 高分解能2D-ARPES
- 内殻光電子回折

  原子立体写真
  原子層分解MCD
  光電子回折分光
  ⇒ 化学シフト分解
- PEEM機能 （(x、y)Δ30 μm）

  拡大像、微小試料
- HAXPES機能（深さ(z)分解 Δnm）

  CrKα 6.4keV、 界面組成・電子状態
- 時間分解 （in future）

  2D情報が一度に
  レンズシステムがTOF tube

1 D, 2D, 3D photoelectron spectroscopy

		電子状態 UPS $h\nu = 7-100$ eV	原子構造 XPS $h\nu > 100$ eV
1D:角度積分 $E_k = h\nu - E_B - \Phi$		状態密度 1D(E_B)	化学シフト 1D(R)
2D:角度分解 (Scienta, DIANA)		バンド分散 2D(k, E_B) 3D(k_x, k_y, (E_B, k_z))	光電子回折・ホログラフィー 立体原子写真 3D(x, y, z)
3D:空間・時間分解	2D:PEEM	*nano*$\Delta x,y+t$ 3D(k_x, k_y, (E_B, k_z))	
	3D:DELMA Stereo-PEEM	*micro*$\Delta x,y+z+t$, 3D(k_x, k_y, E_B), 3D(x, y, z)	