Slow positron applications at Slow Positron Facility of Institute of Materials Structure Science, KEK

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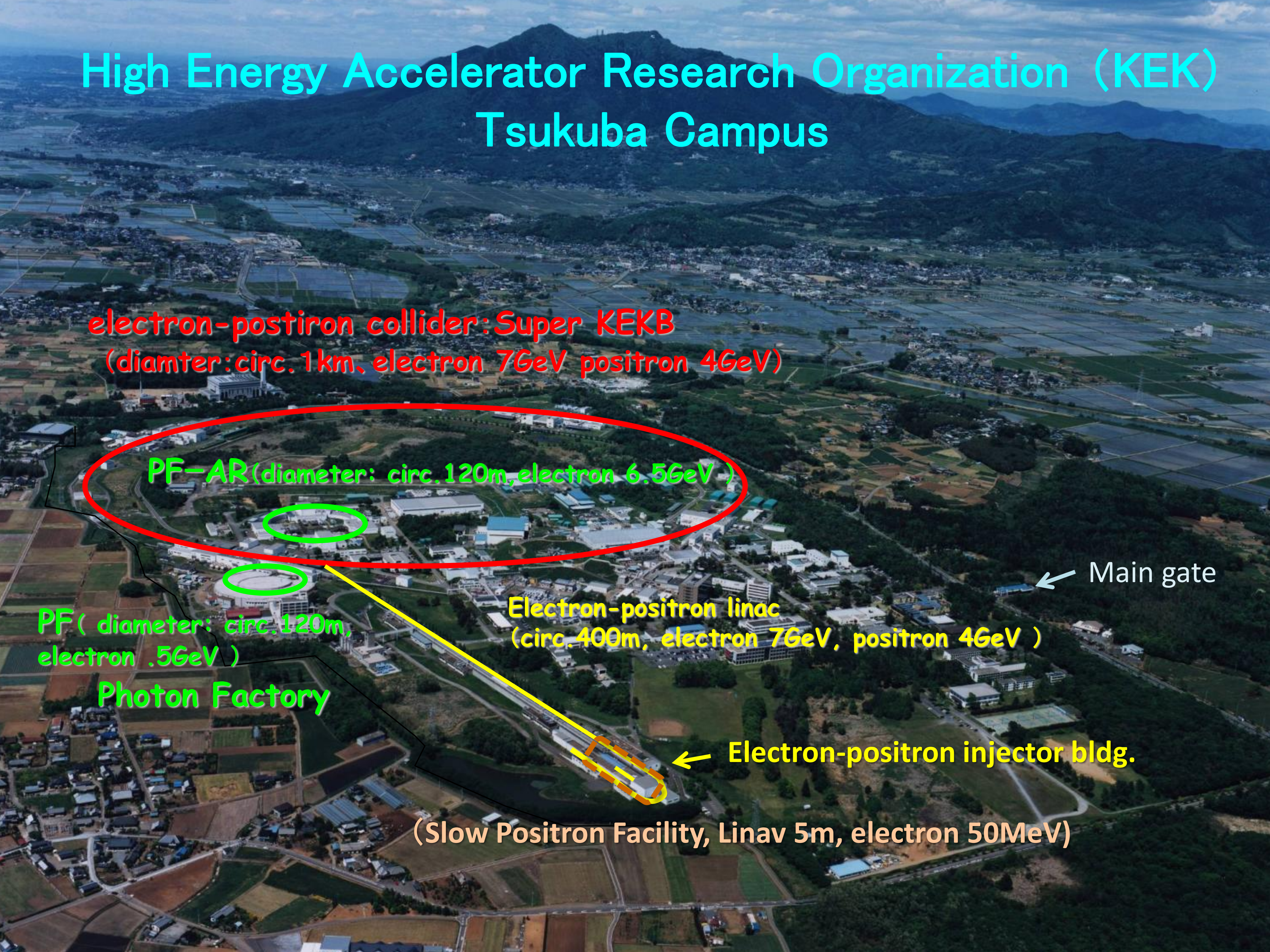
Outline

- Colaborators
- Overview of Slow Positron facility
- Creation of slow positron
- Brightness enhancement
- TRHEPD
- LEPD
- Puls-stretching
- Ps^- (Ps negative ion)
- Ps-TOF

Collaborators

- KEK: (IMSS) T. Kosuge, A. Yagishita, A. Ichimiya;
  (Accelerator) S. Ohsawa, M. Ikeda, A. Shirakawa,
  K. Furukawa, H. Honma;
  (Radiation Control) H. Iwase, T. Sanami
- JAEA: Y. Fukaya (TRHEPD)
- QST: K. Wada, M. Maekawa, A. Kawasuso (TRHEPD, LEPD)
- Chiba Univ.: M. Fujinami (LEPD)
- Tokyo Univ. of Sci.: Y. Nagashima (Ps^-, Ps-TOF),
  T. Tachibana (Ps^-)
- AIST: T. Shirasawa (LEPD), K. Michishio (Ps^-)
- Riken: S. Kuma, T. Azuma (Ps^-)

High Energy Accelerator Research Organization（KEK）
Tsukuba Campus



Slow Positron Facility, KEK

High Intensity

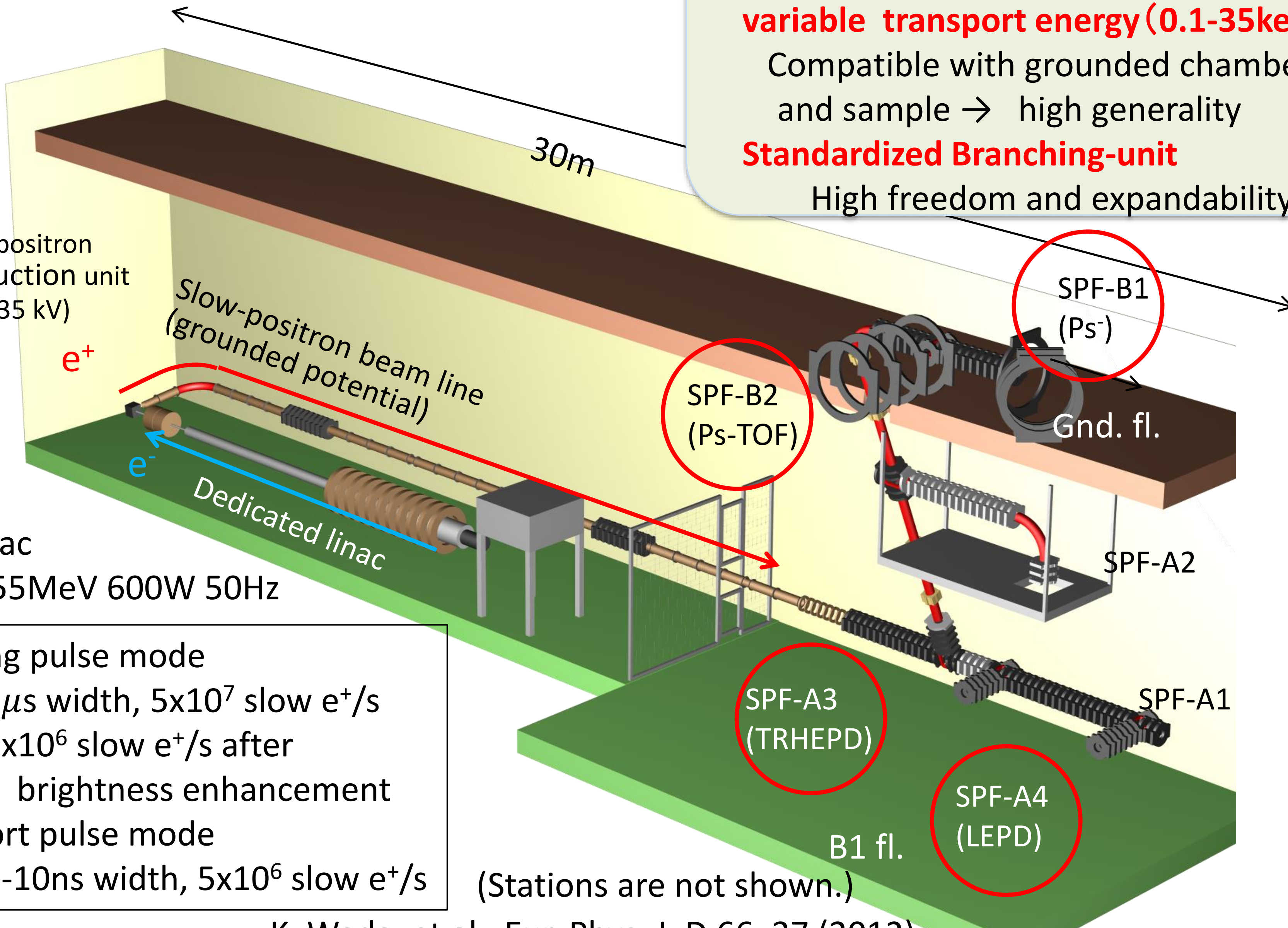
5×10^7 slow e^+/s (long pulse mode)

variable transport energy（0.1-35keV）

Compatible with grounded chamber and sample → high generality

Standardized Branching-unit

High freedom and expandability



Linac
55MeV 600W 50Hz

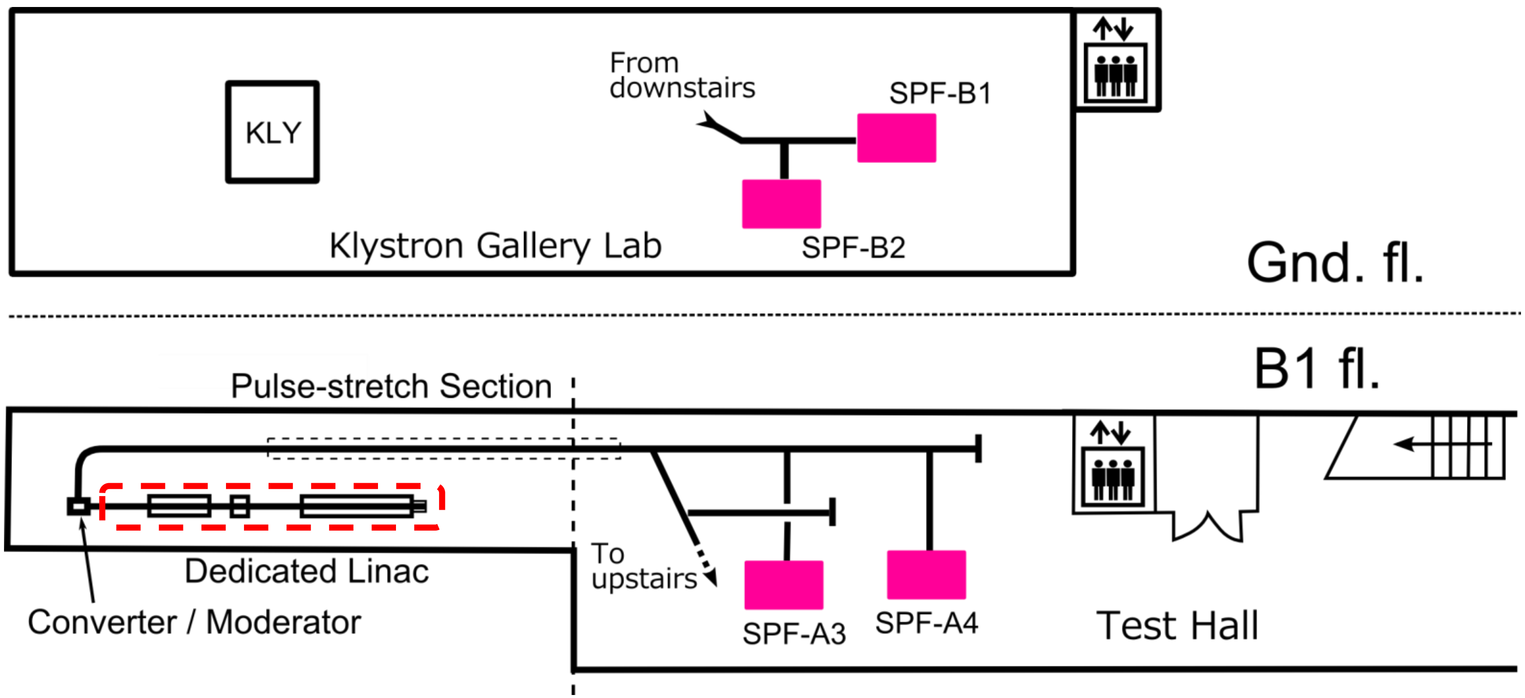
Long pulse mode
- 1μs width, 5×10^7 slow e^+/s
- 1×10^6 slow e^+/s after brightness enhancement

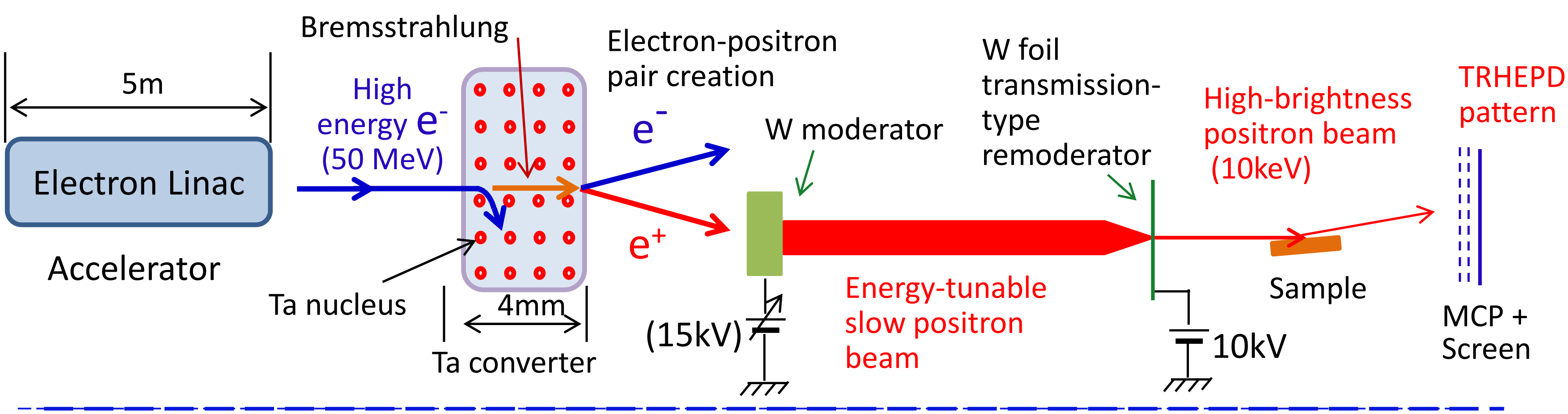
Short pulse mode
- 1-10ns width, 5×10^6 slow e^+/s

(Stations are not shown.)

K. Wada, et al., Eur. Phys. J. D 66, 37 (2012).
K. Wada, et al., J. Phys.: Conf. Ser. 443, 012082 (2013).

Plan view of Slow Positron Facility, KEK





(Example of TRHEPD experiment)

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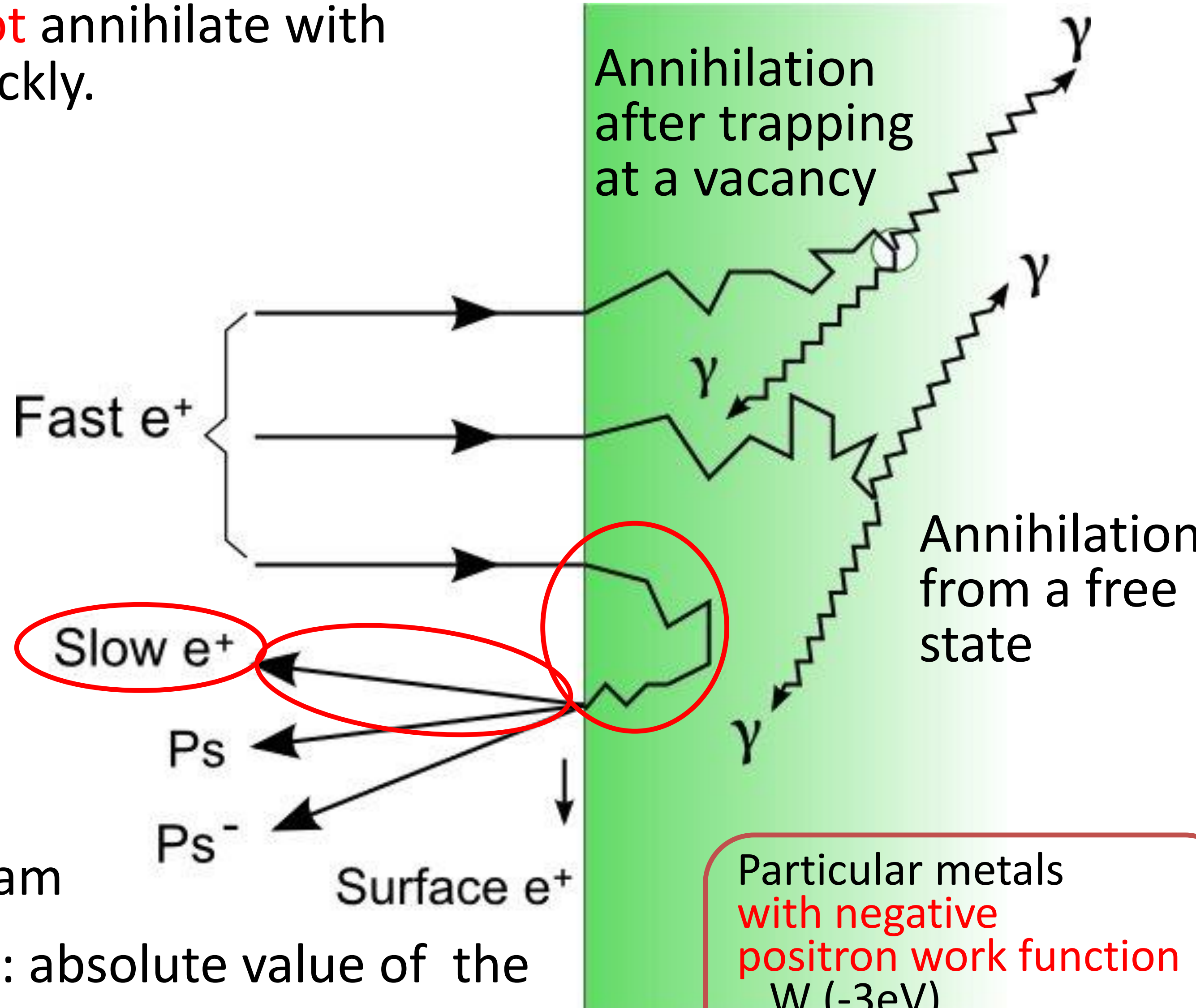
Preparation of monoenergetic slow positrons

Positrons do not annihilate with an electron quickly.

β^+ ray or Positrons from pair creation

Re-emission after thermalization

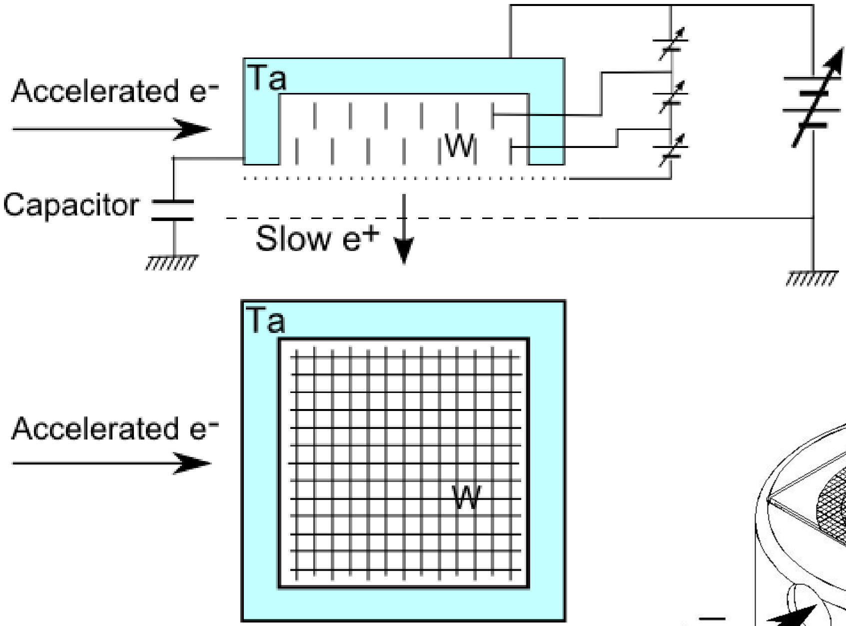
Good quality slow positron beam

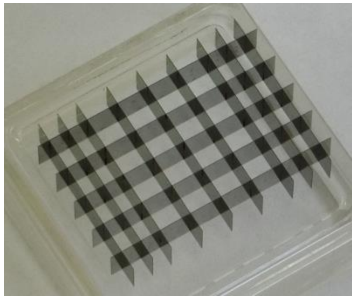


Particular metals with negative positron work function
- W (-3eV)
- Cu (-2eV)
- Ni (-1eV)

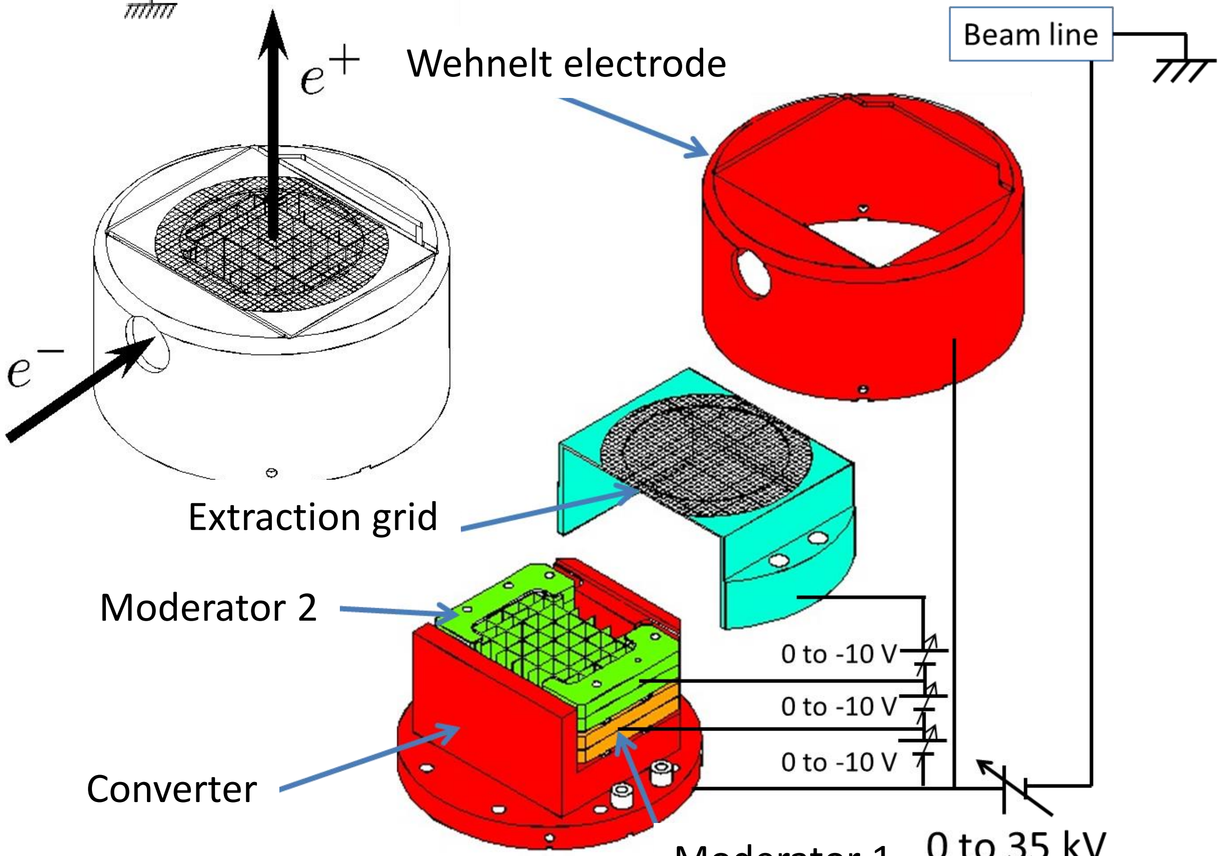
Emission energy : absolute value of the work function
Energy width : order of thermal energy

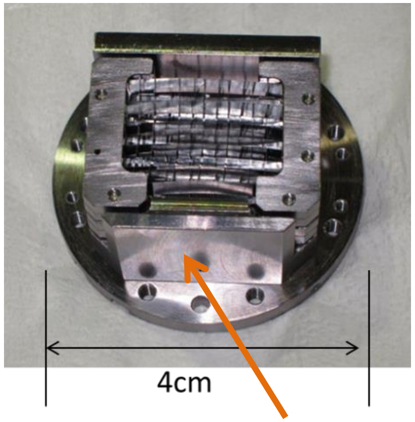
Converter/moderator for slow-positorn production





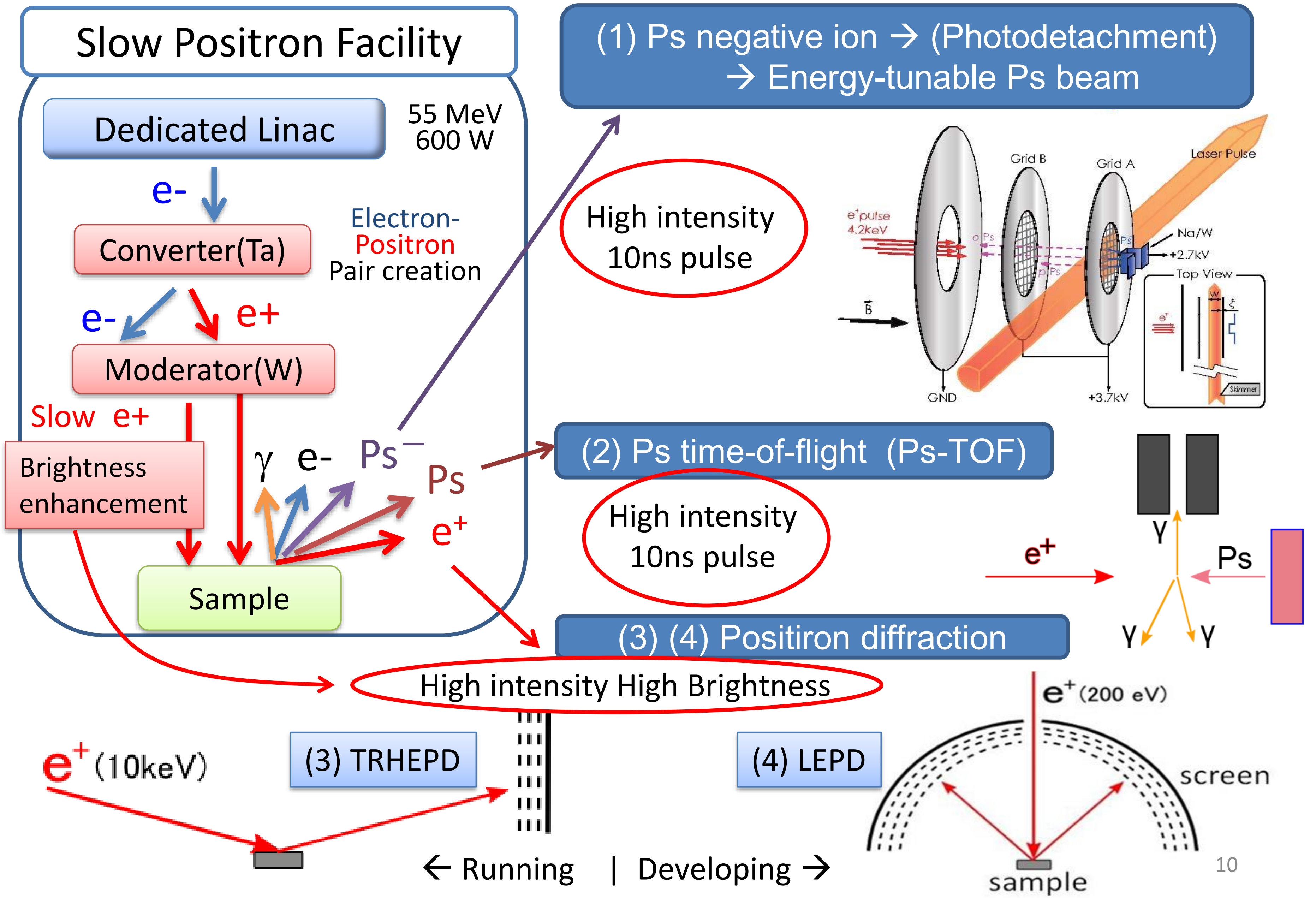
W moderator grid





Ta converter

Available 4 Stations at Slow Positron Facility, IMSS, KEK



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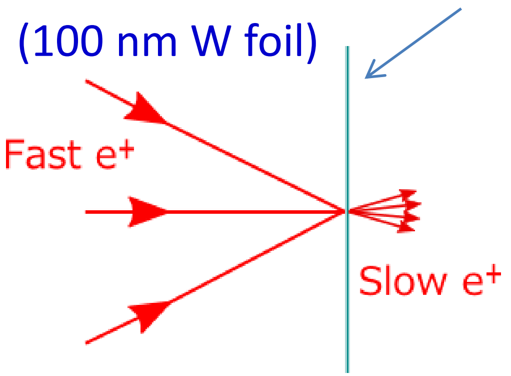
Brightness enhancemet by reemission of positrons from negative-work-function surface after thermalization

Brightness $$B = \frac{I}{Ed^2\theta^2}$$

I : Beam intensity, r : Beam radius
E : Beam Energy, θ : Beam divergence

Focusing on a remoderator foil.
Let dissipative force (thermalization) break the Leuvile's theorem.

Transmission-type remoderator (thin metal foil with negative positron work function) →



$B \rightarrow B \times 10^3$

$I \rightarrow I/10$

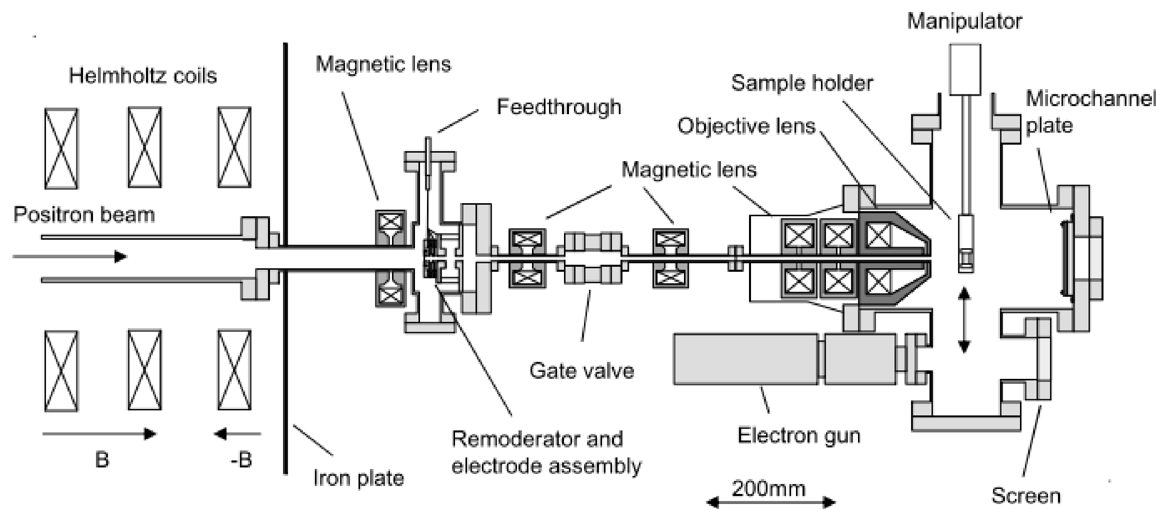
$r \rightarrow r$

$E = 5\text{keV} \rightarrow 3\text{eV}$

$\theta = \sim 50° \rightarrow \sim 10°$

With linac based intense slow positron beam:
Sample orientation by monitoring a TRHEPD pattern is now possible.

1hr for a good TRHEPD pattern
3hrs for a 00-spot rocking curve
← 1 min for a TRHEPD pattern for the rocking curve for an orientation



Brightness enhancement and TRHEPD chambers at KEK
M. Maekawa, K. Wada, *et al.*, Eur. Phys. J. D 68, 165 (2014) .

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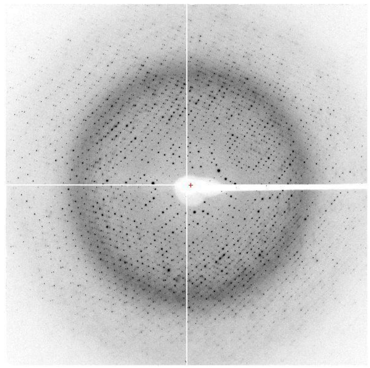
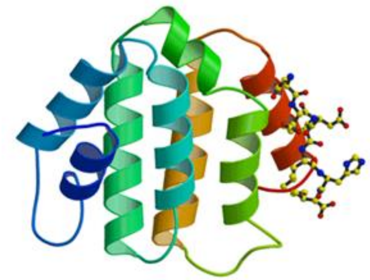
Atomic structure analysis by diffraction

X-ray diffraction pattern using synchrotron radiation

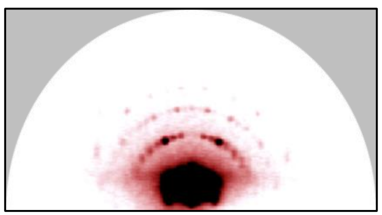


analisys using a computer



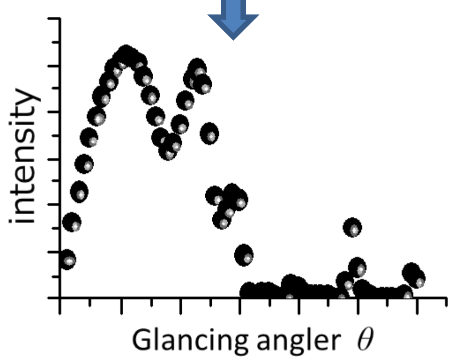
Protein

Positron diffraction pattern usingTRHEP



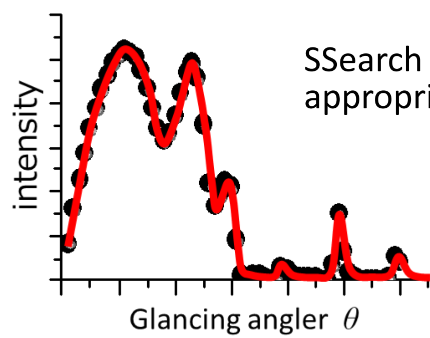
Diffraction data

Microscope (imaging method) observes these directly



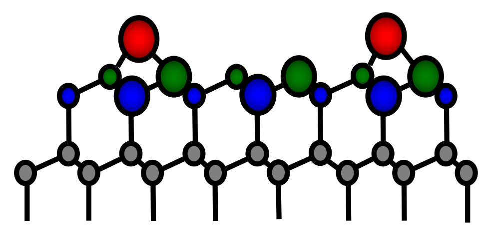
Rocking curve

analisys using a computer



SSearch for an appropriate structure

（Trial 6 error）



Si(111) (7×7)

Status of 3D and 2D structure analysis

Characteristics of materials
←Atomic structure (kinds of atoms and their detailed arrangements)
Structure determination independent from characterization is important.

It is widely practiced to use the methods to recognize the currigation of a surface or the crystal symmetry of the surface.

However, precise determination of the positions of the atoms is difficult.

3D materials (crystal of new material, proteins, etc.)

X-ray diffraction using synchrotron radiation is the standard method.

2D materials and surfaces

No standard method exists.

STM, AFM, SXRD, LEED, RHEED

（Basis and accomplishments)

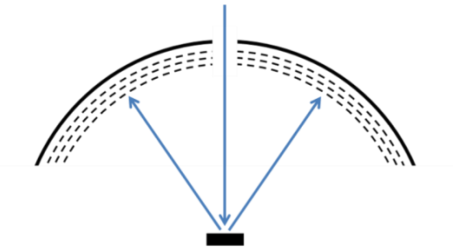
Positron Diffraction（TRHEPD in particular）is emerging to be a standard techinique.

But sufficient intensity of the beam is required, just as the case of X-rays → Use of accelerator for positron production resolves this difficulty.

Origin of the surface sensitivity of electron diffractions

LEED (low-energy electron diffraction)

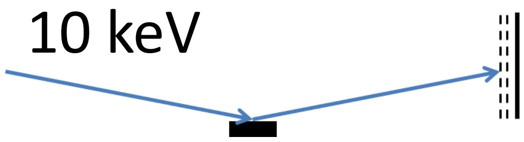
Because of low energy No



$$2d\sin\theta = n\lambda \text{ (Bragg condition)}$$

ex.:Thermal neutron satisfying the condition is not surface sensitive.

RHEED (reflection high-energy electron diffraction)



Because of the grazing angle incidence No

$$\sin\theta_{\mathrm{RHEED}} \sim \sin\theta_{\mathrm{LEED}}/10 \rightarrow \lambda_{\mathrm{RHEED}} \sim \lambda_{\mathrm{LEED}}/10 \rightarrow E_{\mathrm{RHEED}} \sim 100E_{\mathrm{LEED}}$$

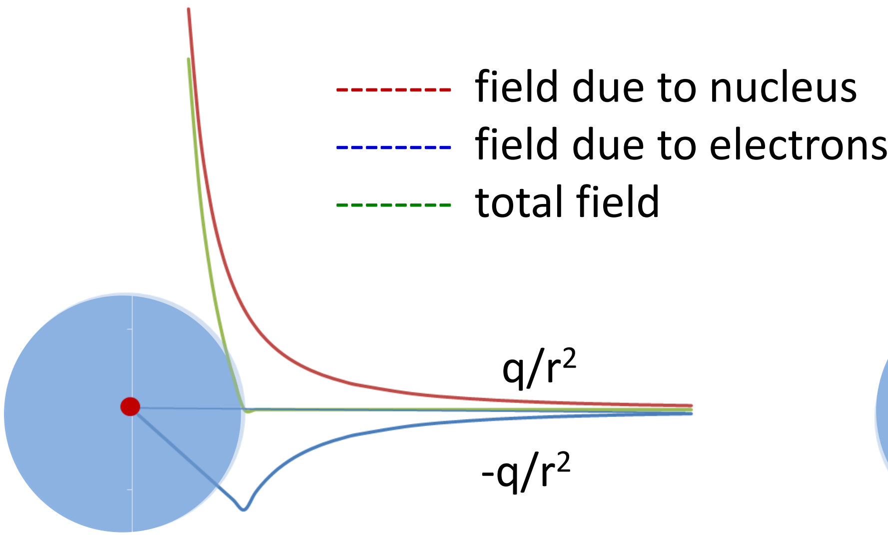
Inelastic scattering is the origin of the surface sensitivity, common to the electron and the positron diffraction.

Just as in Auger electron spectroscopy and photoelectron spectroscopy

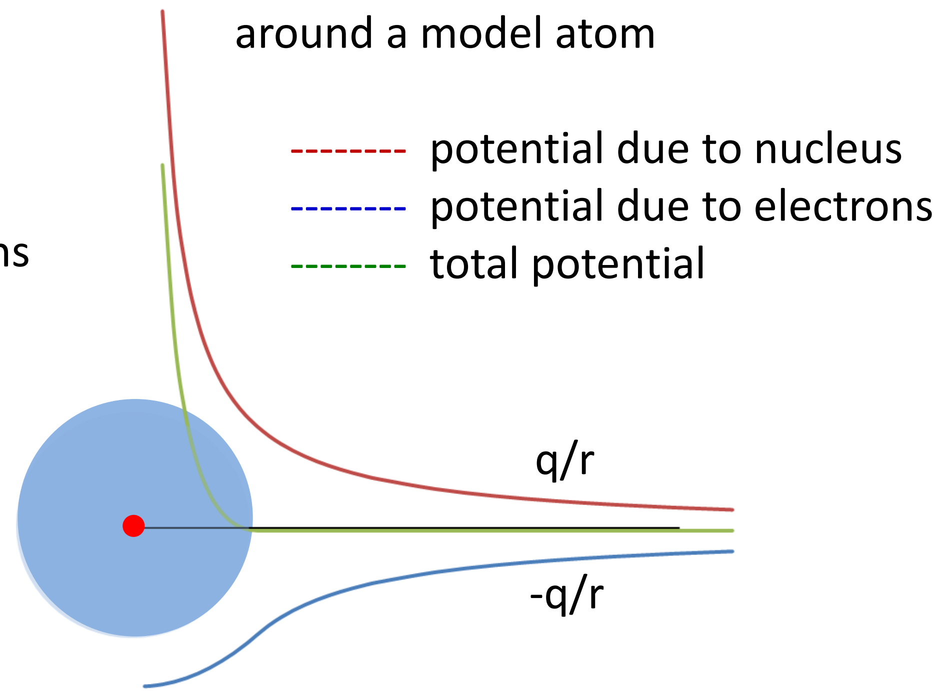
In addition, an origin unique to positron diffraction exists.

Electrostatic field and electrostatic potential around an atom

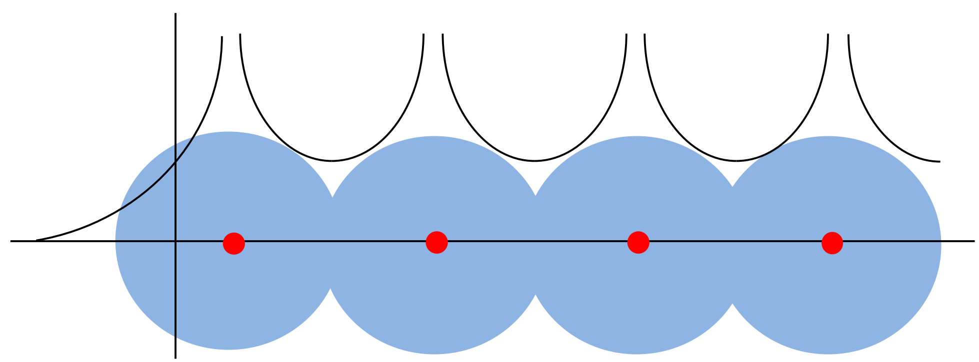
Electric field
around a model atom



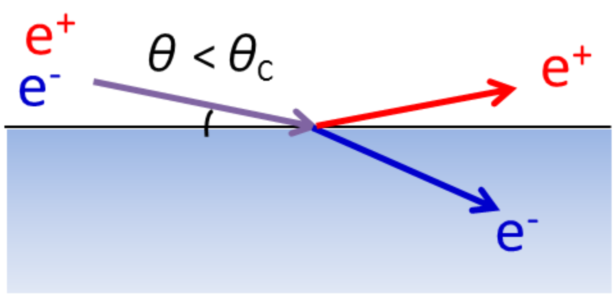
Electrosotatic potential
around a model atom



Electrostatic potential
In every solid



Crystal potential and total reflection of positron



$$E_0 = \frac{\hbar^2 k_0^2}{2m} = \frac{\hbar^2 k_{0\parallel}^2}{2m} + \frac{\hbar^2 k_{0\perp}^2}{2m} = E_{0\parallel} + E_{0\perp}$$

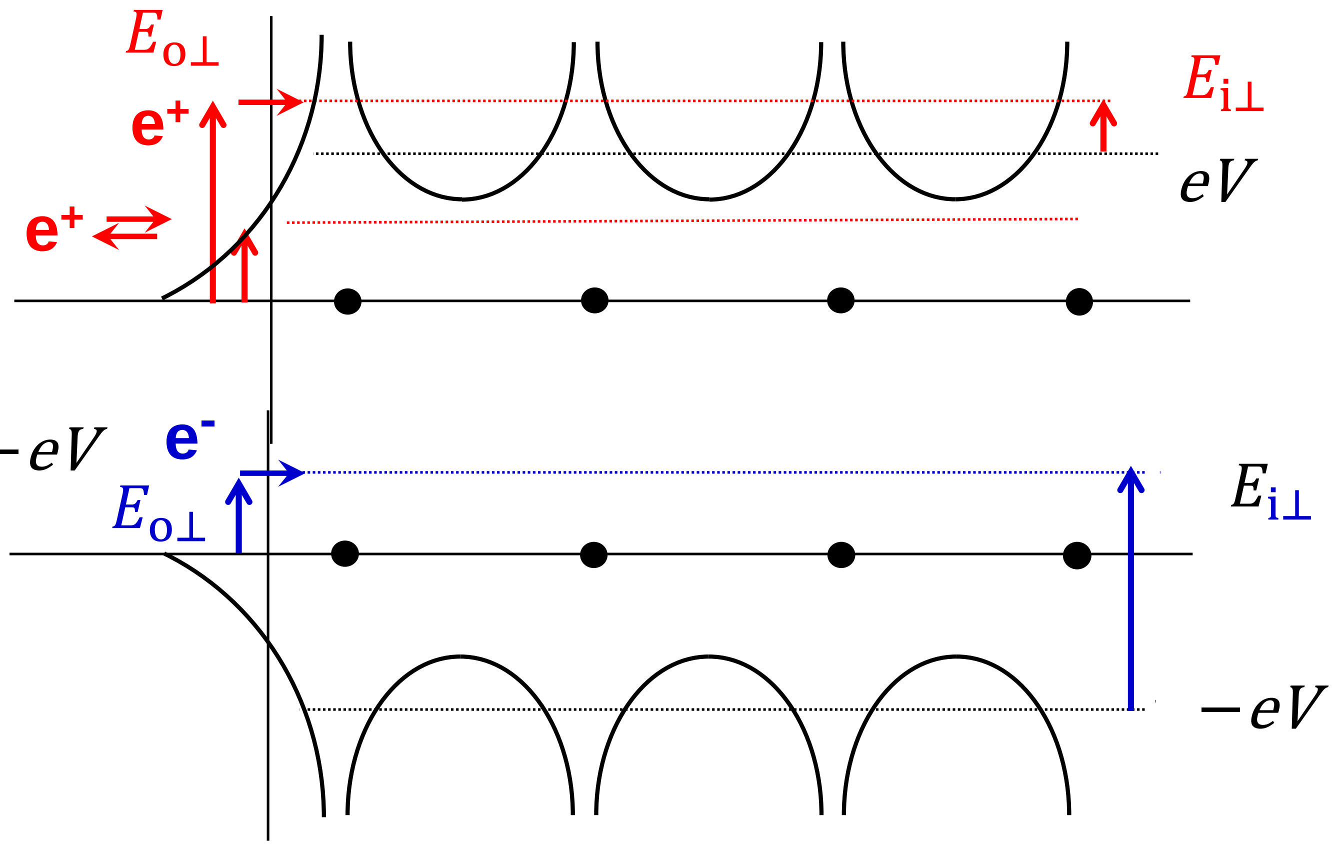
$$E_{0\perp} = \frac{\hbar^2 k_{0\perp}^2}{2m} = \frac{\hbar^2 k_0^2 \sin^2\theta}{2m} = E_0 \sin^2\theta$$

$$E_{0\perp} = eV = E_0 \sin^2\theta_c \rightarrow \theta_c = \sin^{-1}\sqrt{eV/E_0}$$

$E_{o\perp} > eV$

$E_{o\perp} < eV$

$E_{o\perp} > 0 > -eV$



Glancing angle dependence of the paths of positron and electron and their surface sensitivity

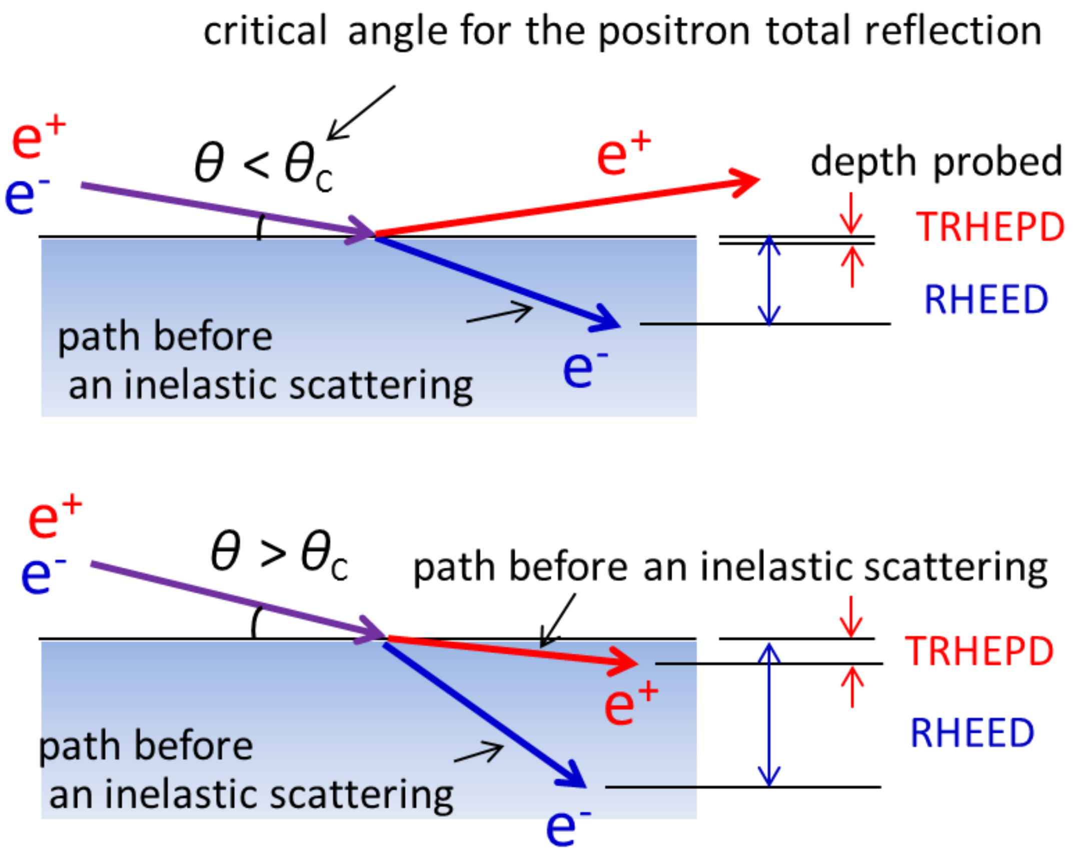
Origin of the surface sensitivity common for electron and positron

-→inelastic scattering

Origin of the surface sensitivity characteristic to positron

-→total reflection
refraction toward
the surface

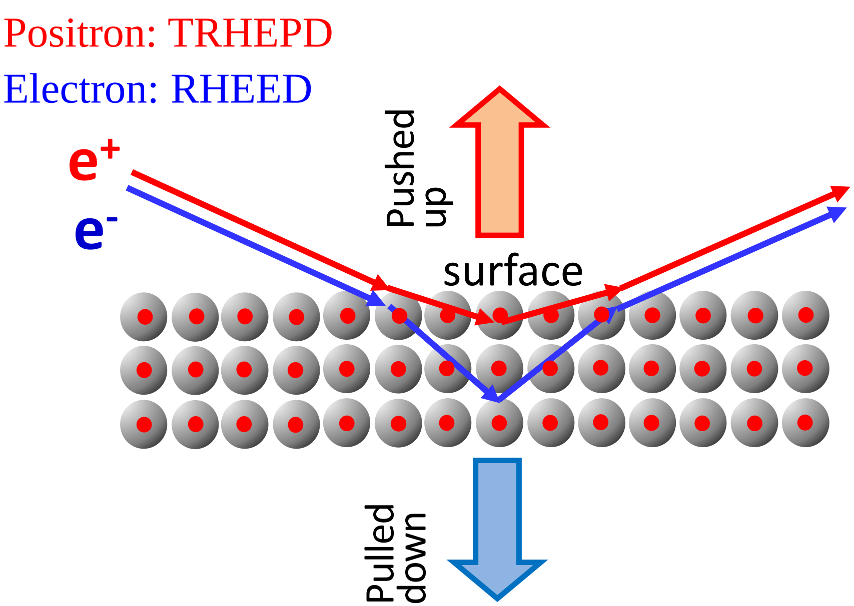
TRHEPD : $\theta < 6^\circ$
toral rfl. : $\theta c = 2^\circ - 3^\circ$

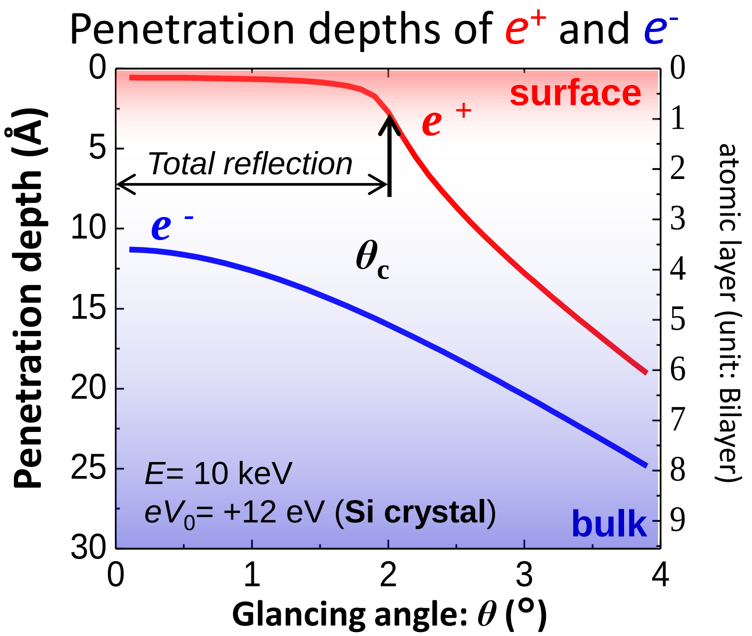


Positron in the only quantum mechanical particle for which angular range for the total-reflection and the Bragg-diffraction overlap.

Data usually include those not satisfying the total reflection condition
TRHEPD is the name of the method.

THREPD is an ideal method for topmost surface and immedeate subsurface

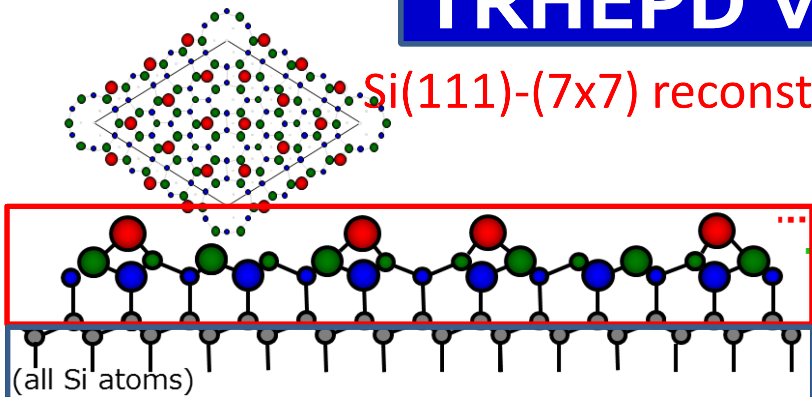




Features of TRHEPD

1. Positrons undergo pure or ideal total reflection.
2. The critical angle for total reflection θ_C (2°-3°) lies in the middle of the TRHEPD measurement region (→ unique property of the positron).
3. $\theta_{in} < \theta_C$: positrons are totally reflected and see the topmost surface only.
4. $\theta_{in} > \theta_C$: positrons also see the immediate subsurface.
5. Width of interest from the surface is adjustable with varying θ_{in}.
6. No background from the deeper, bulk part at all.

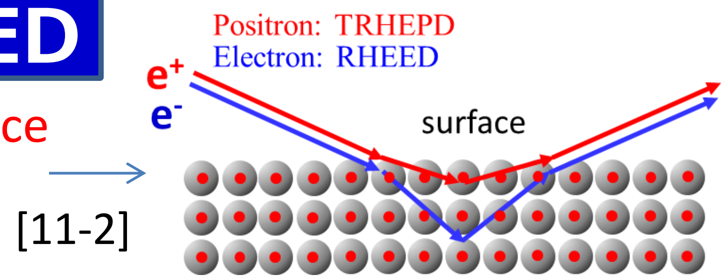
TRHEPD vs RHEED



Si(111)-(7x7) reconstructed surface

reconstructed and relaxed

unrelaxed bulk



Exp: 10keV $\theta = 1.3^\circ$

Calc:

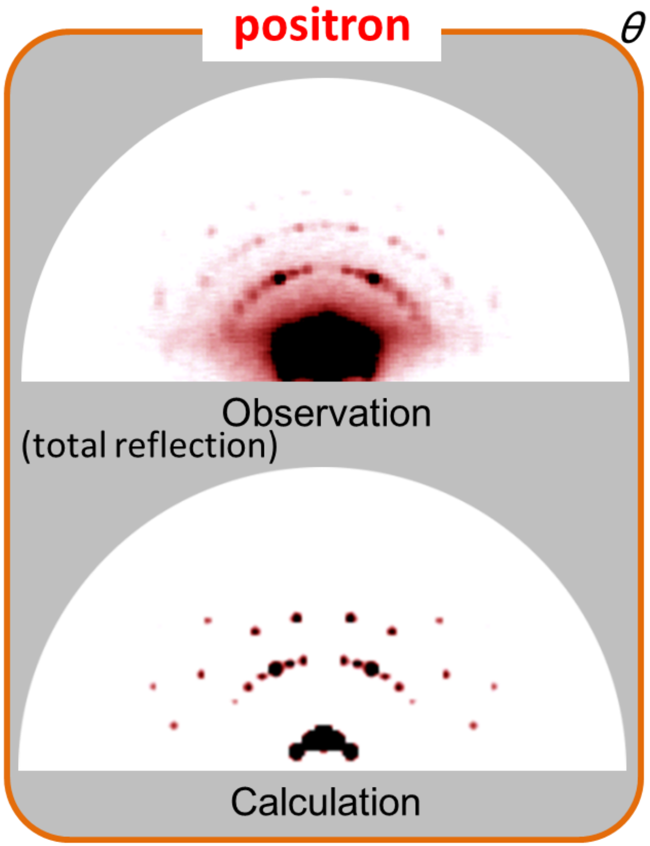
Same code

(only signs of the charge different)

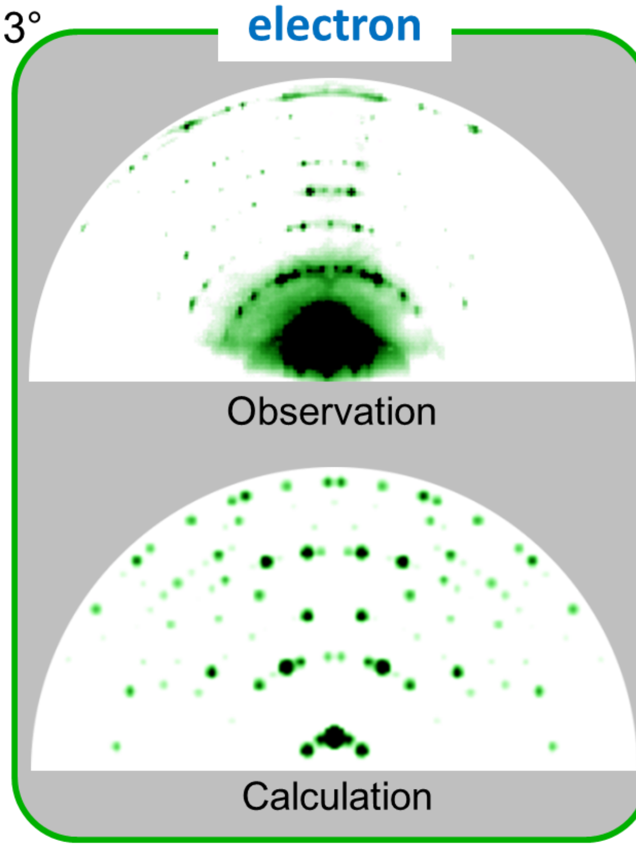
Structural model

down the 3rd layer: reconstructed and relaxed (literature)

below: unrelaxed bulk structure (literature)



$\theta = 1.3^\circ$



Y. Fukaya, et al., Appl. Phys. Express 7, 056601 (2014).

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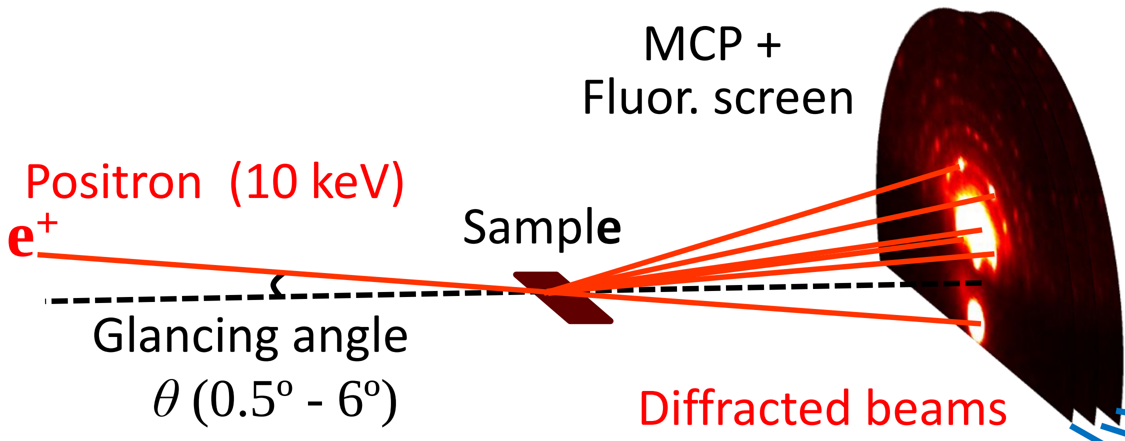
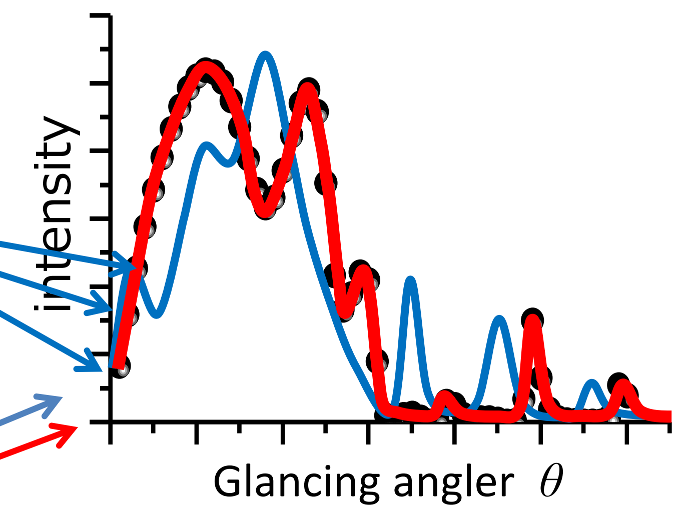
Rocking curve analysis of the surface structure

A. TRHEPD measure ment

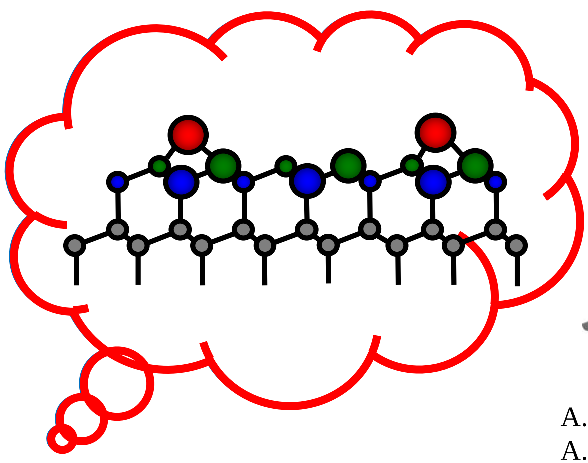


Obtain a rocking curve from patterns for various θ



B. Calculation of TRHEPD rocking curve

Surface structure model



Calculation with
full dynamical theory

A.Icimiya and Cohen:"RHEED" (Cmbridge U. P.)
A. Ichimiya, *JJAP* (1983)

Compare goodness of fit

- **R-factor**

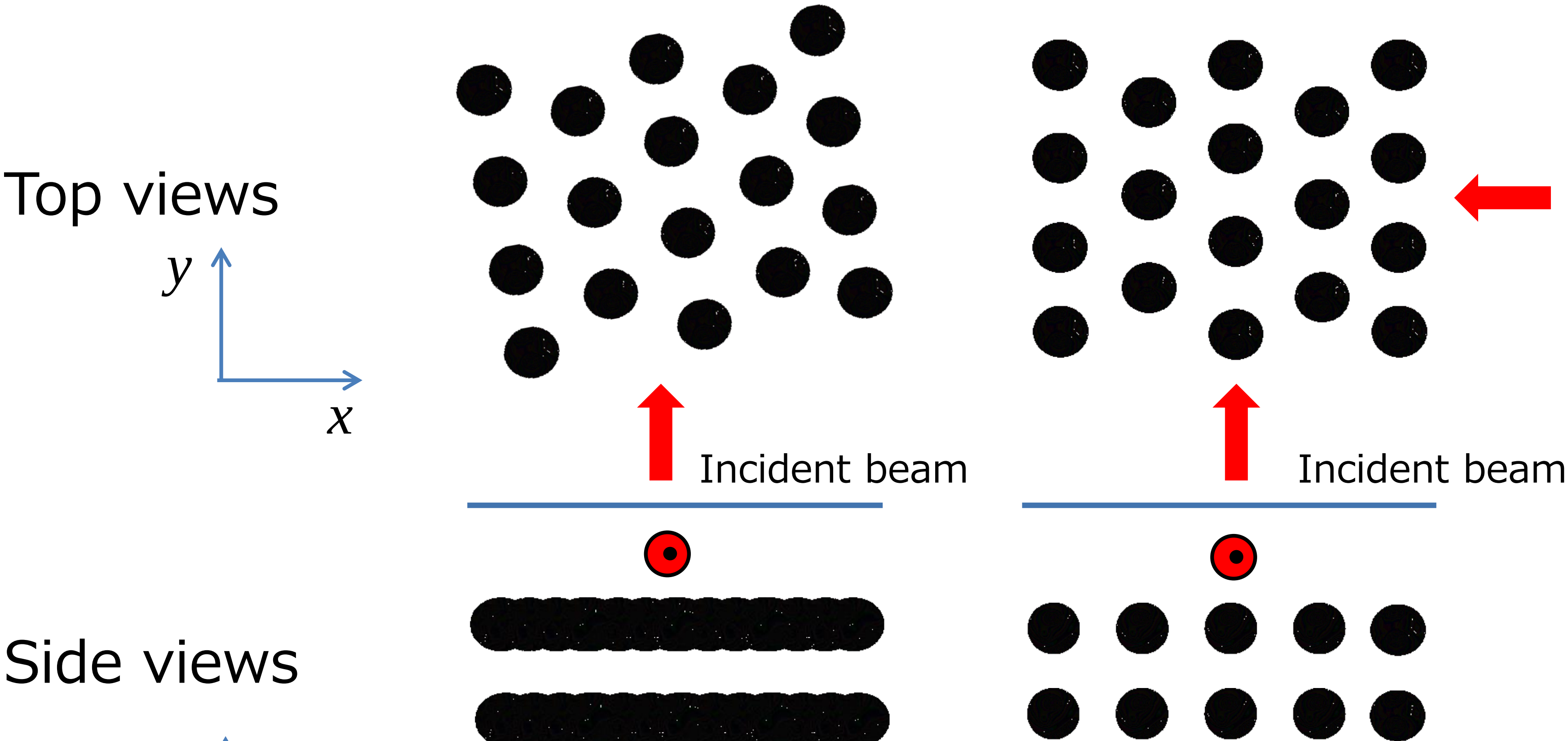
$$R = \sqrt{\sum_{\theta} \left(I_{\theta}^{\mathrm{exp}} - I_{\theta}^{\mathrm{cal}}\right)^2}$$

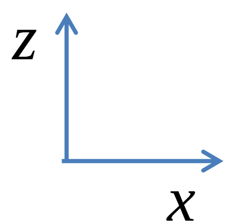
$$\sum_{\theta} I_{\theta}^{\mathrm{exp}} = \sum_{\theta} I_{\theta}^{\mathrm{cal}} = 1$$

Conditions for rocking curve mesaurements

One-beam condition
Incident from non-symmetrical direction

Many-beam condition
Incident from non-symmetrical direction

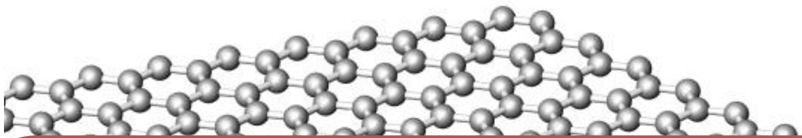




Only sensitive to the Coordinate perpendicular to the surface and atomic density

Sensitive to the coordinate in plane also

2D atomic layer materials



Graphene(C)

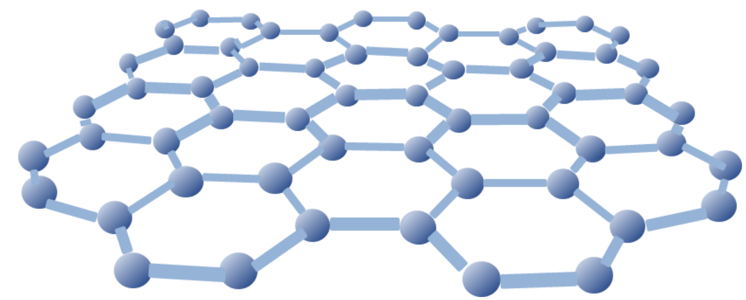
- Single layer of C atoms
- High electron mobility, thermal conductivity, stifness
- Prospective new material for energy-saving, fast devices
- Usually synthesized on a substrate

Silicene (Si) ･Germanene(Ge)

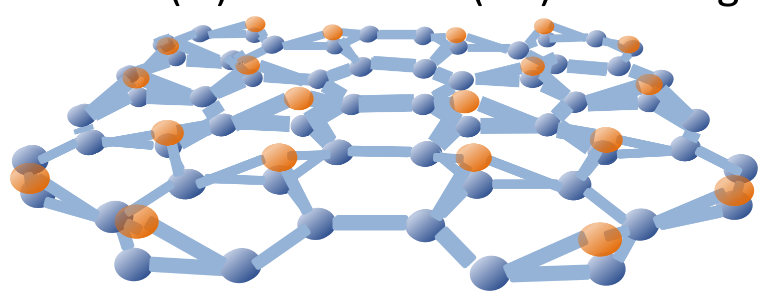
- Graphene-like material
- Wilth buckling
- Electronic properties depend on amount of buckling

周期｜族	13	14	15
2	B	C	N
3	Al	Si	P
4	Ga	Ge	As
5	In	Sn	Sb
6	Tl	Pb	Bi

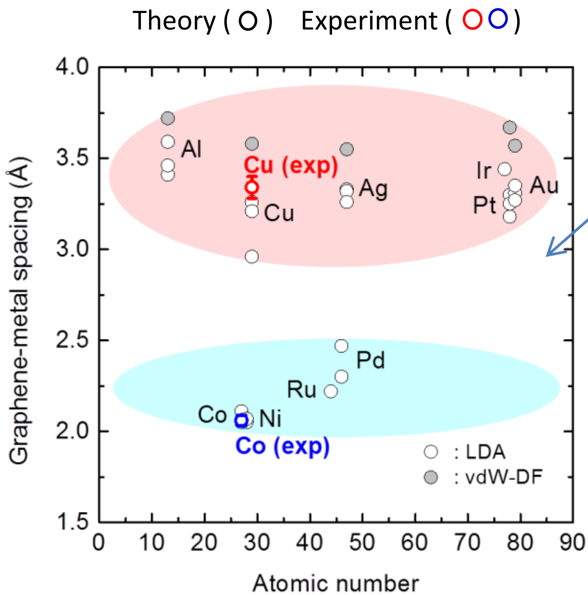
Graphene(C)：planar



Silisene (Si) Germanen (Ge): buckling



graphene on Cu(111) and Co(0001)



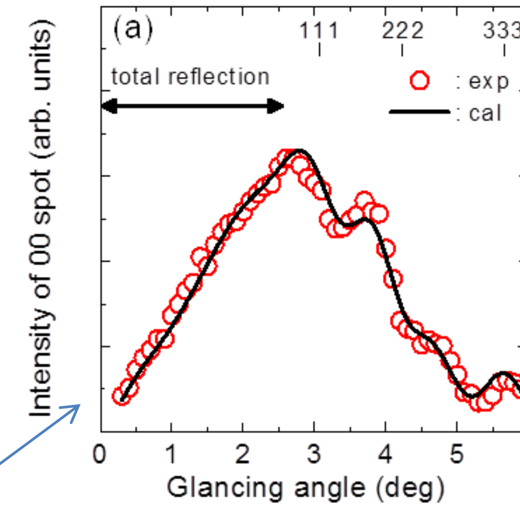
Giovannetti, Phys. Rev. Lett. (2008).
Vanin, Phys. Rev. B (2010).
Gong, J. Appl. Phys. (2010).

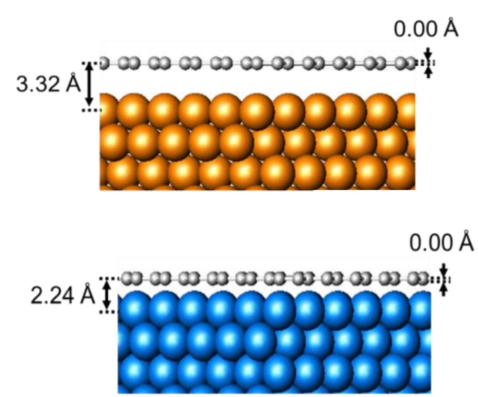
Theory
Graphene-substrate distance is classified depending on the kind of metal substrate.

Charge transfer from the substrate to graphene depends on the Graphene-substrate distance and affect the band structure

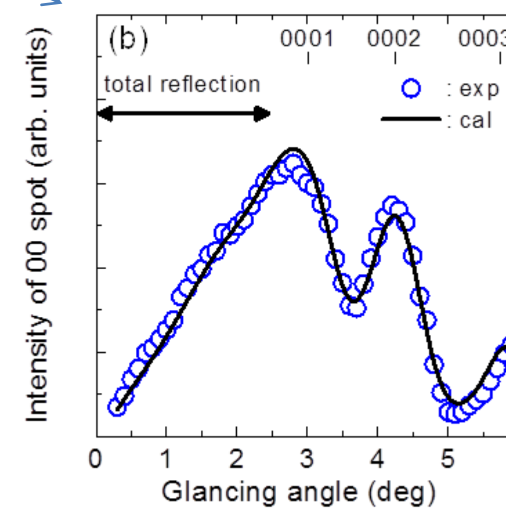
TRHEPD
Measured
Graphene-Cu(111) distance
Graphene-Co(0001) distance
Verified theoretical prediction

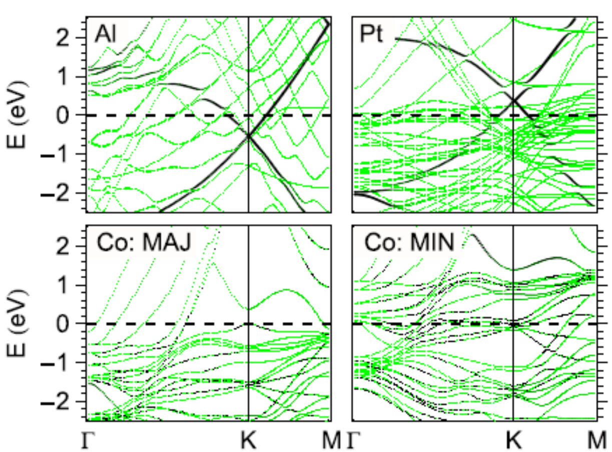
Graphene on Cu(111)





Graphene on Co(0001)

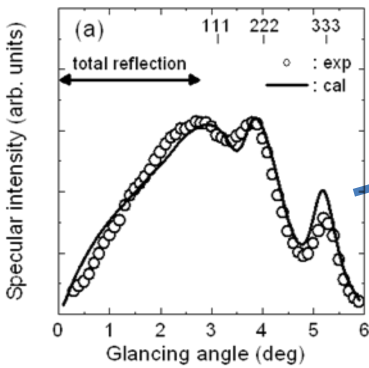




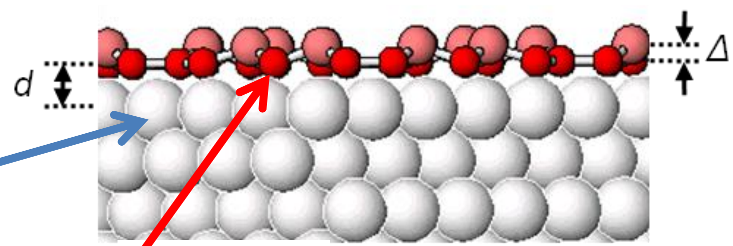
Giovannetti, Phys. Rev. Lett. (2008).

Y. Fukaya, et al., *Carbon* **103** (2016) 1.

Silicene on Ag(111) suraface



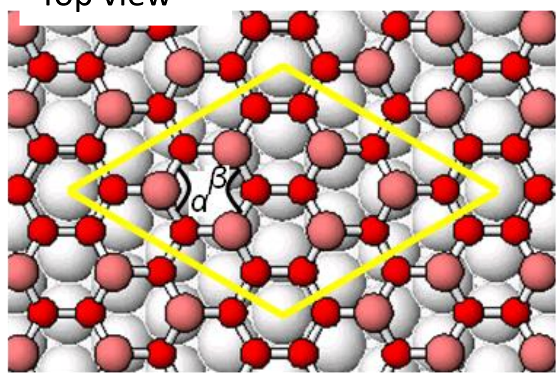
(one -beam condition)

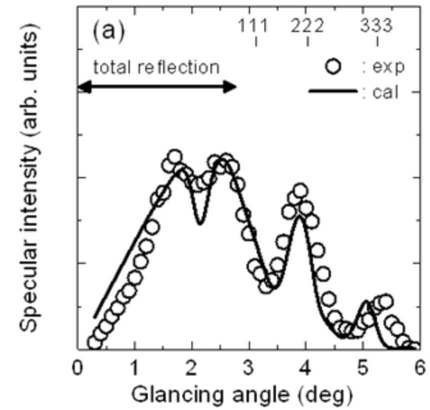


Side view

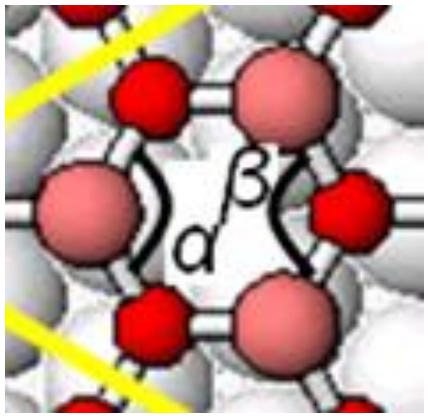
Red: Si atoms
Grey: Ag atoms

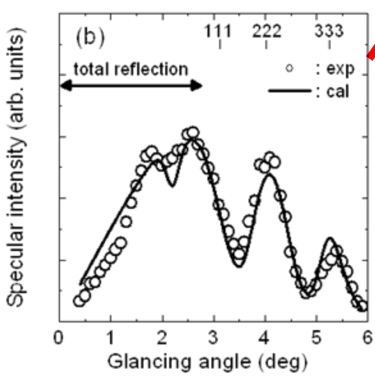
Top view





(Many-beam comndition)





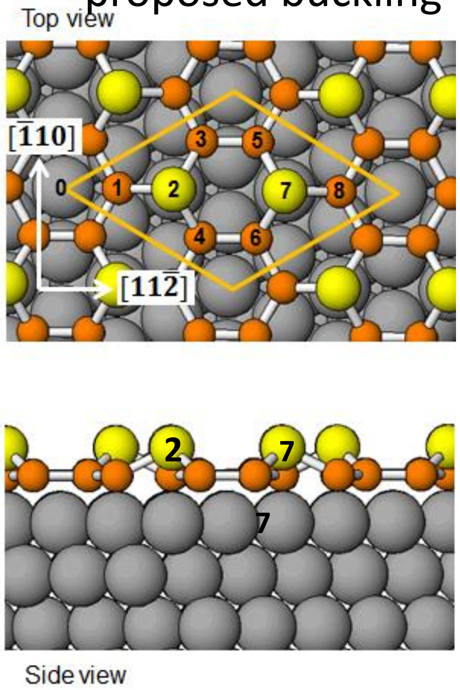
(one -beam condition)

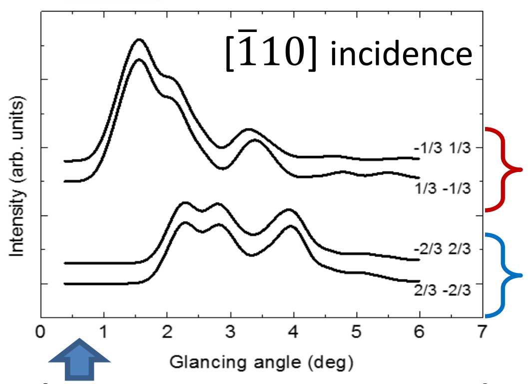
	Δ (Å)	d (Å)	α (°)	β (°)
RHEPD	0.83	2.14	112	119
Theory	0.78	2.17	110	118

TRHEPD: Fukaya et al., Phys. Rev. B 88, 205413 (2013)
Theory:: Vogt *et al*., Phys. Rev. Lett. **108**, 155501 (2012).

Germanen on Al(111) : Buckling is asymmetric.

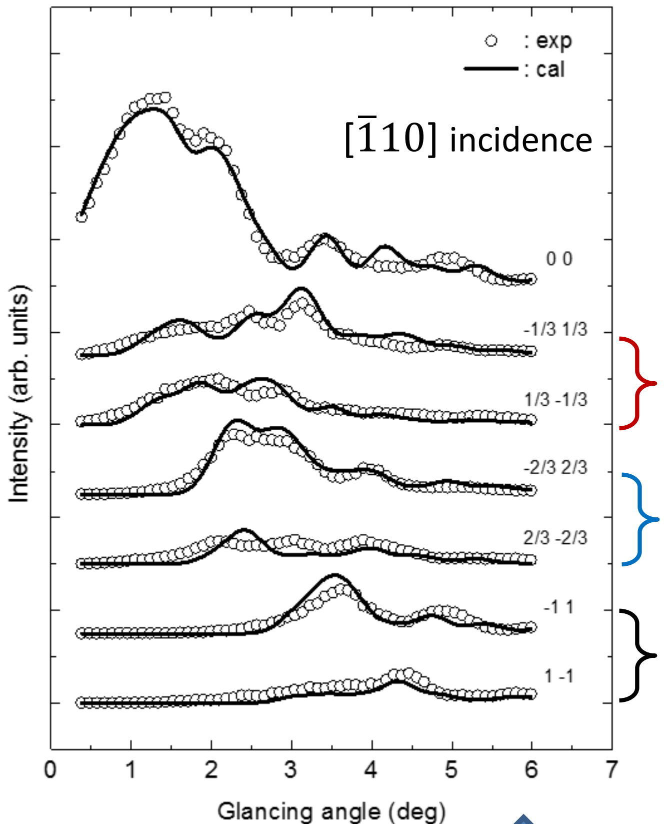
Previously proposed buckling





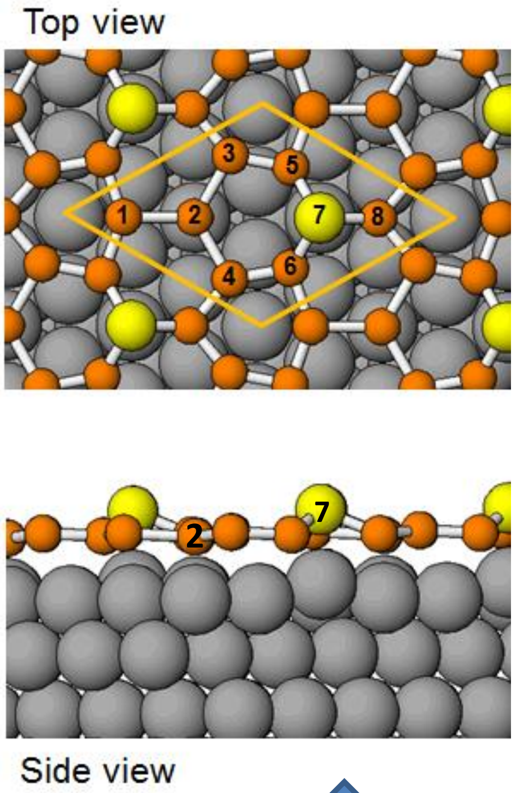
Rocking curves expected from this model, which disagree with the data.

Experimental data and fitted curves



Measured rocking curves and fitted curves.

Buckling proposed by TRHEPD

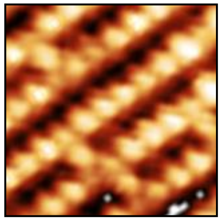


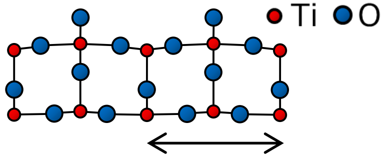
Structure proposed to explain the TRHEPD data.

Y. Fukaya, et al. *2D Materials* 3, 035019 (2016).

Structure analysis of rutile-TiO_2(110)-(1×2) surface

Rutile-TiO_2(110)(1×1) surface



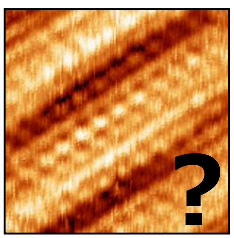


(1×1) unit

already establishied (most stable)

annealing at ~1200K

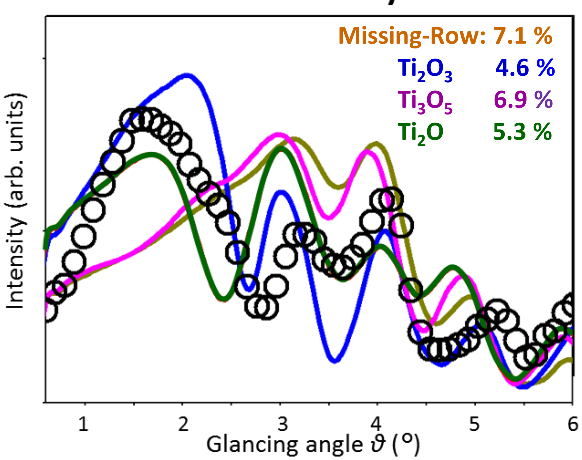
Turns into (1×2) surface

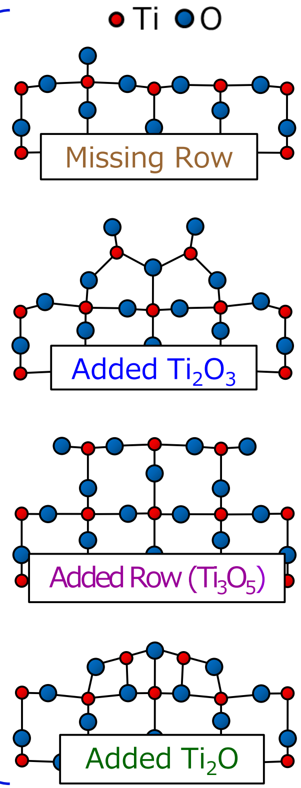


Many possible structures proposed.

No proposed structure explained TRHEPD data.

One-beam analysis





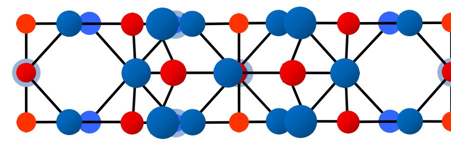
TiO_2 (titania)

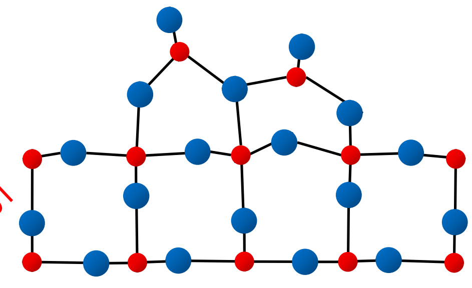
Photocatalyst, Gas sensor,
Catalyst support
Standard substance
for metal-oxide catalysis
Single-crystalized TiO_2 surface
(jmpotant for catalytic process
studies at atomic scale)

allowing for asymmetric structure

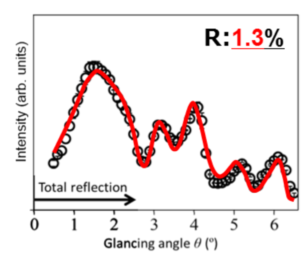
Wang et al. (Oganov group) Phys. Rev. Lett. 113 (2014) 266101 (USPEX code)

Structure determined by TRHEPD

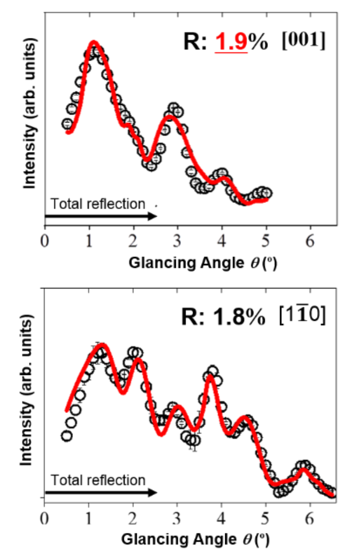




One-beam analysis



Many-beam analysis



Asymmetric-Ti_2O_3 Model is correct.

Controversy for 30 years settled

I. Mochizuki et al. PCCP **18** 7085 (2016)

Chemistry World : https://www.chemistryworld.com/research/9591.article

History of (T)RHEPD

- 1992: Proposal and Basic Theory of RHEPD by A. Ichimiya
  (an expert in RHEED) , Solid State Phenom. 28/29 , 143.
- 1998: First experimental data published
  (A. Kawasuso and S. Okada: Phys. Rev. Lett. 81, 2695.)
- 2000- : Many publications (about 40) by Kawasuso group (JAEA)
  (K. Hayashi, Y. Fukaya, et al.)
  10^3-10^4 slow-positrons/s (with ^{22}Na) for RHEPD.
- 2010: RHEPD station moved from JAEA to KEK:
  10^6 slow-positrons/s (with Linac) for RHEPD.
- 2012: Brightness Enhancement with remoderation of the positrons. Construction of a new station (→ TRHEPD)
- 2014: Station at KEK is still the only one of the kind in the world.
  → We encourage other positron facilities to implement positron diffractions (TRHEPD and LEPD).
  Technical University Munich is now constructing one.

Recent inquiries from outside positron community

April 2016
We are trying to make extremely thin layer of some oxide. We already succeeded in a few kind of thin layer (about 0.5 nm thick)。We want to use TRHEPD to analyze their detailed structures.

April 2016
We are investigating superconductivity of a system of bilayer graphene with intercalated alkali metals. We want to analyze the structure of the bilayer graphene, that after intercalation, and that after removal of the intercalated metals.

September 2016
We are trying to make a novel monatomic 2D material. It appears that we already succeeded in making one, but we have no way to identify the structure. TRHEPD must be capable of doing it.

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- Colaborators
- Creation of slow positron
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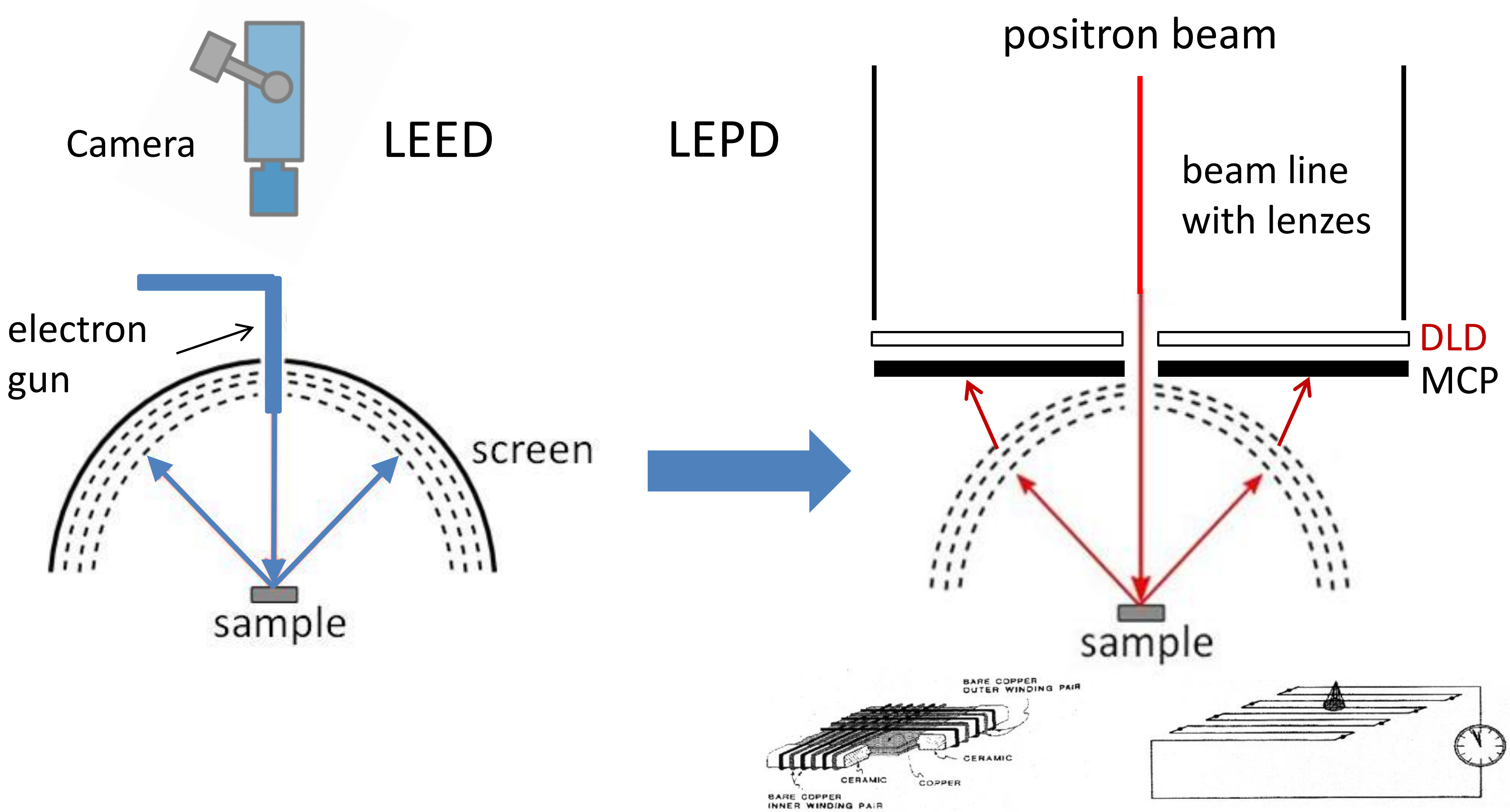
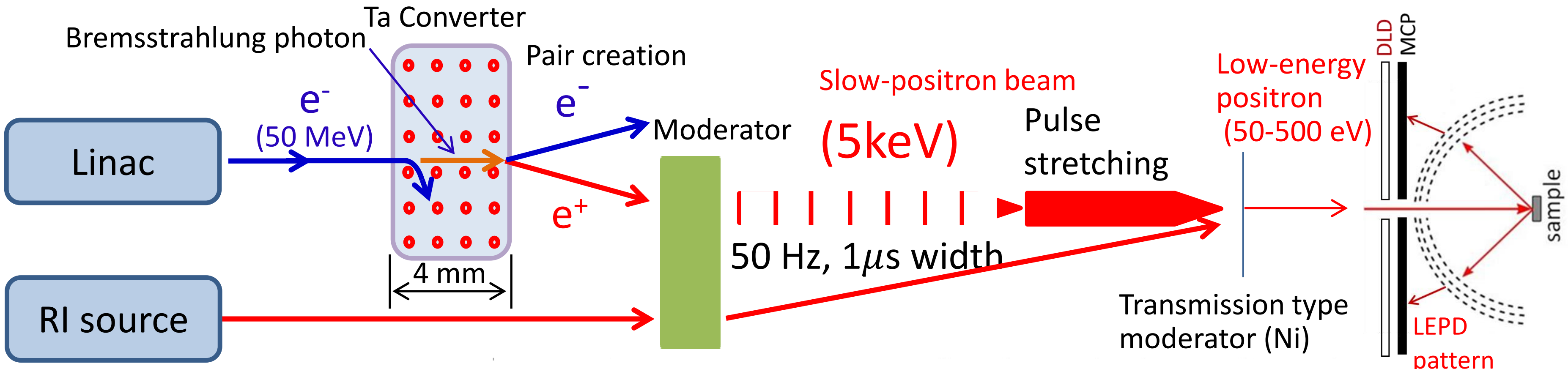
- TRHEPD
- LEPD
- Puls-stretching
- Ps^- (Ps negative ion)
- Ps-TOF

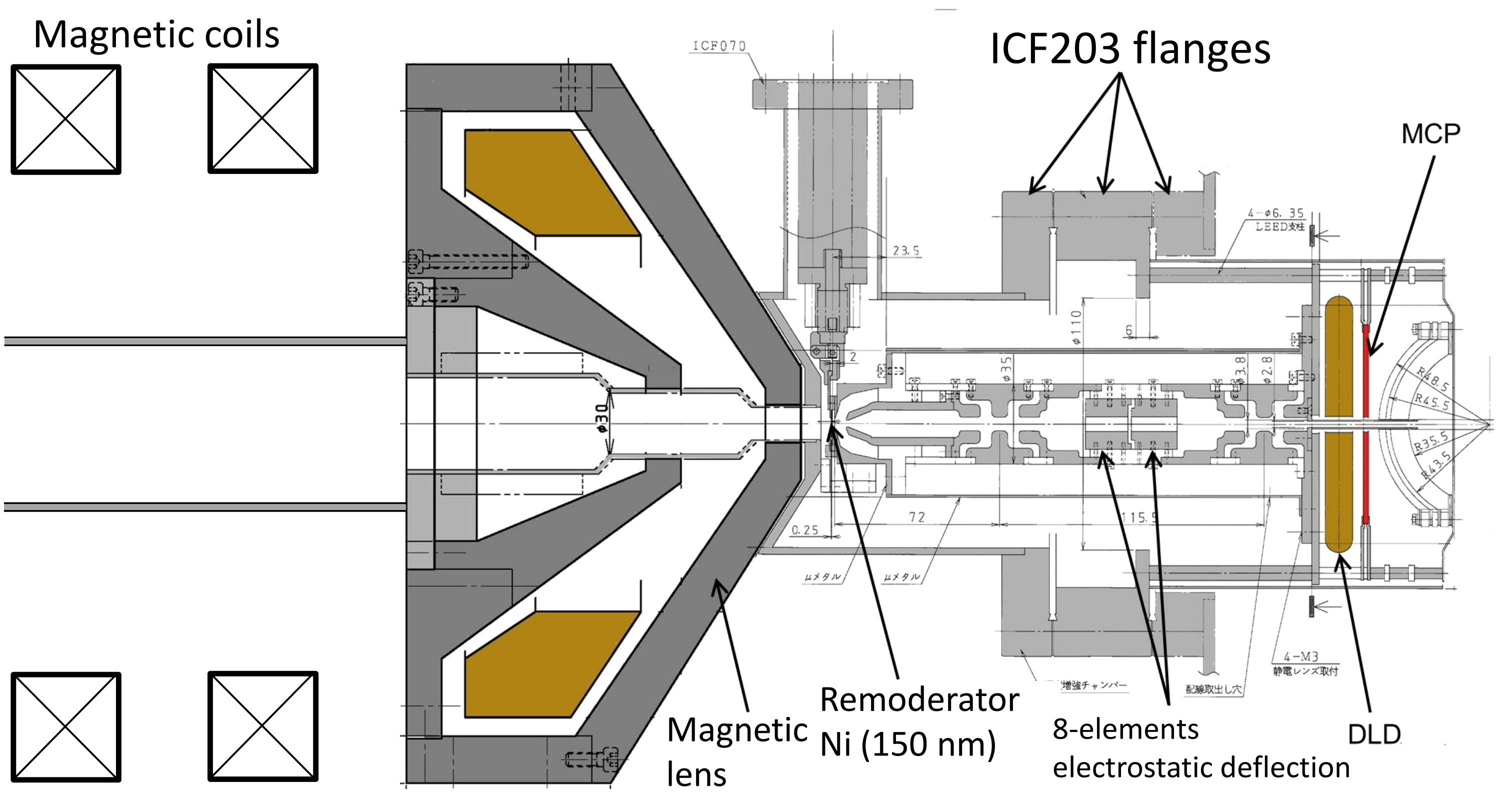
Needs for pulse-stretching system

- LEED patterns are recorded by a CCD camera from the back.
- LEPD system is not compatible with similarly using a camera because of the interaction of the camera and beam line containing lenzes.
- Use of delay-line-detector (DLD) to record LEPD patterns.
- DLD cannot process too many particle in a short pulse .

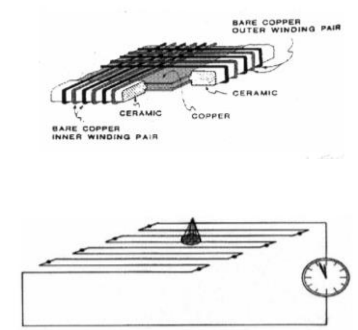


LEPD system at KEK and QST Takasaki

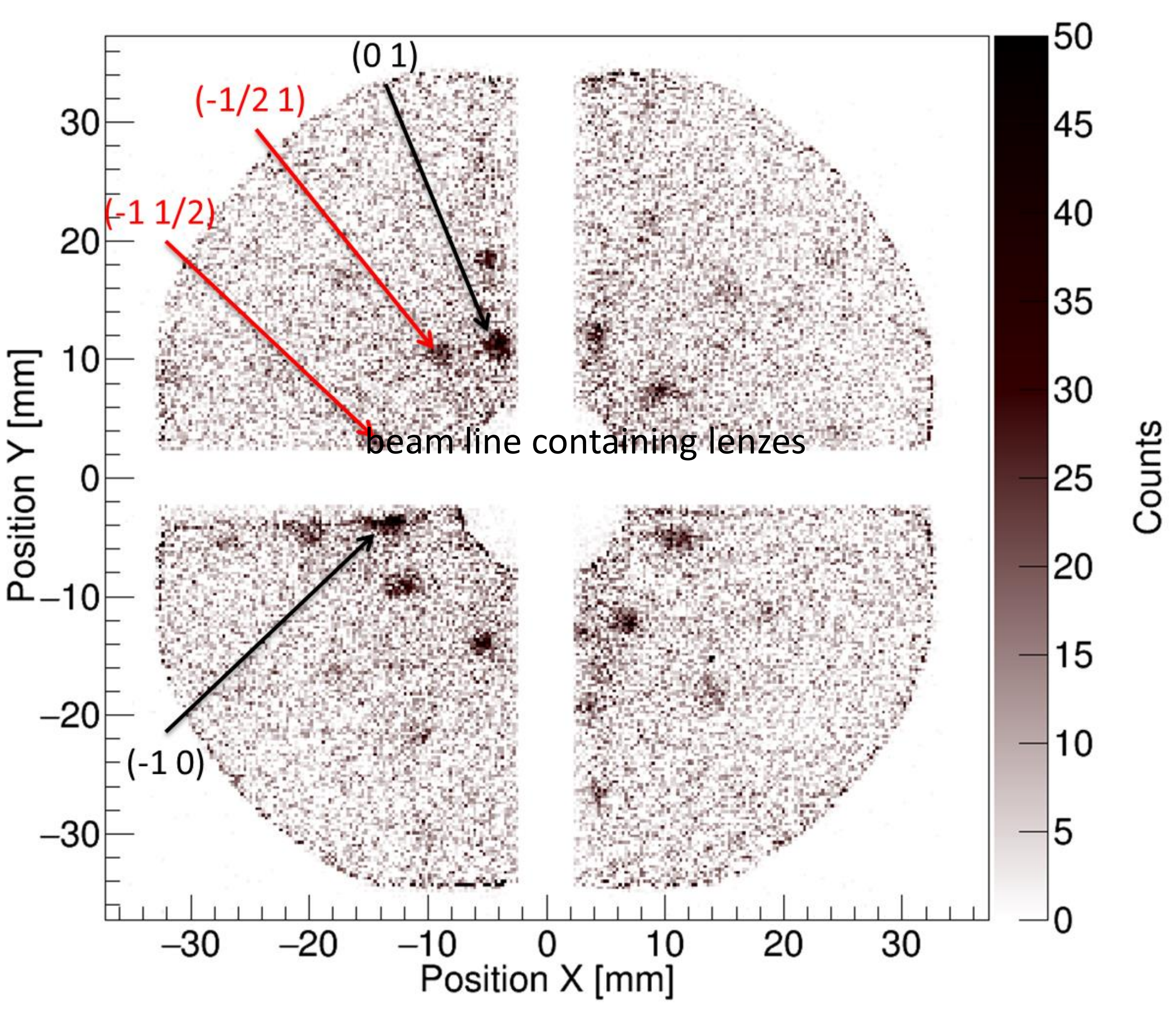




Delay-line detector (DLD)



Our first LEPD pattern from Ge(001)-(2×1) at 140 eV



Scattering factor for positron and electron

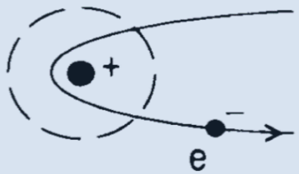
Positron

$R_t = Ze^2/E_p$

Classical turning point at large radii due to the repulsive force from the nucleus.

Weak LS coupling

Electron

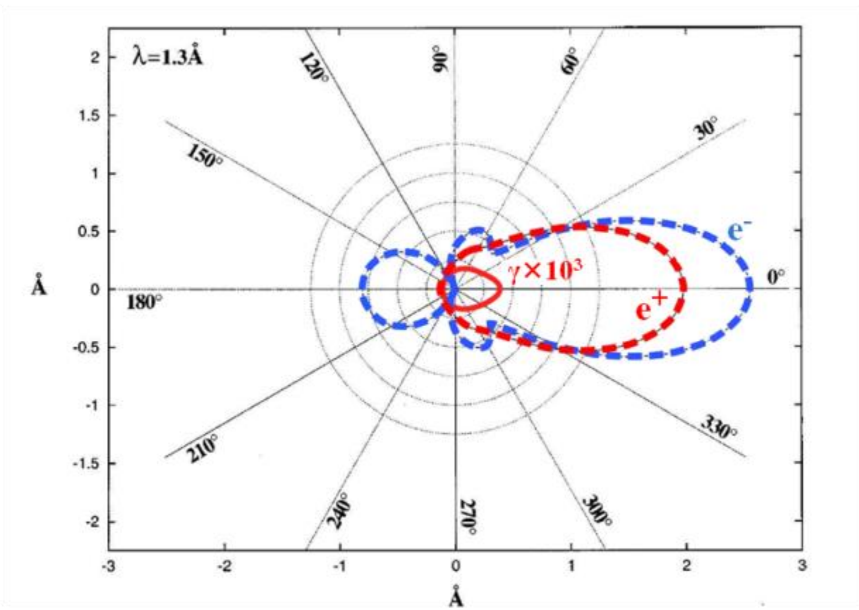


Tong et al., Phys. Rev. Lett. **69**, 3654 (1992)

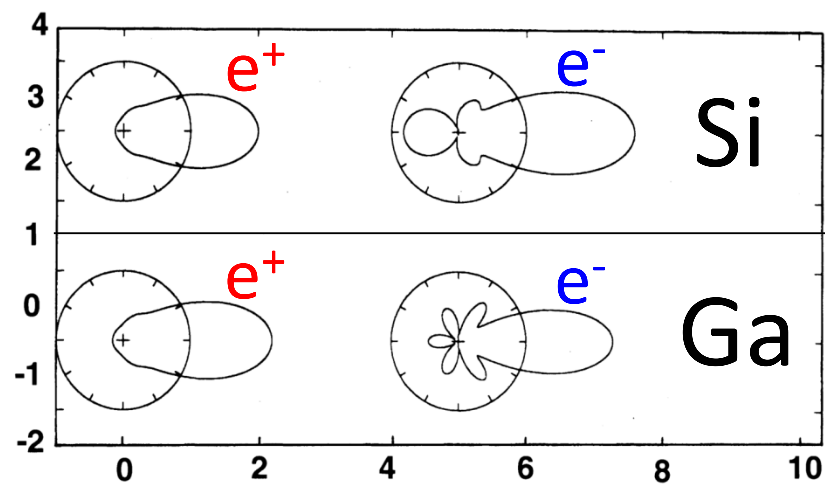
Electron is accelerated toward nucleus resulting in strong relativistic effects.

Strong LS coupling

Angular dependence os $]f(\theta)]$ for Si



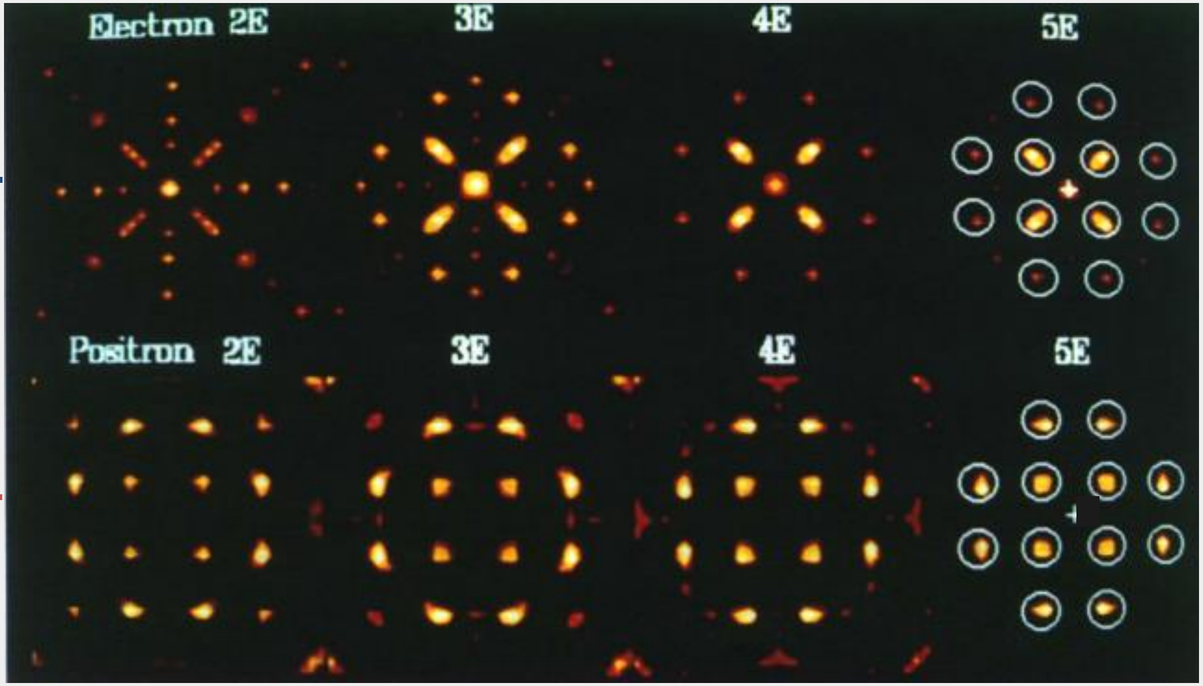
Angular dependence os $]f(\theta)]$ for Si and Ga



LEPD holography by using adatoms as beam splitters

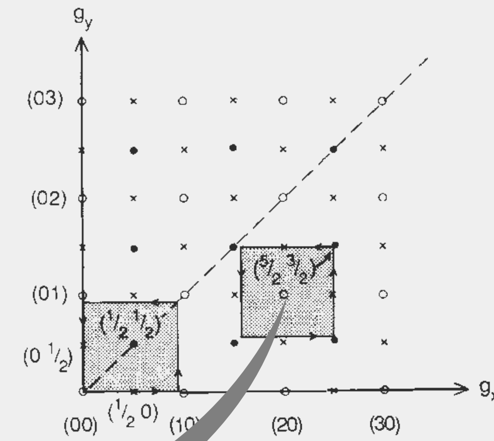
e^{-}

e^{+}



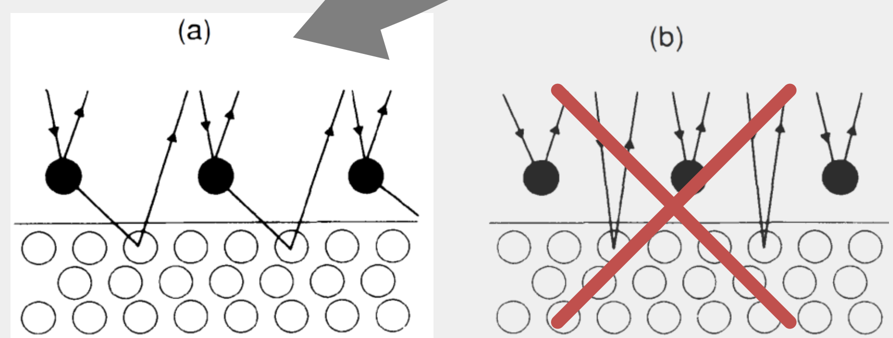
Tong et al., Phys. Rev. Lett. **69**, 3654 (1992)

Calculated for Cu(001)-*p*(2x2)Se



Theoretical prediction

LEPD spot intensities of (5/2 3/2) with **5 incident energies** from 114 eV to 166 eV gives sufficient information for holographic reconstruction.



We are going to try to prove it experimentally

Outline

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- LEPD
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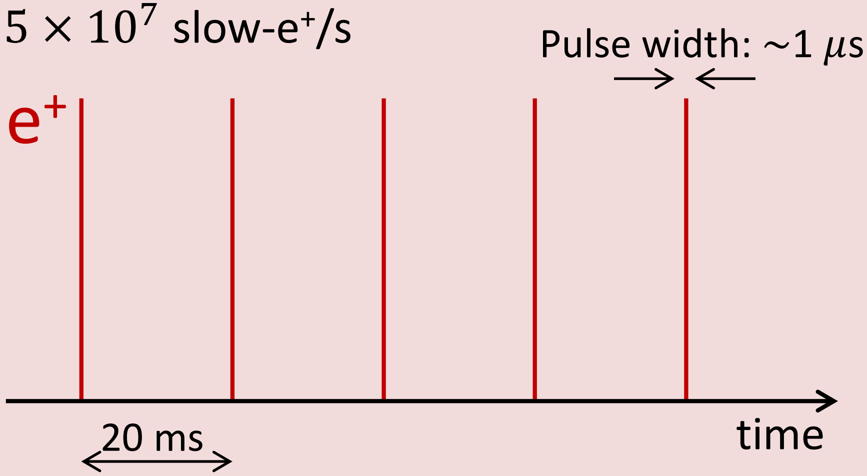
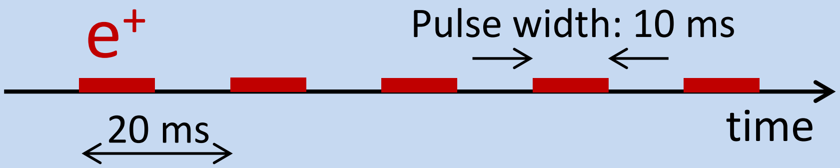
- Ps^- (Ps negative ion)
- Ps-TOF

Pulse stretching solves pile-up problem

5×10^7 slow-e⁺/s



5×10^7 slow-e⁺/s



Pulse stretching (10^4 times)

Difficulty in γ-ray spectroscopy

10^6 slow-e⁺/μs in a pulse

2γ-annihilation detection with
Solid angle: 1/100
Detection efficiency: 1/10

2×10^3 γ-rays detected per 1 μs

Pile-up problem at a detector

DLD detector has similar difficulty

Solved

10^2 slow-e⁺/μs in a pulse

2γ-annihilation detection with
Solid angle: 1/100
Detection efficiency: 1/10

0.2 γ-rays detected per 1 μs

No pile-up problem at a detector

Slow Positron Facility, KEK

Features

High Intensity

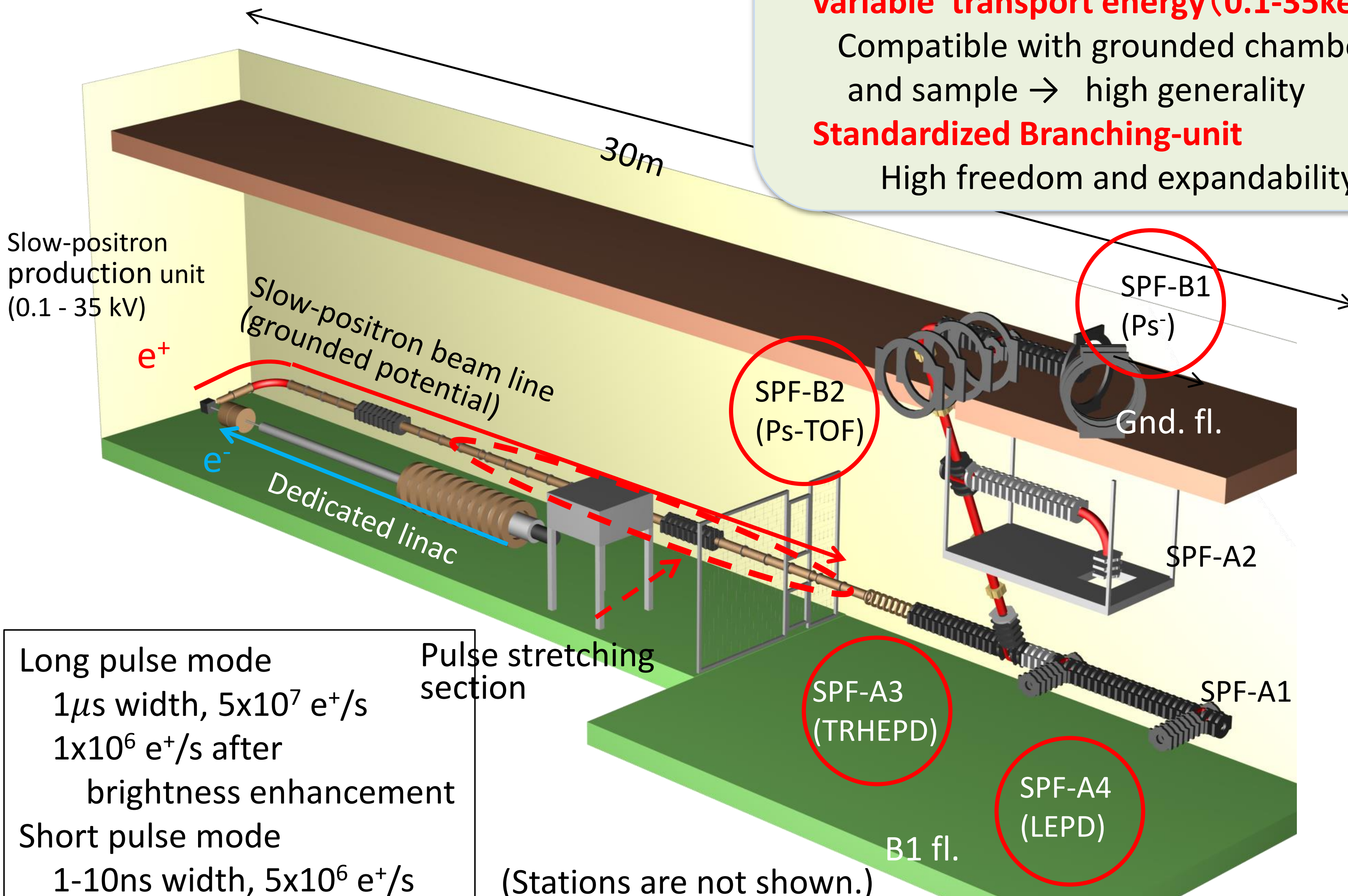
5×10^7 slow e^+/s (long pulse mode)

variable transport energy（0.1-35keV）

Compatible with grounded chamber and sample → high generality

Standardized Branching-unit

High freedom and expandability



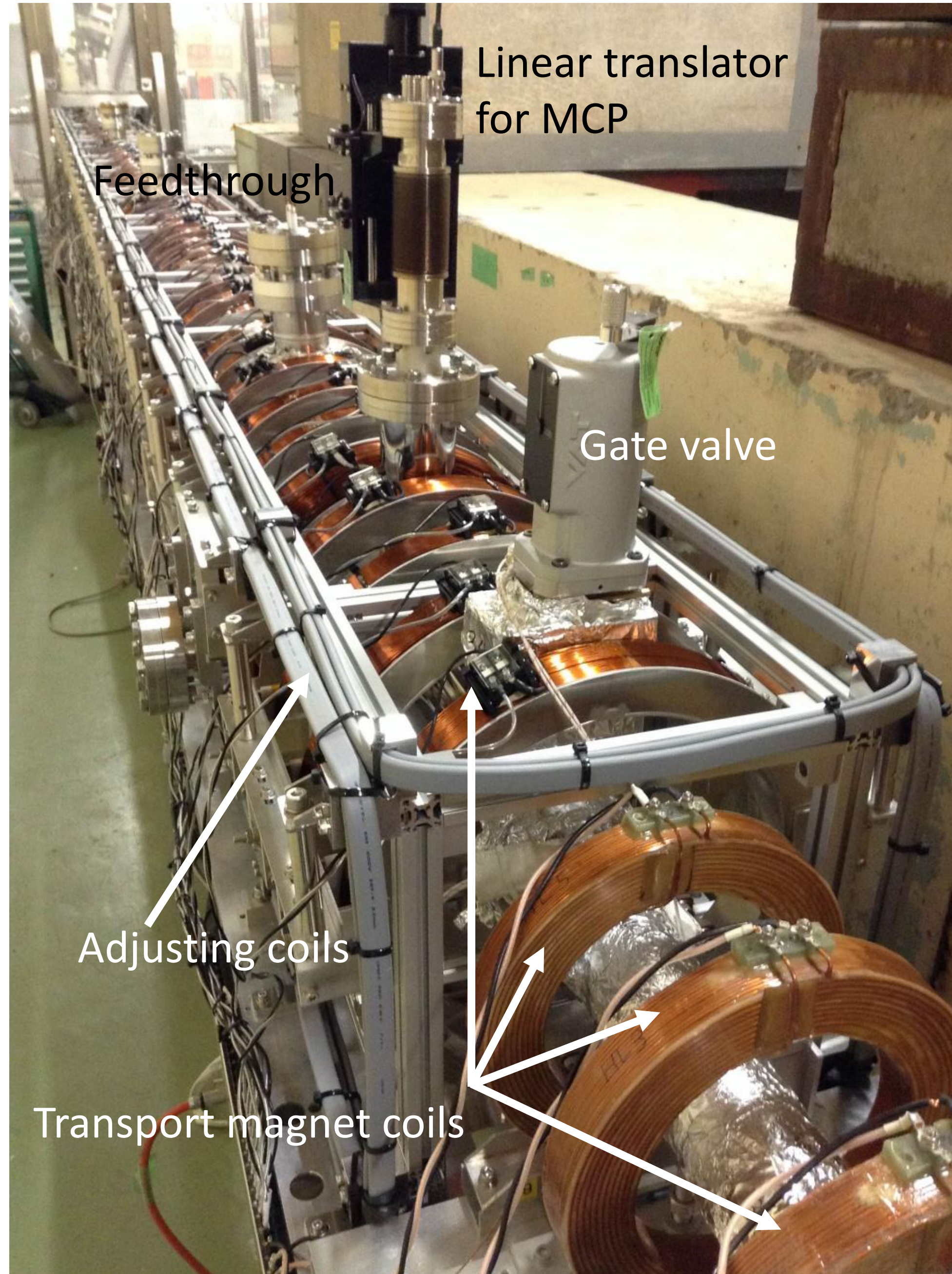
Long pulse mode

$1\mu s$ width, 5×10^7 e^+/s

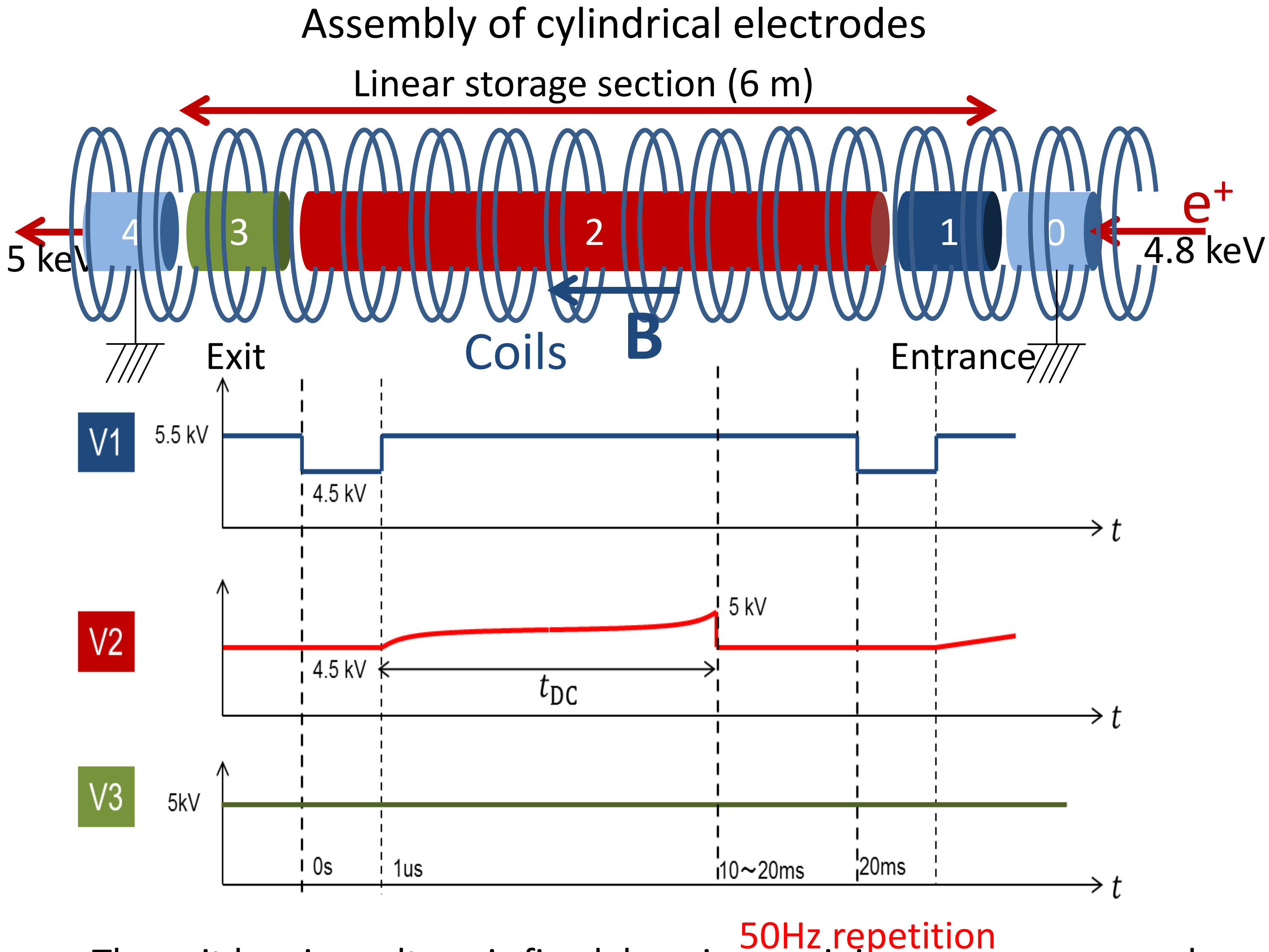
1×10^6 e^+/s after brightness enhancement

Short pulse mode

1-10ns width, 5×10^6 e^+/s

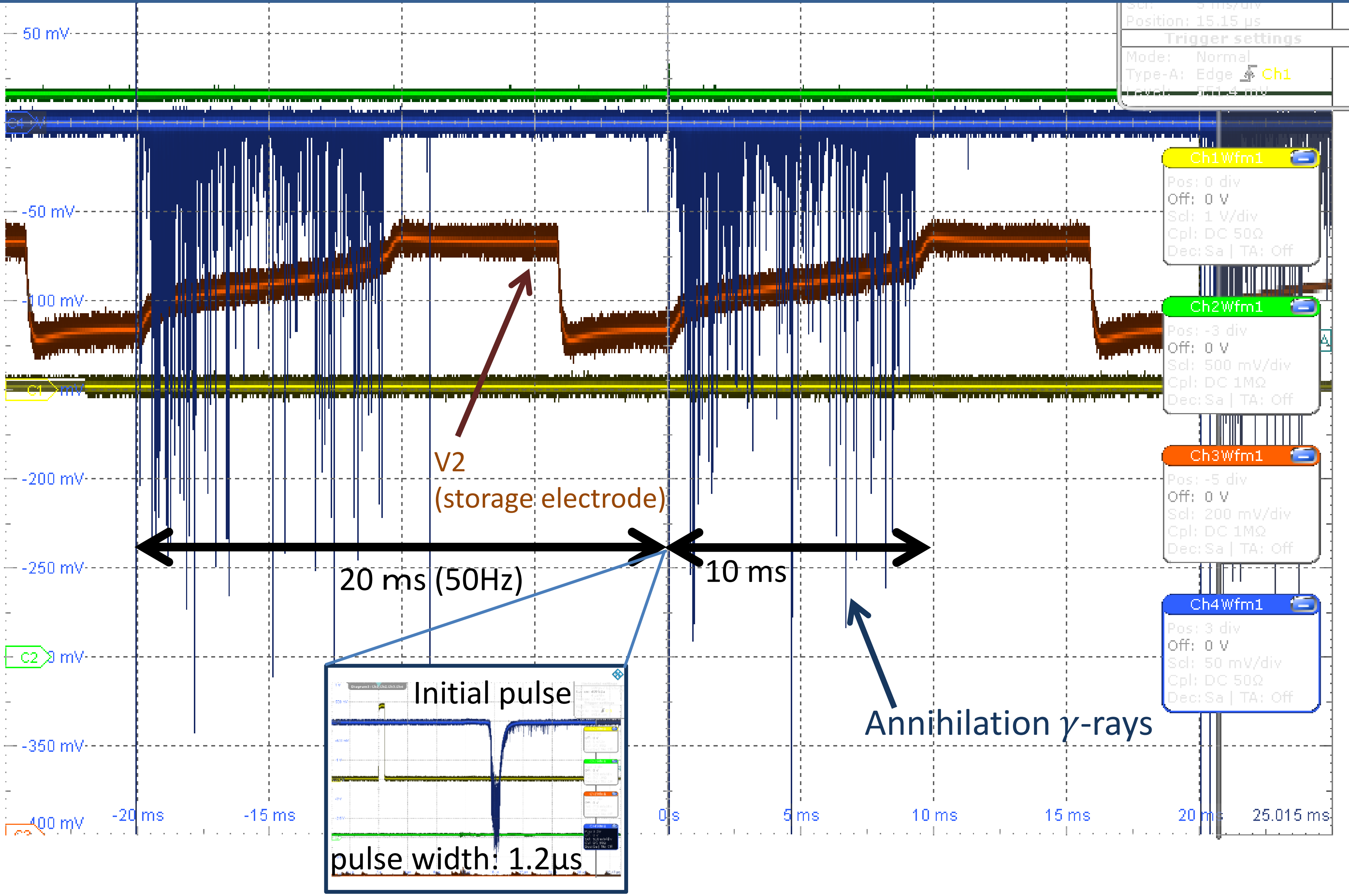

Linear translator
for MCP
Feedthrough
Gate valve
Adjusting coils
Transport magnet coils

Pulse stretching system newly developed at KEK Penning-Malmberg trap



- The exit barrier voltage is fixed, keeping a minimum energy spread.
- High-energy (up to 5 keV) pulse stretching

Stretched 5 keV pulse beam (pulse width: ～10 ms)



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Alkali metal coating enhances Ps emission

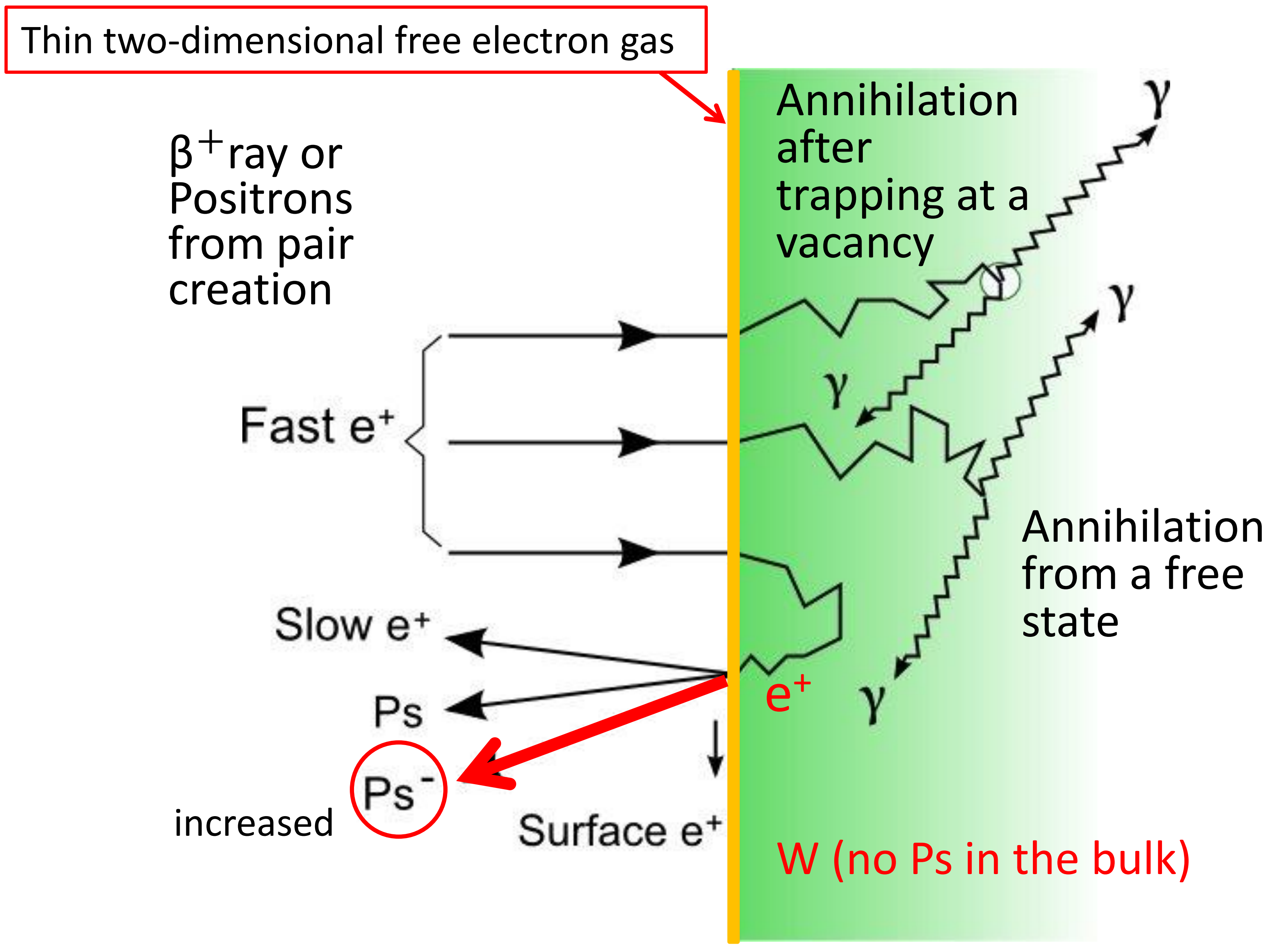
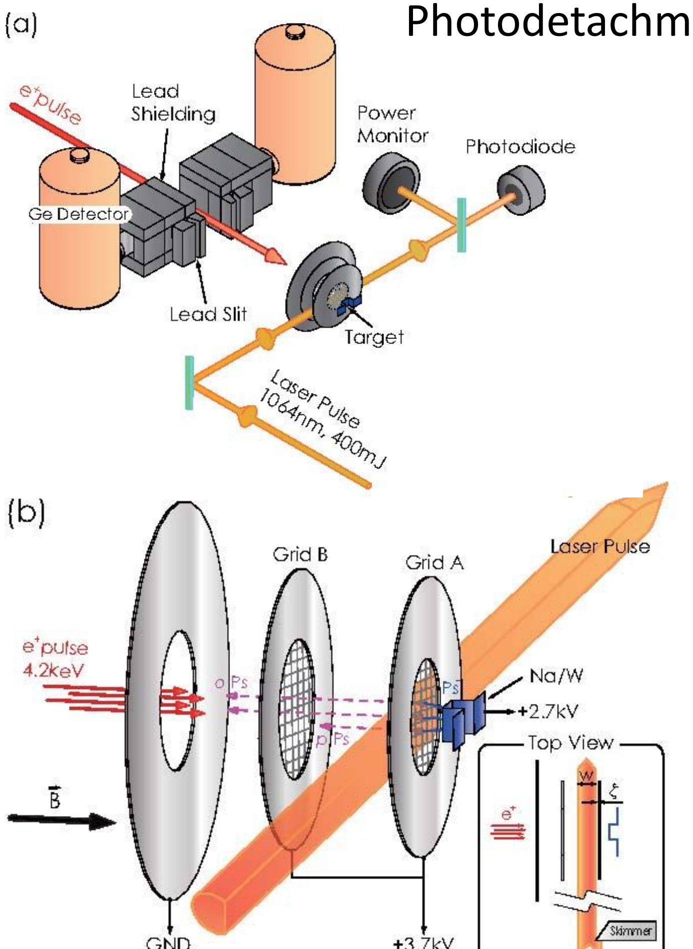


Photo detachment of Positronium negative ion (Ps^-)

Ps^- (pure leptonic three-body system, $e^- e^+ e^-$) is efficiently produced at alkali-metal-coated W surface, confirmed by Doppler-shift of the 511keV annihilation γ.

Ps^- is made into Ps by photodetachment using a laser beam.

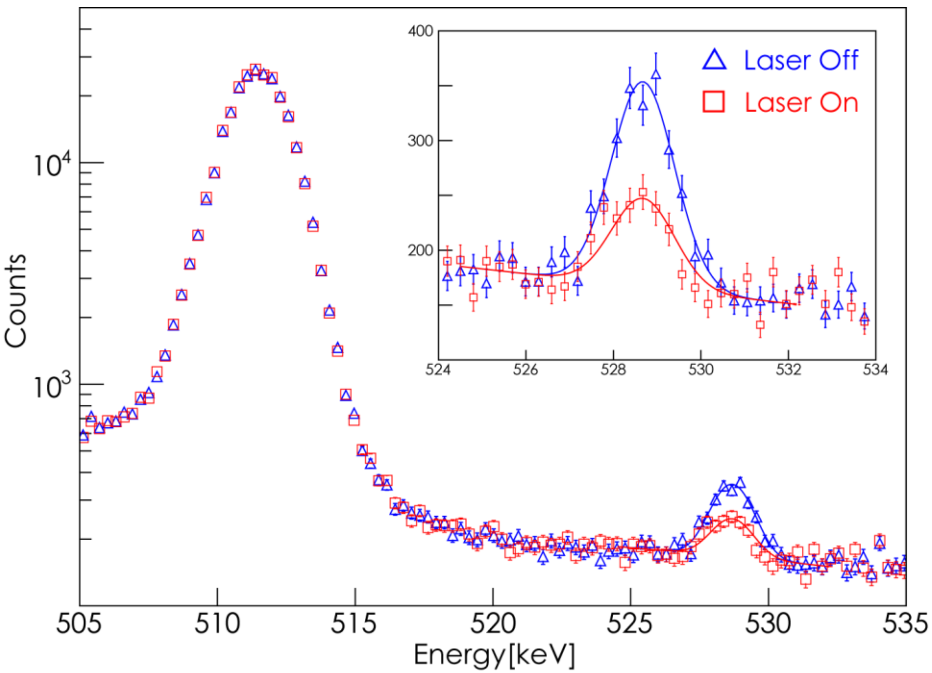


Photodetachment of Ps^-

$$Ps^- + h\nu \rightarrow Ps + e^-$$

$(e^- e^+ e^-)$ $(e^+ e^-)$

(o-Ps : p-Ps = 3 : 1)

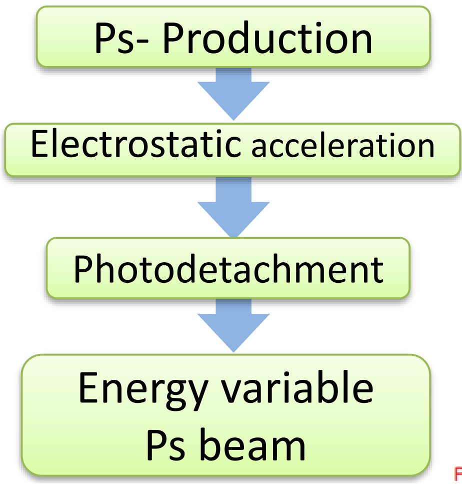


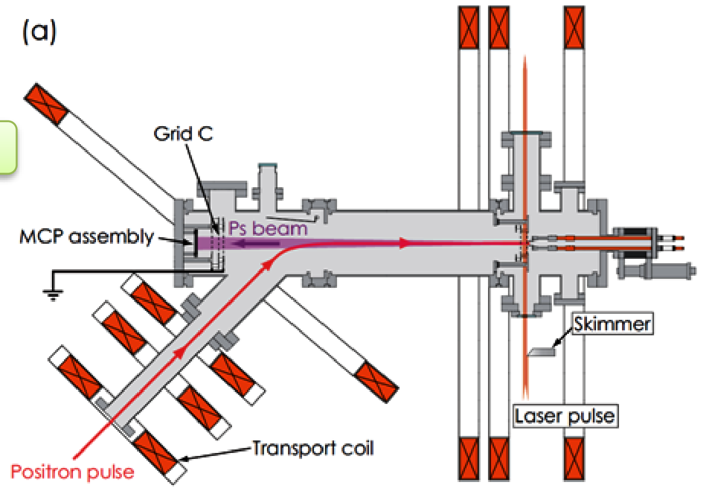
Michishio, *et al.*, Phys. Rev. Lett., 106, 153401 (2011).

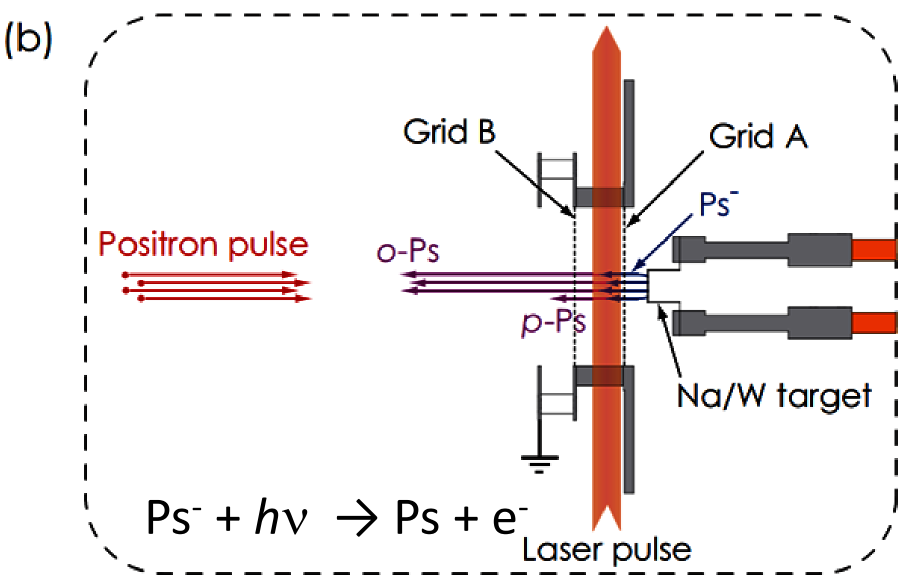
Production of energy-tunable Ps beam

Ps^- is accelerated to a desired energy , and then photodetachied to be neutral Ps.

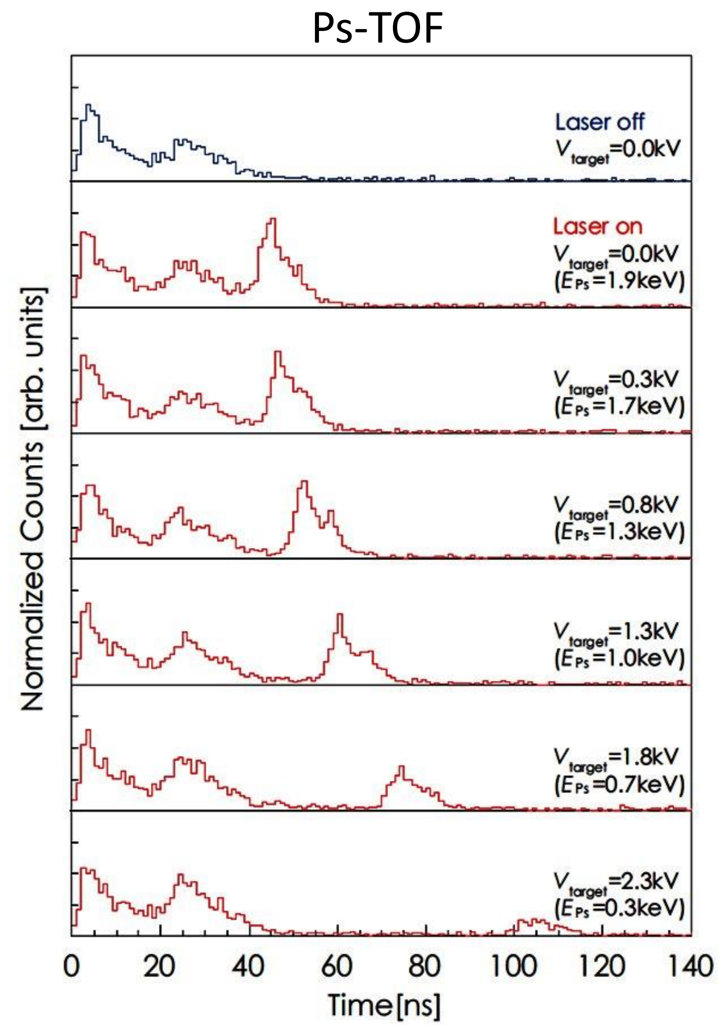
Congfermed by Ps-TOF measurements.





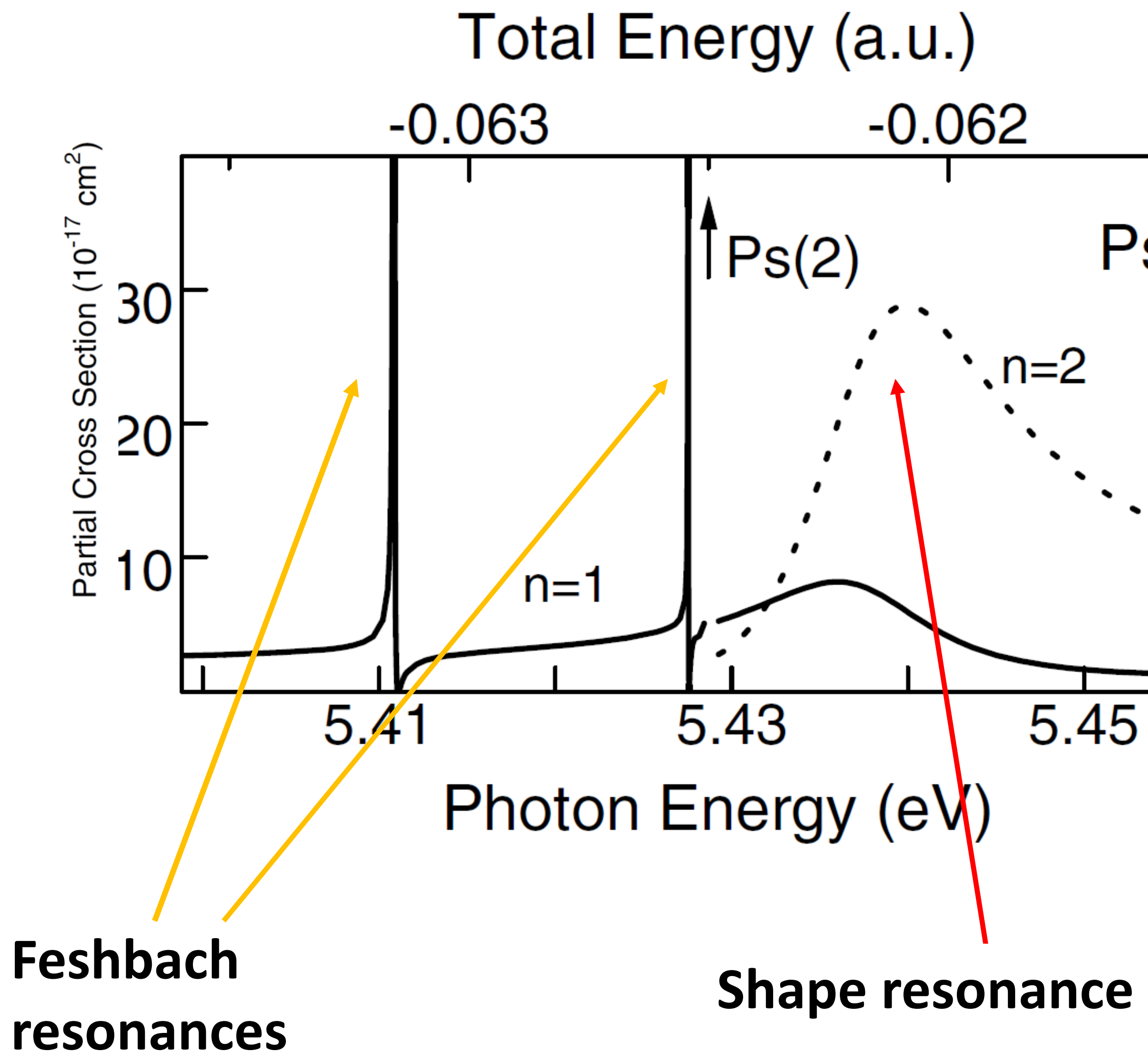


$$Ps^- + h\nu \rightarrow Ps + e^-$$



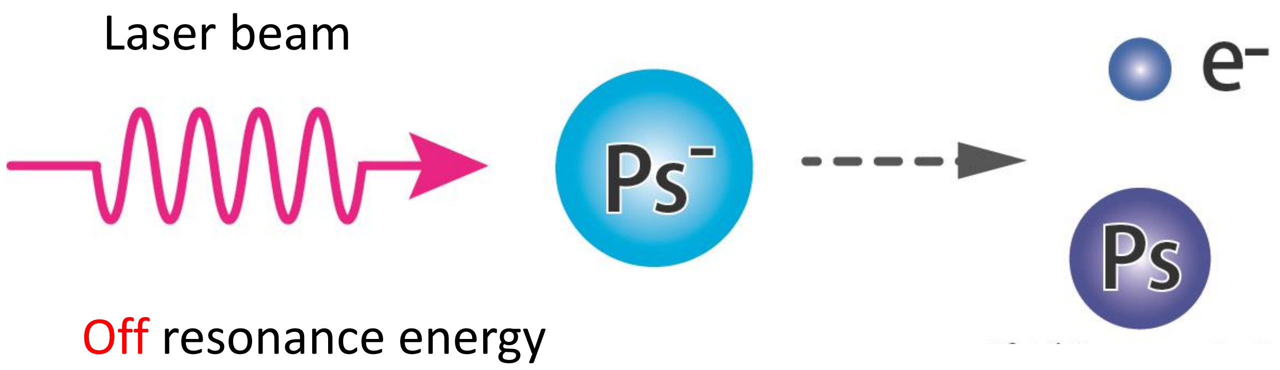
Michishio, *et al*., Appl. Phy. Lett. 100, 254102 (2012).

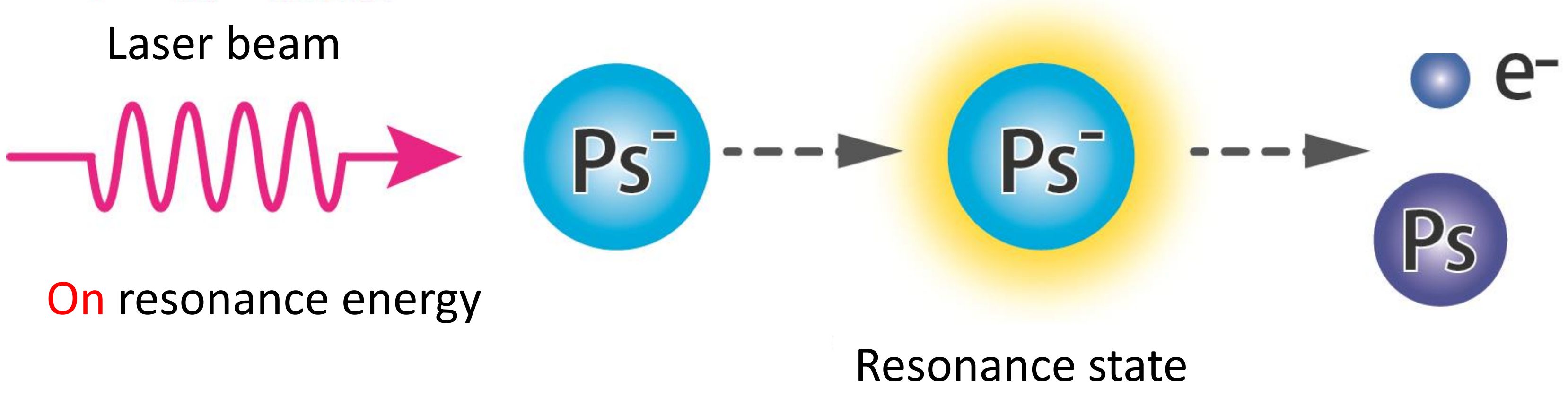
Theoretical prediction of resonances in photodetachment cross section of Ps^-



Resonances in photodetachment cross section of Ps^-

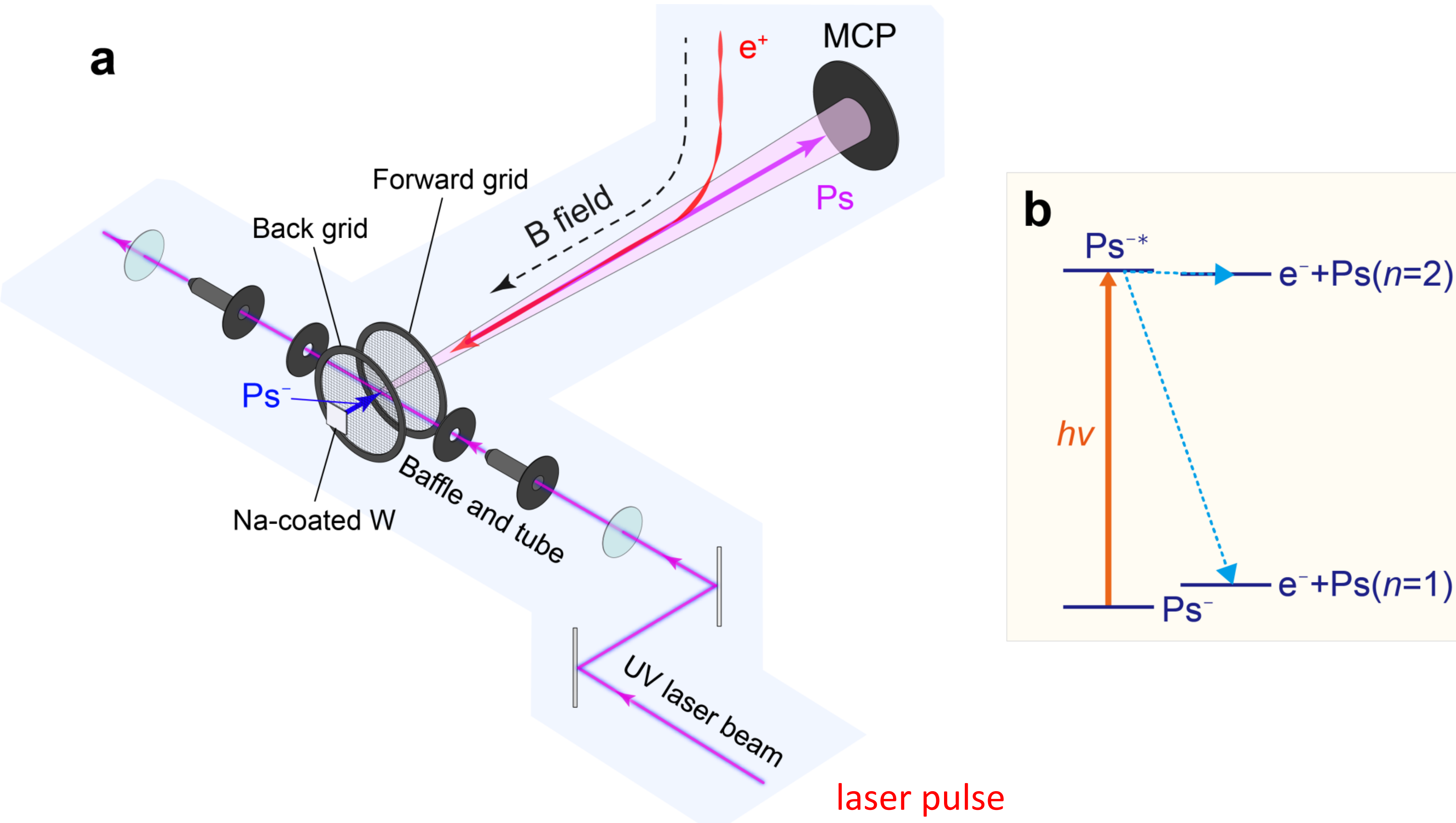
Ps^- does not have an excited state.
But resonant states exist which enhance photodetachiment.





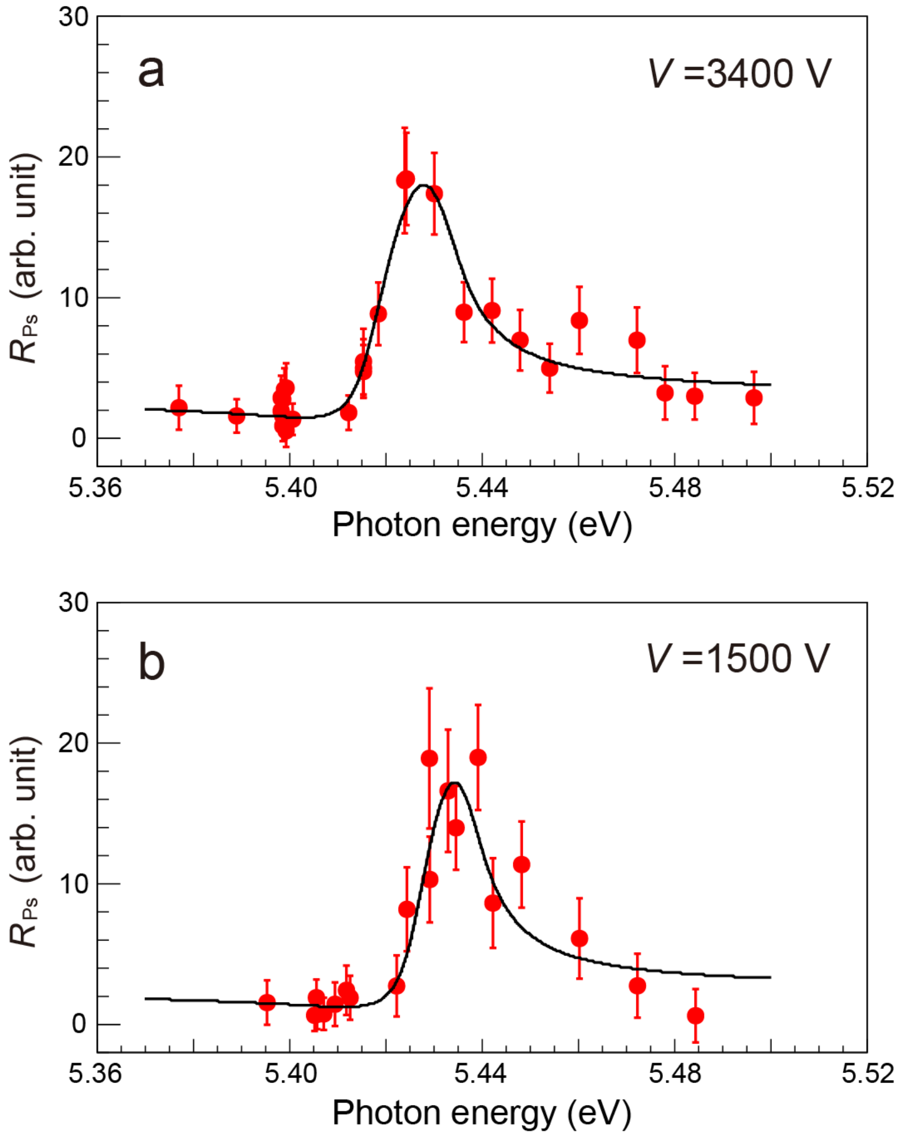
Measurement of photodetachment cross section of Ps⁻

pulsed e⁺ beam



laser pulse

Results of shape resonance measurements



K. Michishio, et al., Nature Communications 7, 11060 (2016) doi:10.1038/ncomms11060.

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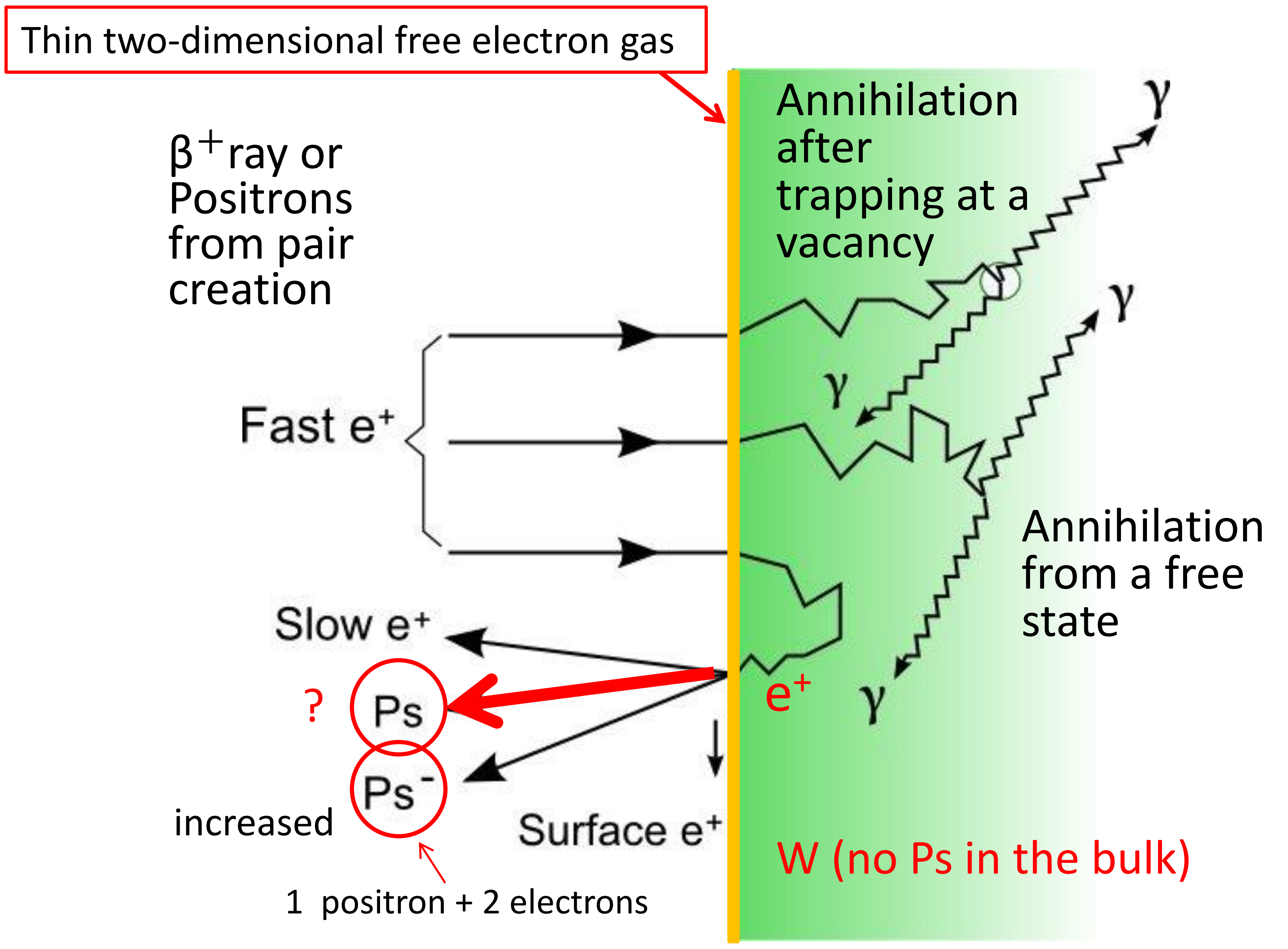
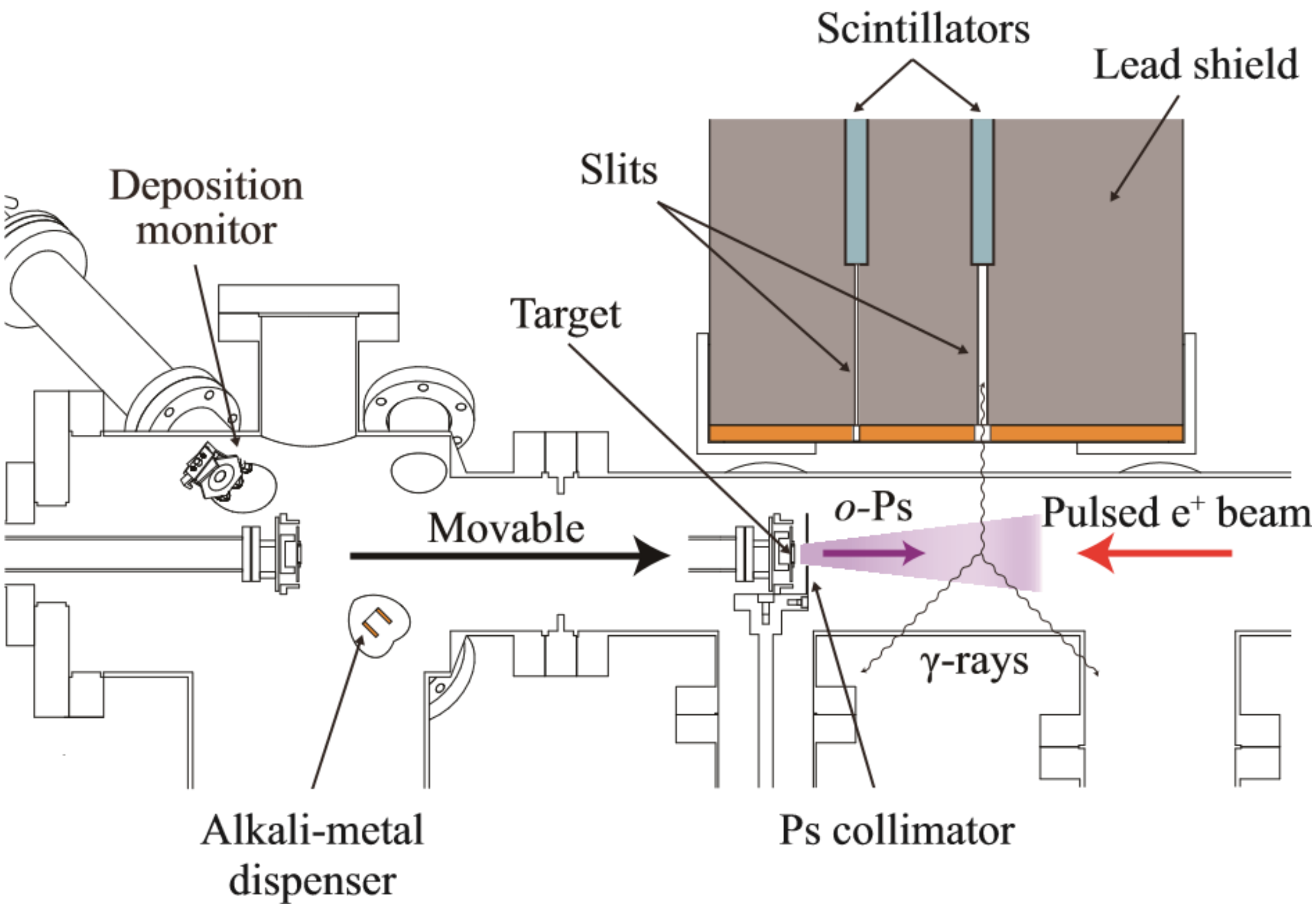


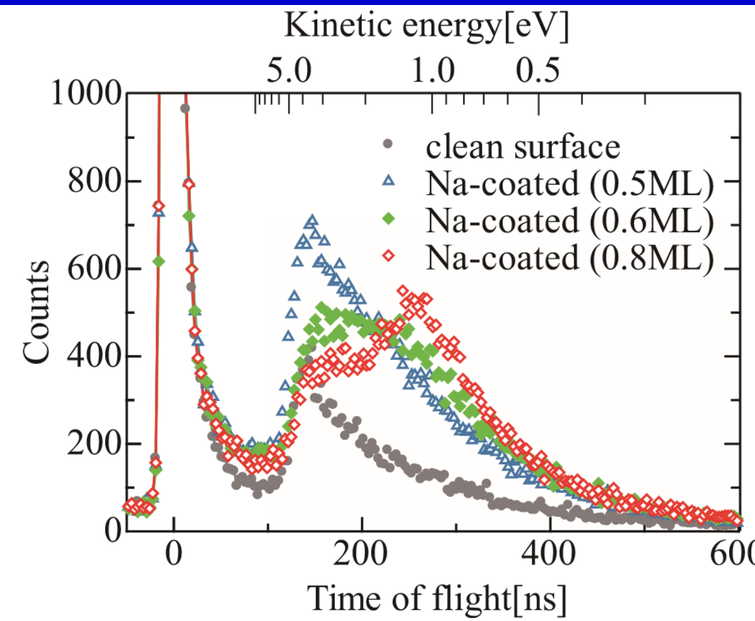
Ps-TOF station

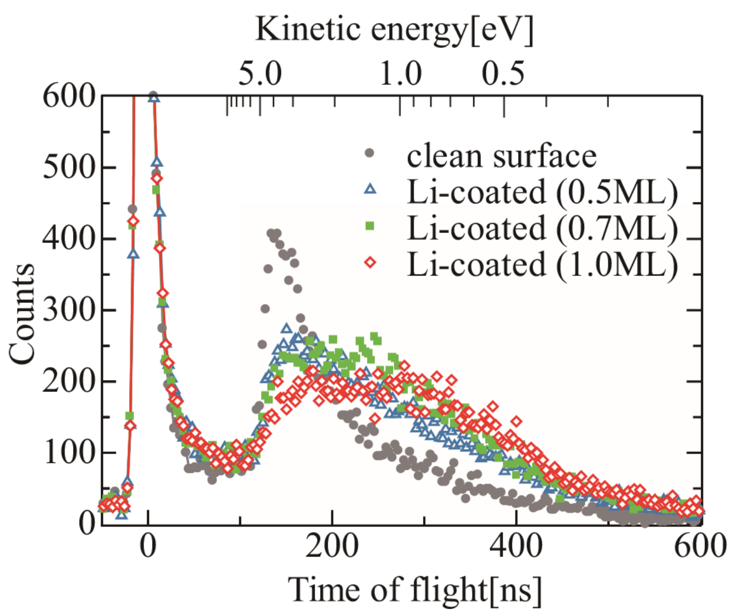


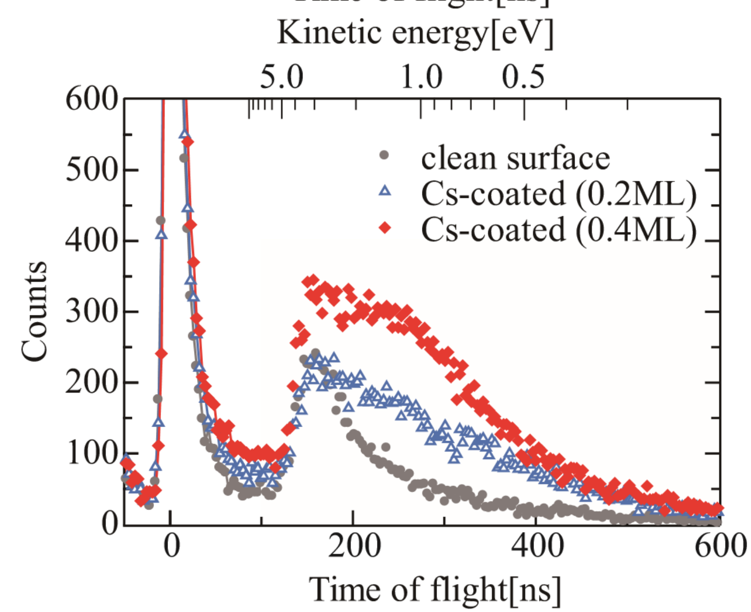
Ps-TOF form clean and alkali-metal-coated W

Ps formation increases on coating W surface with alkali metals (sub-monolayer).

Almost 90% of the positrons which come back to the surface are emitted as Ps.



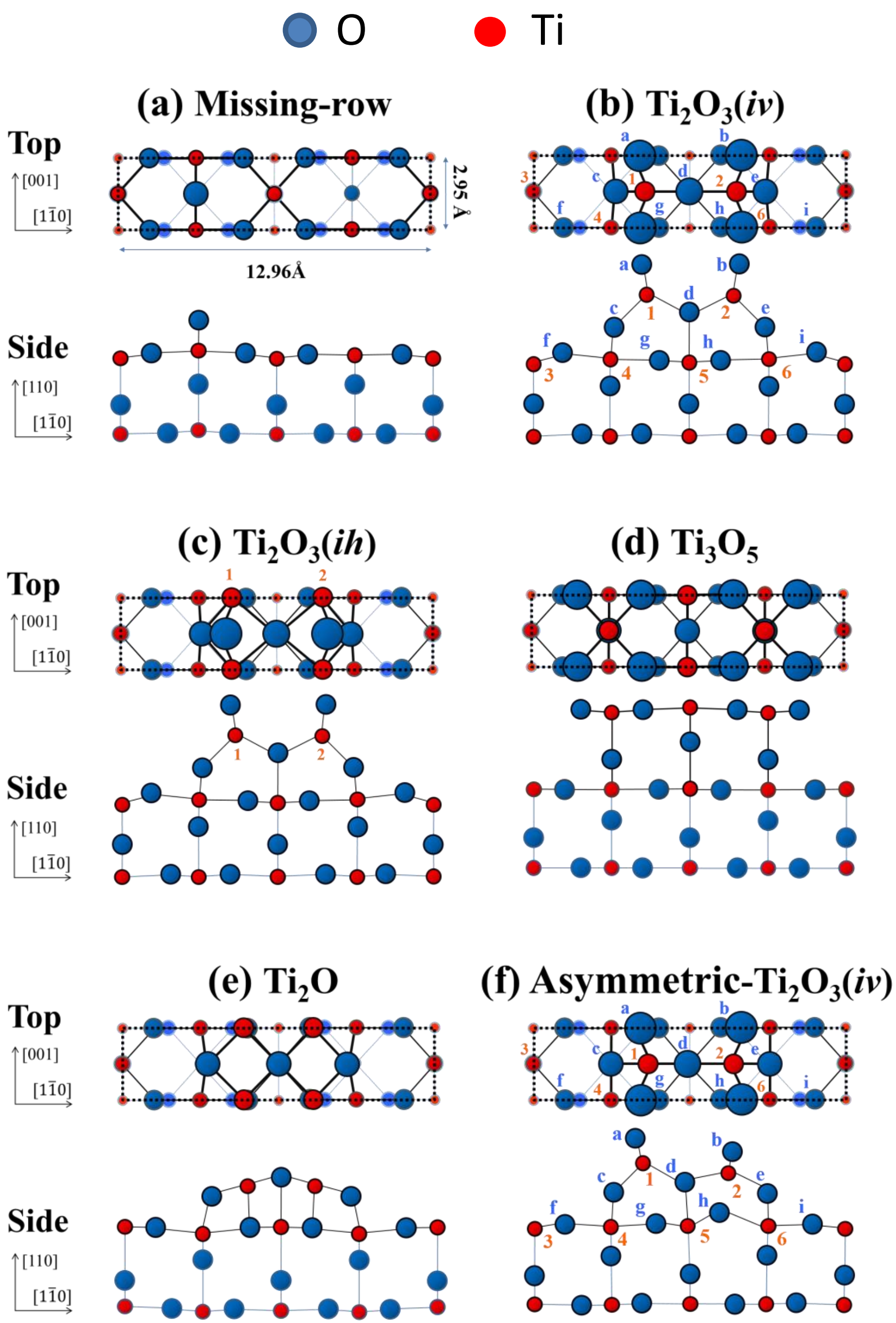


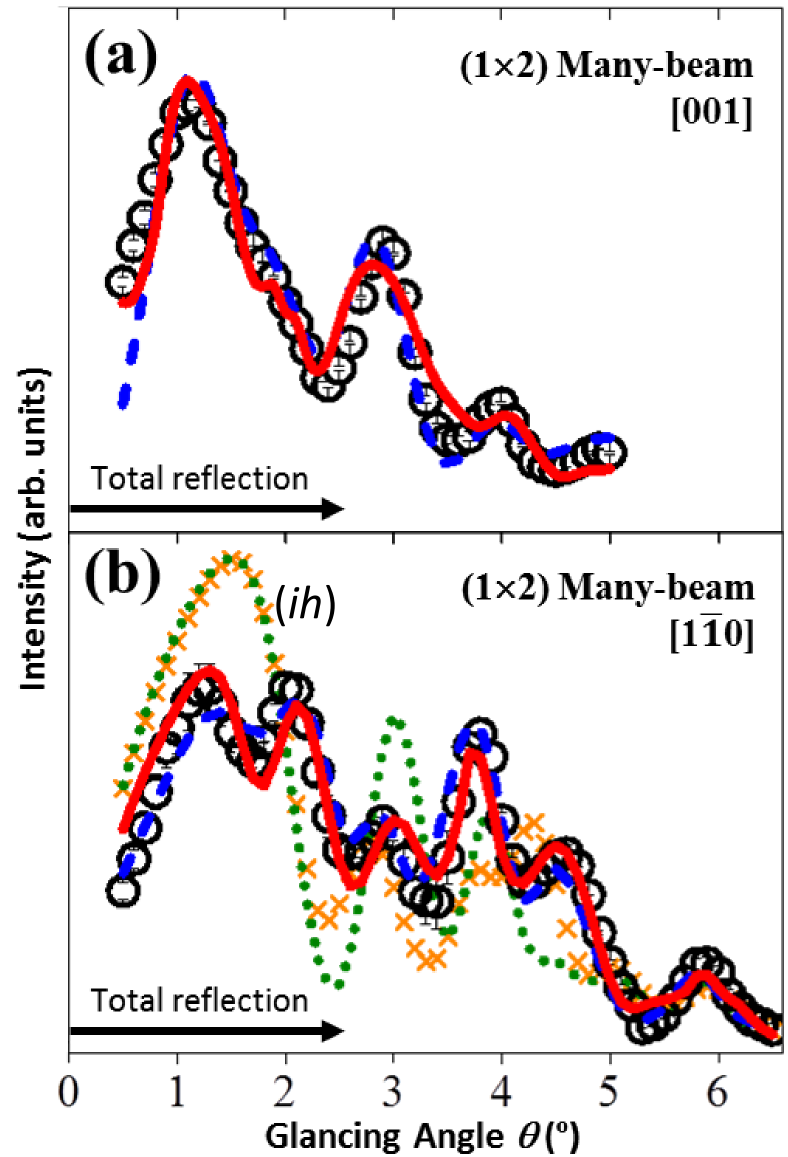


S. Iida, et al., J. Phys.: Condens. Matter 28, 475002 (2016)

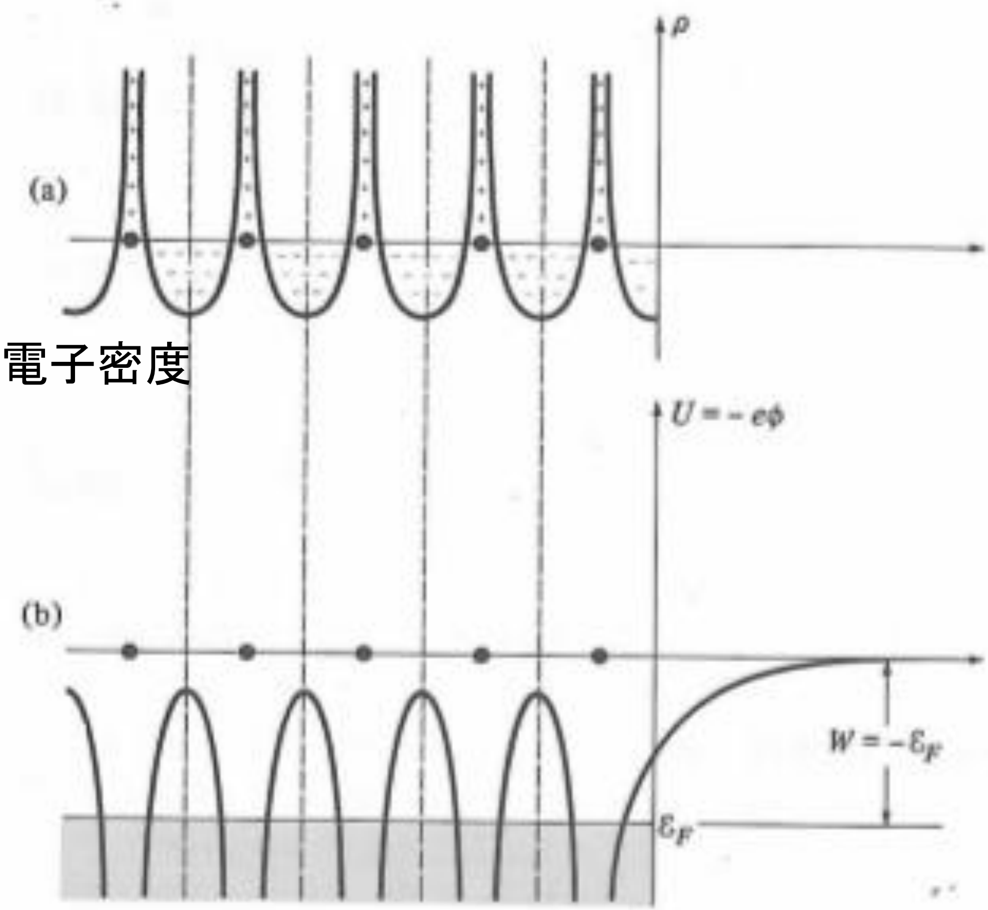
Summary

- Energy-tunable slow-positron beam is successfully used at SPF, IMSS, KEK
- Intensities are 5×10^7/s in long-pulse mode (width 1.2 μs) and 5×10^6/s in short-pulse mode (width 1-10 ns, variable).
- 5 keV pulse may be stretched to 200μs-20ms (variable).
- Surface structure study by positron diffraction (TRHEPD and LEPD), Surface science by Ps-TOF and science motivated by Ps^- are currently conducted.


O
Ti
(a) Missing-row
(b) $Ti_2O_3(iv)$
(c) $Ti_2O_3(ih)$
(d) Ti_3O_5
(e) Ti_2O
(f) Asymmetric-$Ti_2O_3(iv)$
Top
Side
[001]
[1$\bar{1}$0]
[110]
2.95 Å
12.96Å


(a)
(1×2) Many-beam
[001]
(b)
(1×2) Many-beam
[1$\bar{1}$0]
(ih)
Total reflection
Total reflection
Intensity (arb. units)
Glancing Angle θ (º)
0
1
2
3
4
5
6

Ashcroft/Mermin の教科書の電子密度と結晶ポテンシャルの図



電子密度

電子のポテンシャル・エネルギー

$$\left(-\frac{\hbar^2}{2m}\nabla^2 + U\right)\psi = E\,\psi$$

$$(U = -e\phi)$$

Figure 18.1
(a) The electric charge density near the surface of a finite crystal if there were no distortion in cells near the surface. The density is plotted along a line of ions. Vertical dashed lines indicate cell boundaries. (b) The form of the crystal potential U (or the electrostatic potential $\phi = -U/e$) determined by the charge density in (a), along the same line. Far from the crystal U and ϕ drop to zero. The (negative) Fermi energy is indicated on the vertical axis. The shading below the Fermi energy is meant to suggest the filled electronic levels in the metal. Since the lowest electronic levels outside the metal have zero energy, an energy $W = -E_{\mathrm{F}}$, must be supplied to remove an electron.
(p.356)

Crystal Potential and Work Function

$$\left(-\frac{\hbar^2}{2m}\nabla^2 + qV\right)\psi = E\psi$$

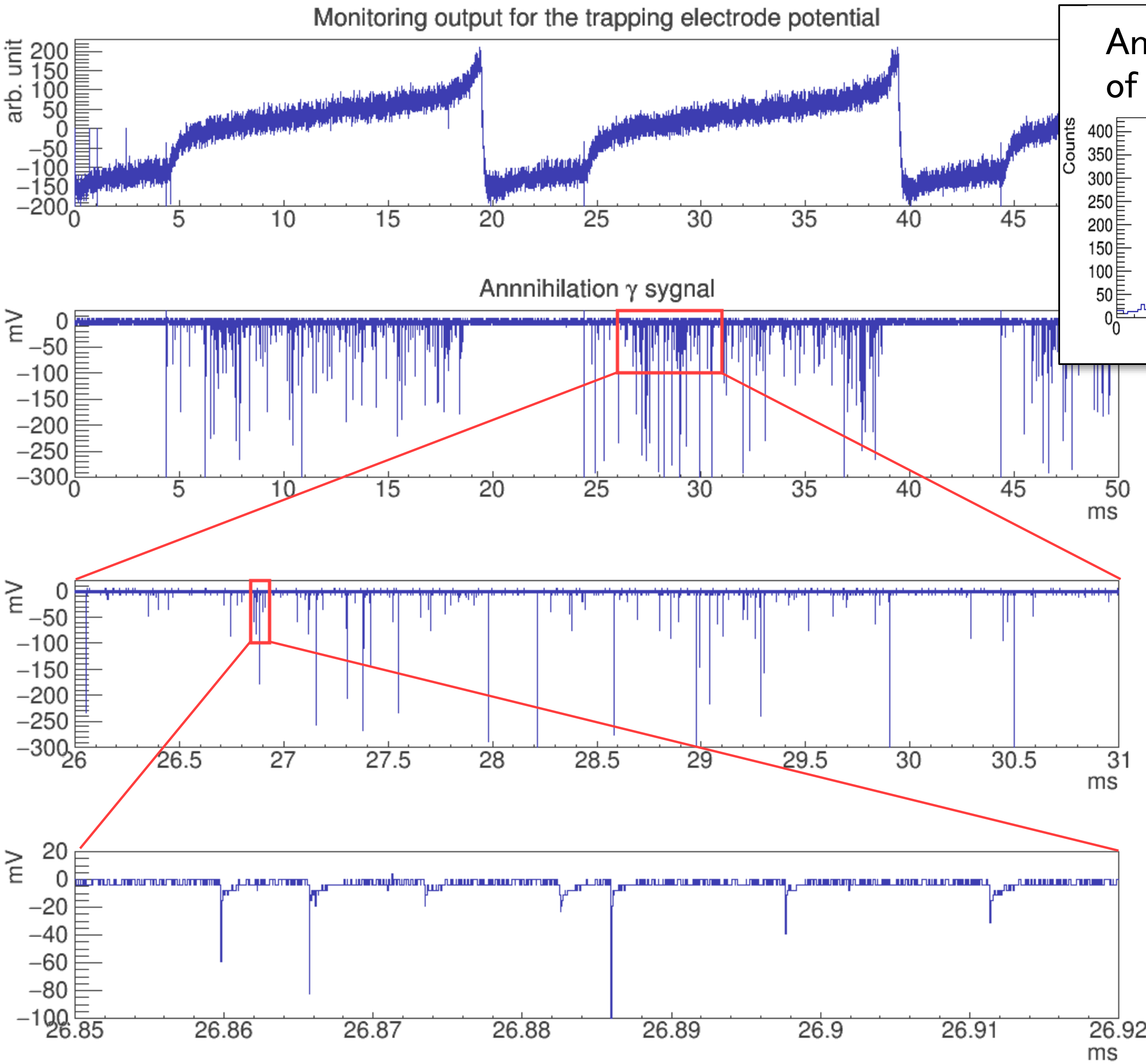
Average crystal potential: V (> 0: always)

Average potential energy: $qV = \begin{cases} -eV & (< 0 \text{ for the electron}) \\ eV & (> 0 \text{ for the positron}) \end{cases}$

Total reflection in TRHEPD

Work function: Ground state energy of a particle in fully interacting system (with respect to vacuum level)

5-keV pulse stretched beam (15 ms)



An example of time distribution of the γ signals.