Evaluation of energy-resolved spin polarization of surface electrons by spin-polarized positronium time-of-flight method (SP-PsTOF)

M. Maekawa, S. Sakai, A. Miyashita and A. Kawasuso

Quantum Beam Science Research Directorate,
Takasaki institute,
National Institutes for Quantum and Radiological Science and Technology

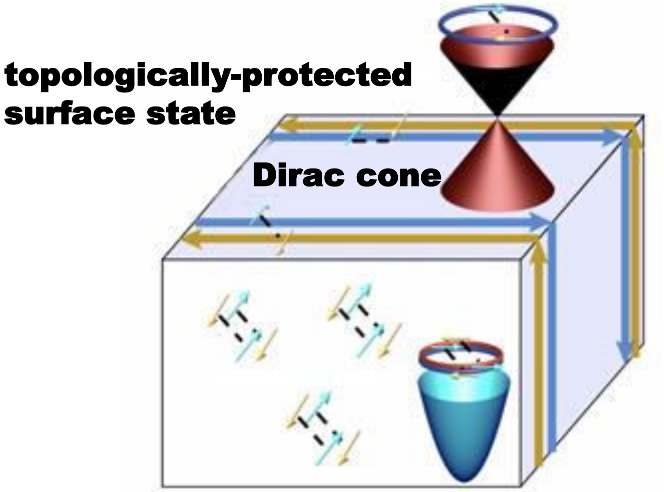
Topmost spin phenomena

- ✓ **Novel quantum spin phenomena on the topmost surface are important for realizing next-generation devices.**

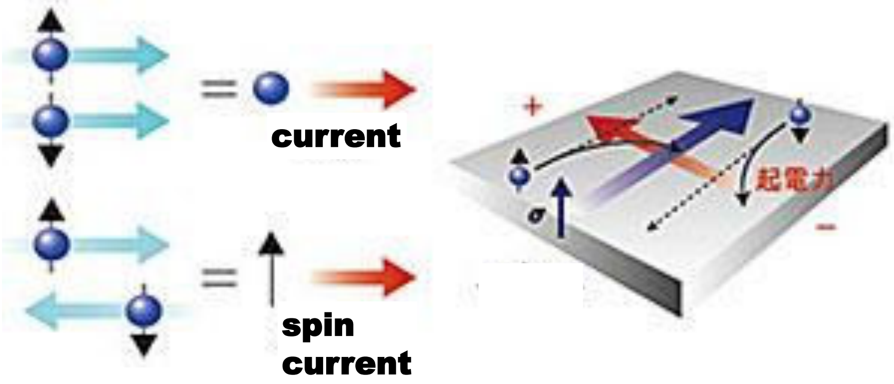
- ✓ **To understand these phenomena, the spin polarization of the topmost layer must be evaluated correctly.**

Spin phenomena on the topmost surface

ex. Topological insulator



ex. spin-Hall effect



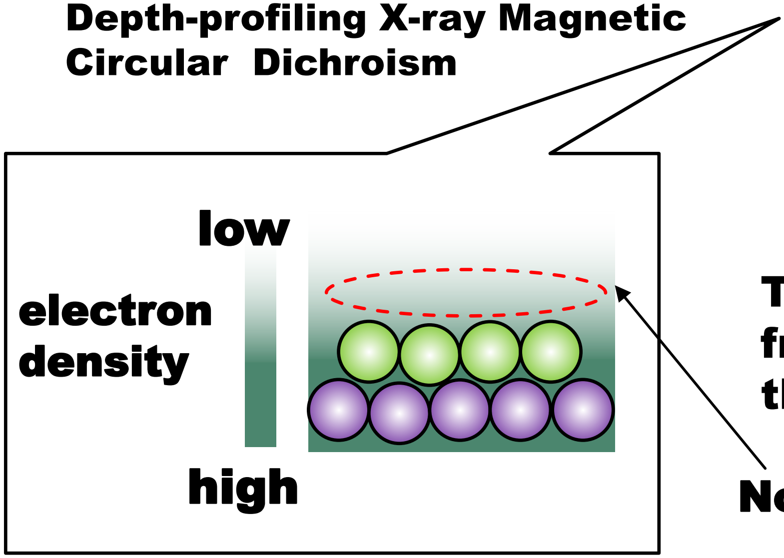
Surface spin evaluation

- ✓ There are several methods to evaluate surface spins.
- ✓ It is very difficult to evaluate only such the spins.

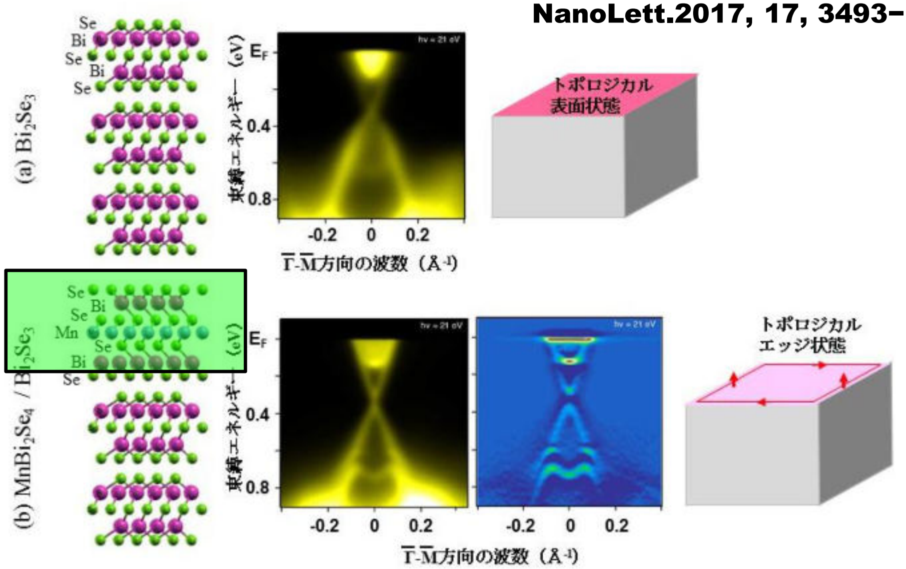
Commonly used techniques:

S-ARPES
Spin- and Angle-Resolved
Photoemission Spectroscopy

DP-XMCD
Depth-profiling X-ray Magnetic
Circular Dichroism



NanoLett.2017, 17, 3493−3500



These results include information not only from the surface electrons but also from the several bulk layer.

Novel quantum spin phenomena occurs here

Positronium spectroscopy can provide such information

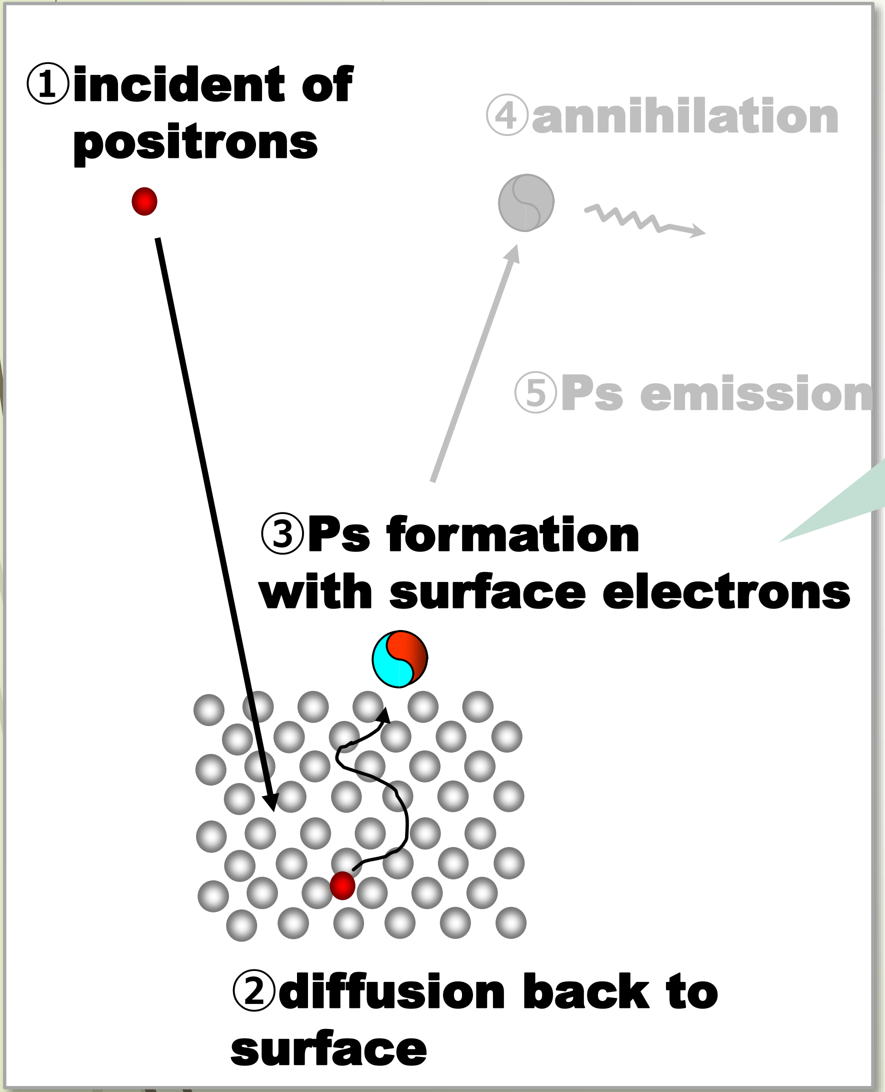
Advantages of positronium spectroscopy:

1. **Ps is formed only at the vacuum side of the surface.**

2. **Ps annihilation changes depending on the direction of electron spin. (p-Ps and o-Ps)**

3. **Ps emission affected by the state of electrons.**

1. Ps formation only at surface



①**incident of positrons**

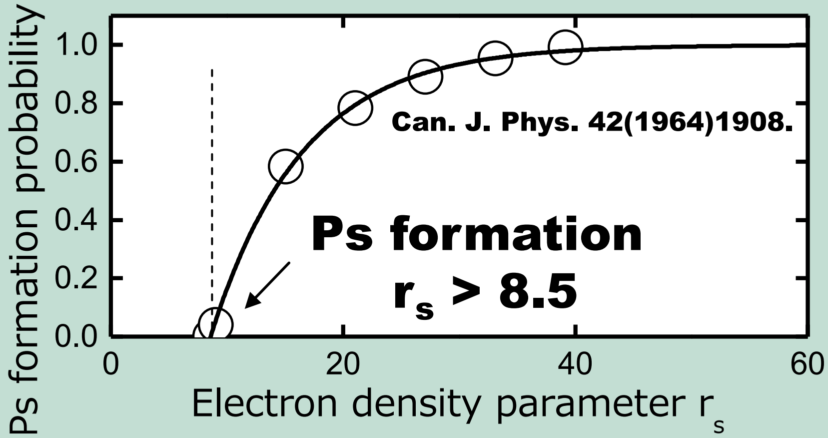
④**annihilation**

⑤**Ps emission**

③**Ps formation with surface electrons**

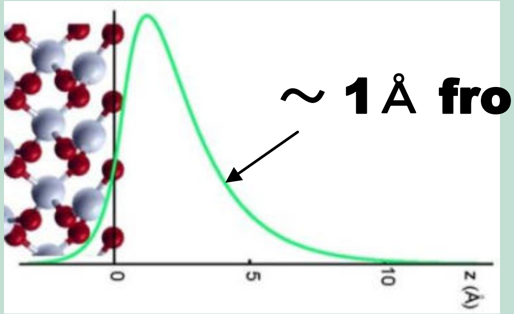
②**diffusion back to surface**

Ps can form only at surface



Can. J. Phys. 42(1964)1908.

Ps formation
r_s > 8.5

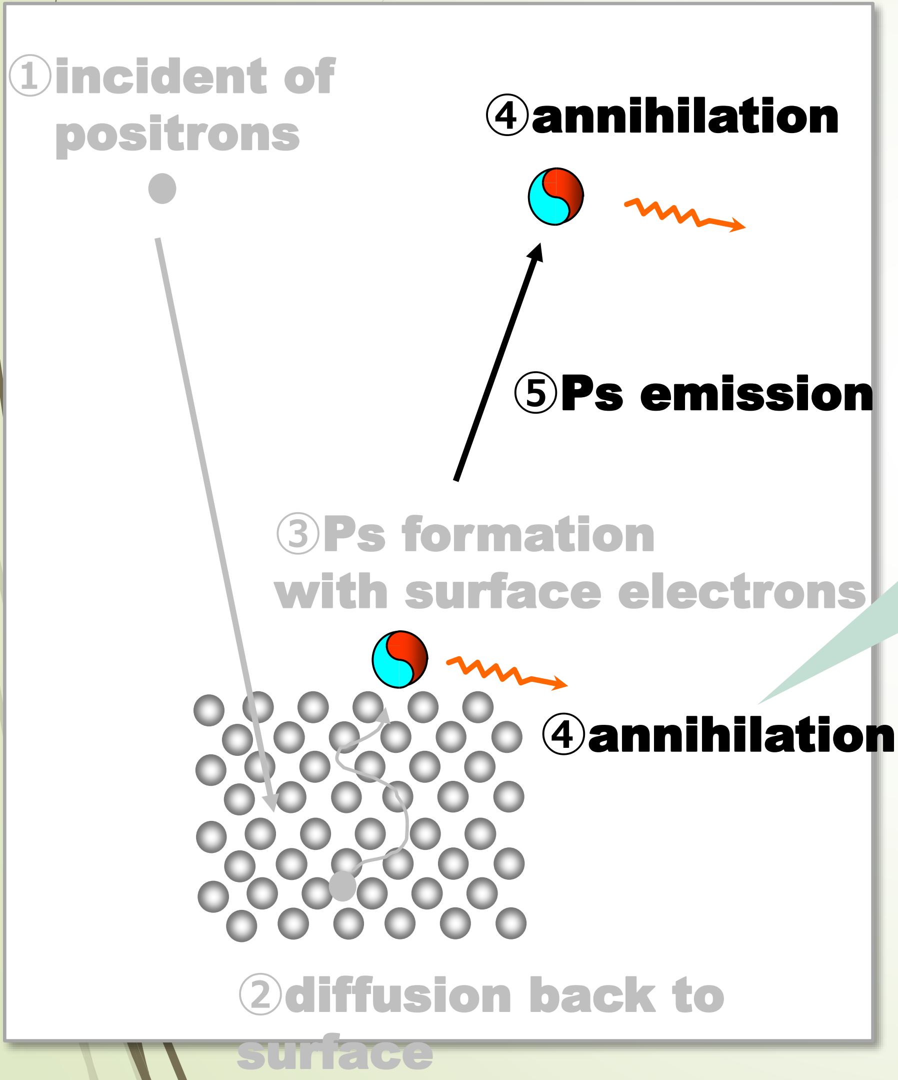


～1Å from surface

Saints, PRL100(2008)019902

Ps observation = surface electron observation

2. Ps state changes by electron spin



①incident of positrons

④annihilation

⑤Ps emission

③Ps formation with surface electrons

④annihilation

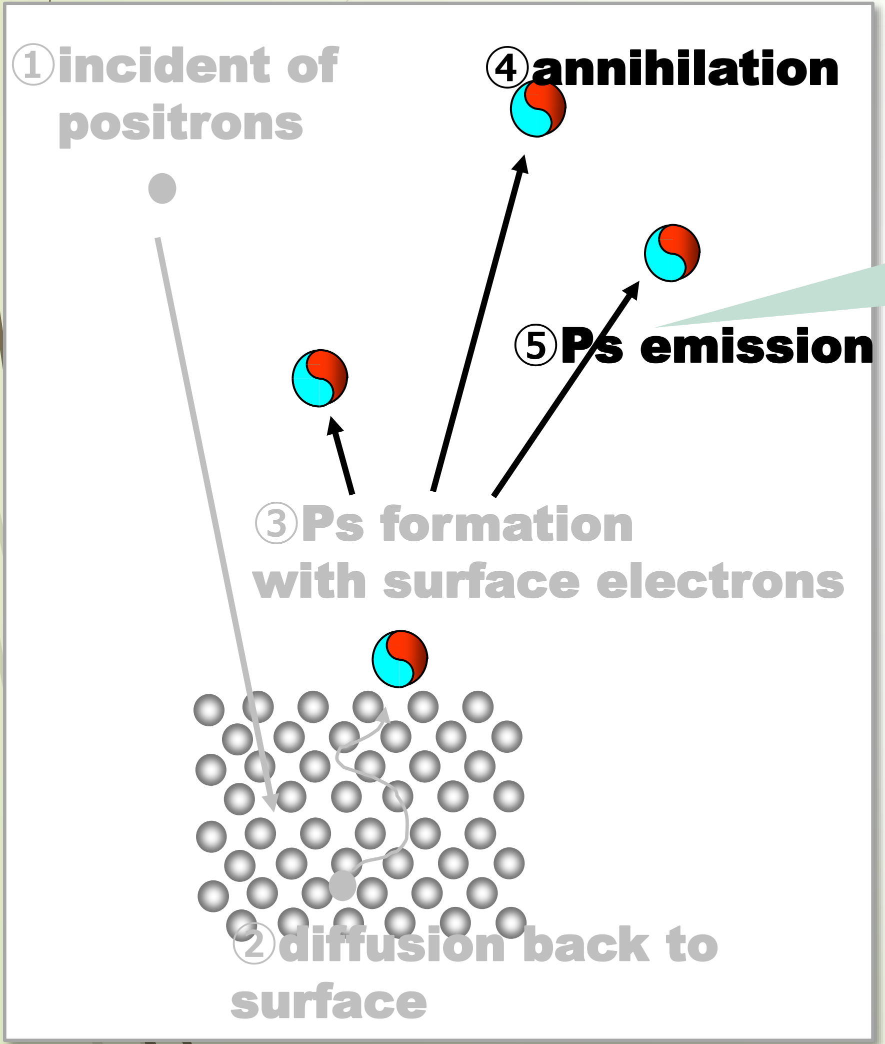
②diffusion back to surface

Ps state changed by electron spin

spin parallel	spin anti-parallel
ortho-Ps	para-Ps
3 photon decay (~511keV×3)	2 photon decay (511keV×2)
lifetime 142ns	125ps

Determination of electron spins
(i.e. spin polarization)

3. Ps emission affected by electron state



①incident of positrons

②diffusion back to surface

③Ps formation with surface electrons

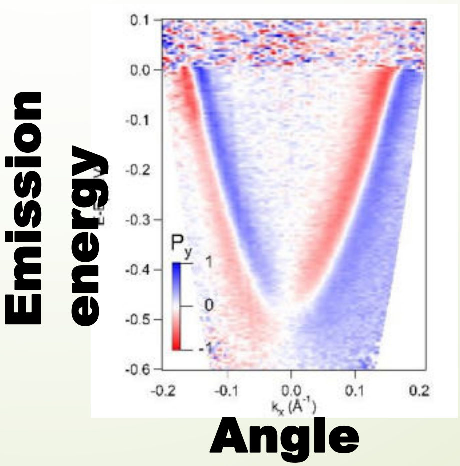
④annihilation

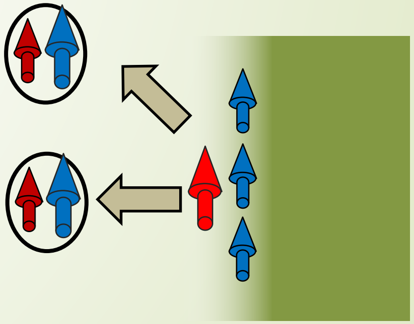
⑤Ps emission

Ps emission energy and angles are affected by electron state

Ps emission energy spectrum

$$\frac{d(N(\theta\varphi))}{d\varepsilon_\perp} \propto \frac{\alpha}{\sqrt{\varepsilon_\perp}} \int \gamma \left[\tan(\theta) - \frac{|k_\parallel|}{q}\right] |\nabla_\perp(\varepsilon \boldsymbol{k})|^{-1} d\boldsymbol{k}_\parallel$$

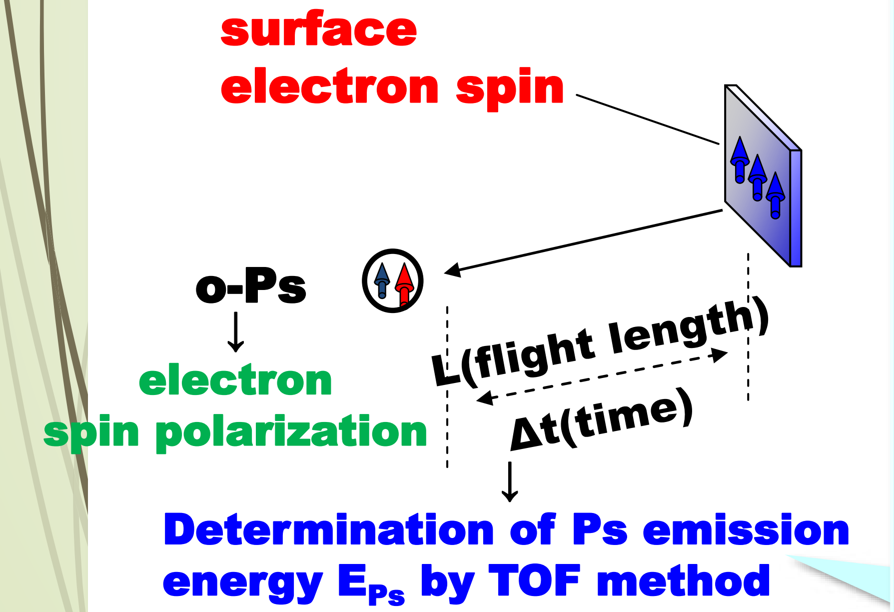




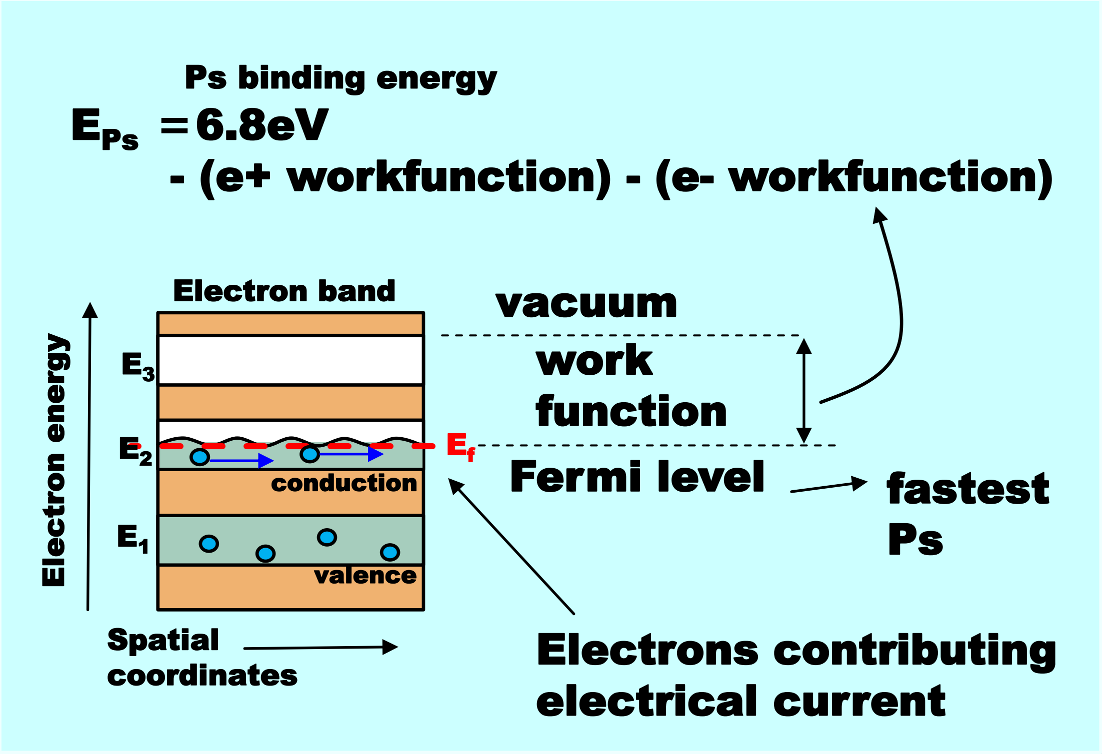
Spin-polarized electron band structure might be obtained !

Energy-resolved Ps spectroscopy

- ✓ **Energy of emitted Ps can be determined by Ps time-of-flight(TOF) .**
- ✓ **The spin-polarized electron density of states associated only with the topmost layer of metals will be obtained.**



Determination of Ps emission energy E_{Ps} by TOF method



Ps binding energy

E_{Ps} = 6.8eV - (e+ workfunction) - (e- workfunction)

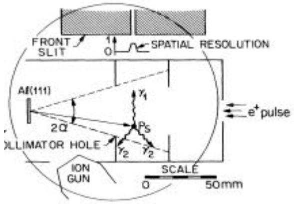
work function

Fermi level → fastest Ps

Electrons contributing electrical current

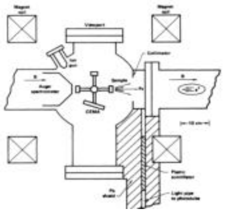
PsTOF apparatus

<table>
<tr><td colspan="2" rowspan="2"></td><td colspan="2">Timing signal</td></tr>
<tr><td>beam pulsing (high efficiency), spin depolarization</td><td>secondary electron spin pol.</td></tr>
<tr><td rowspan="2">Positron source</td><td>Large-scale facility
high intensity</td><td>
Bell lab.



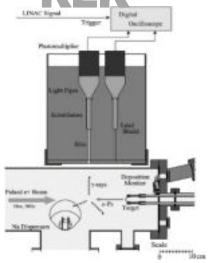
PRB34(1986)3069

LLNL



PRL51(1983)1085

KEK



Surf. Sci. 641(2015)68.
</td><td>
Torrent univ. → NEPOMUC

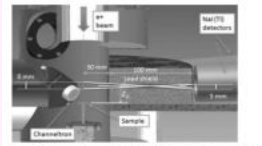
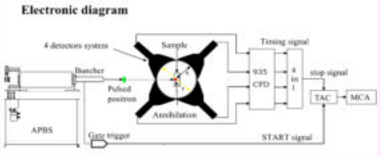
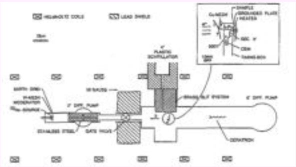


Fig. 1. Experimental geometry of TOF apparatus: a section view of TOF chamber is shown.
</td></tr>
<tr><td>RI source
low intensity, spin pol.</td><td>
IHEP



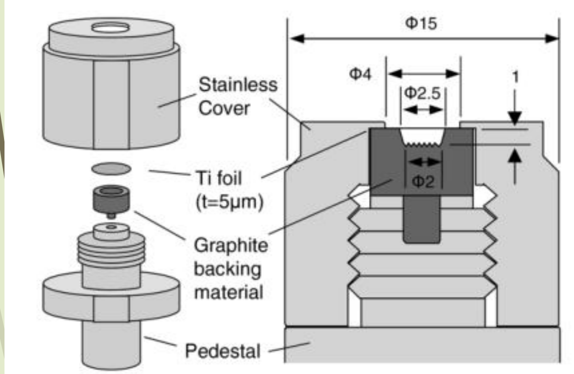
London univ, ETHZ


</td><td>no one</td></tr>
</table>

Harder than we thought…(extremely low counting rate)

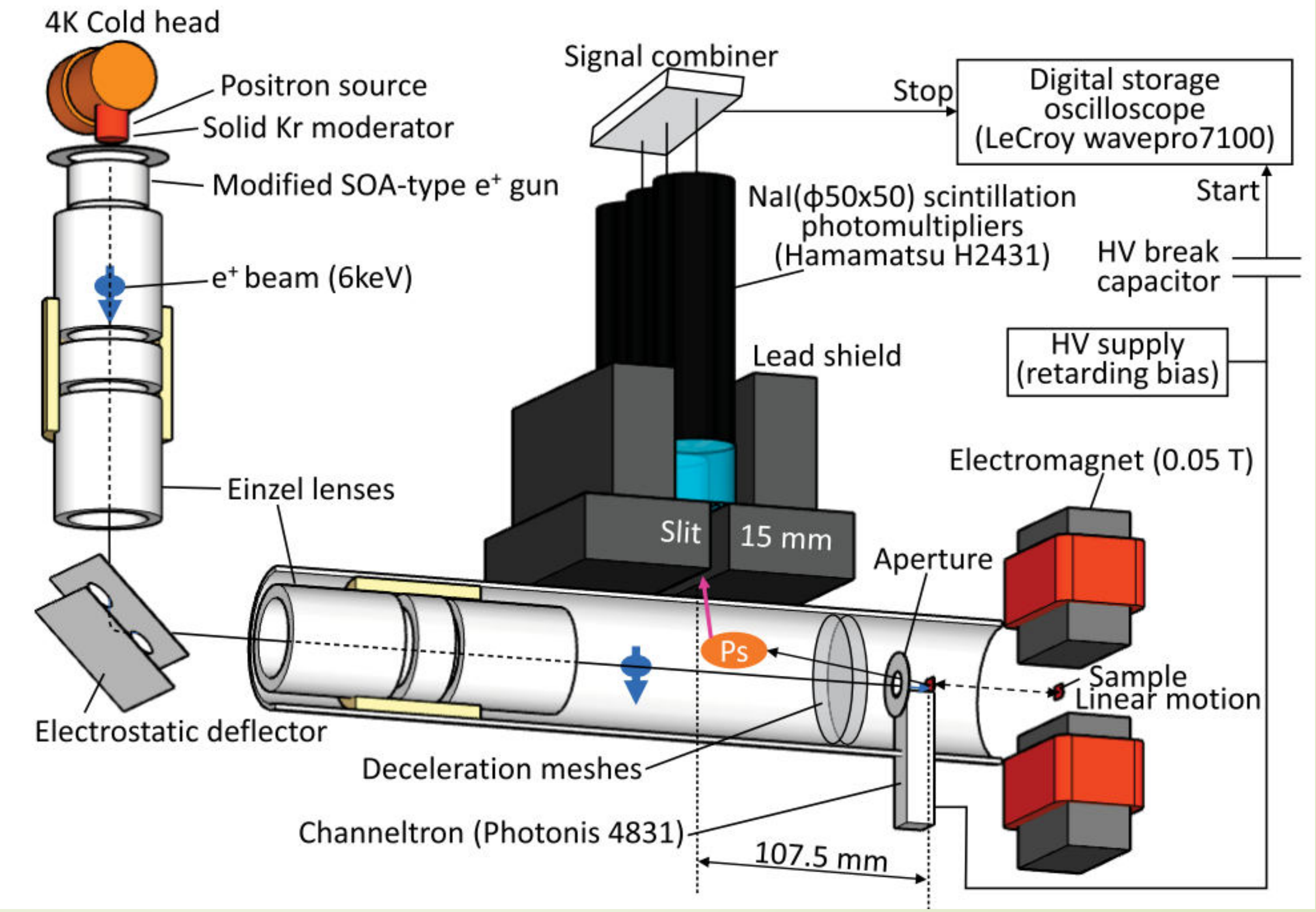
Spin-polarized Ps-TOF (SP-PsTOF)

✓ **Construction a Ps-TOF apparatus using spin-polarized positron beam.**



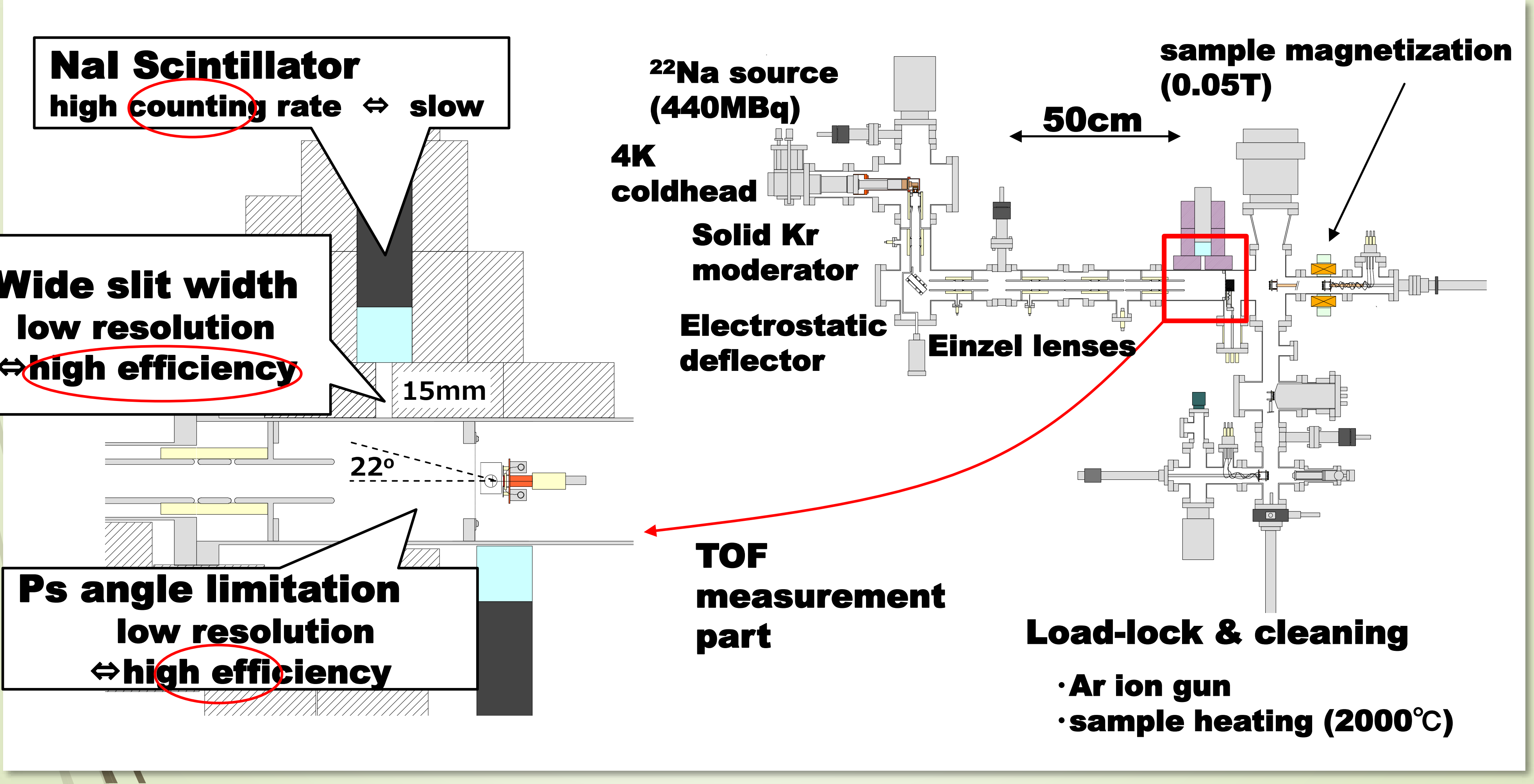
Spin-polarized positron source
NIMB480(2020)49

Phys. Rev. Lett.126(2021)186401



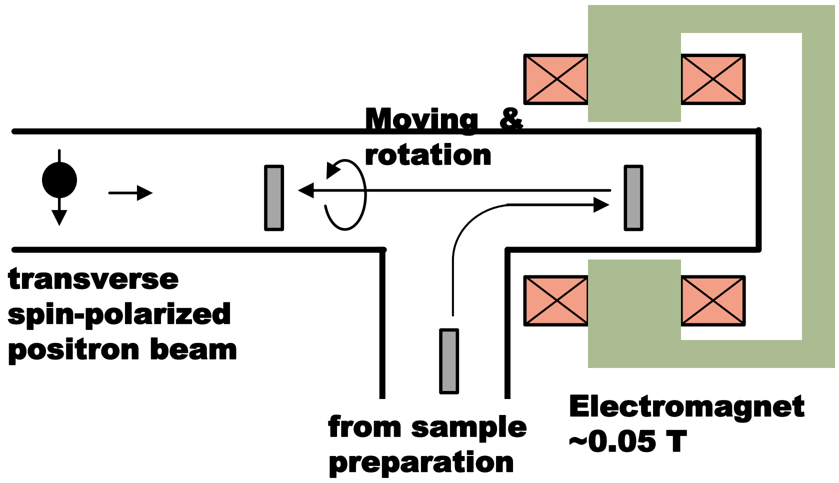
TOF detector

- ✓ **Keeping counting rate is the highest priority.**



Sample magnetization

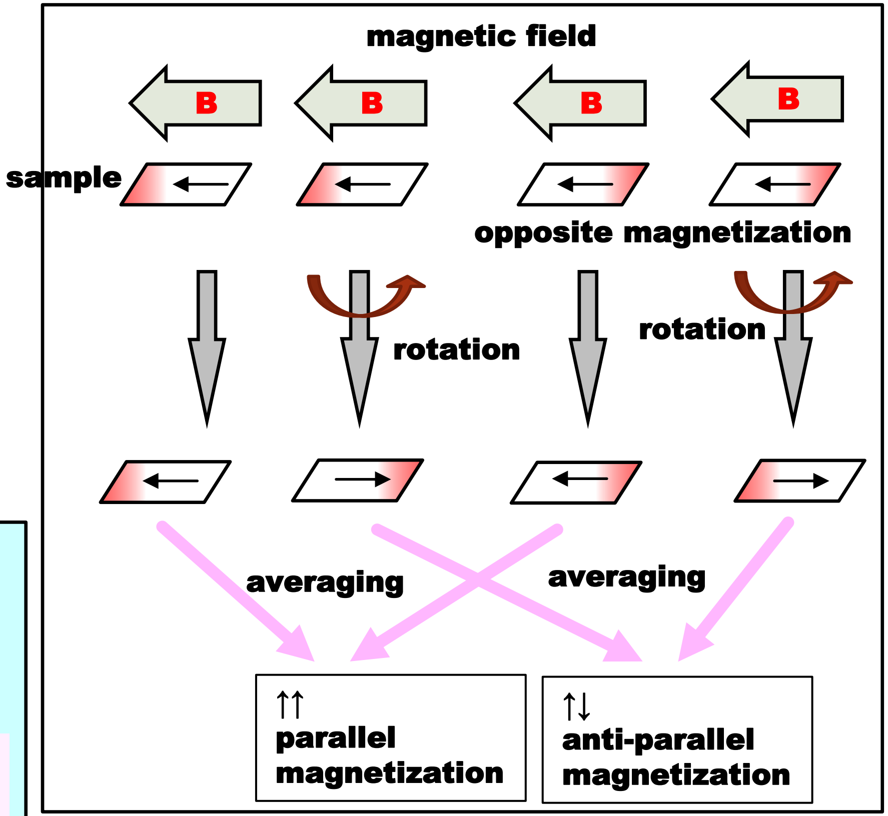
- ✓ **Sample magnetization by external magnetic field of 0.05T.**
- ✓ **The spectrum asymmetricity was obtained from the spin parallel / antiparallel TOF spectra.**



TOF spectrum Neg. field : $I^{\uparrow}(E)$
Pos. field : $I^{\downarrow}(E)$

↓

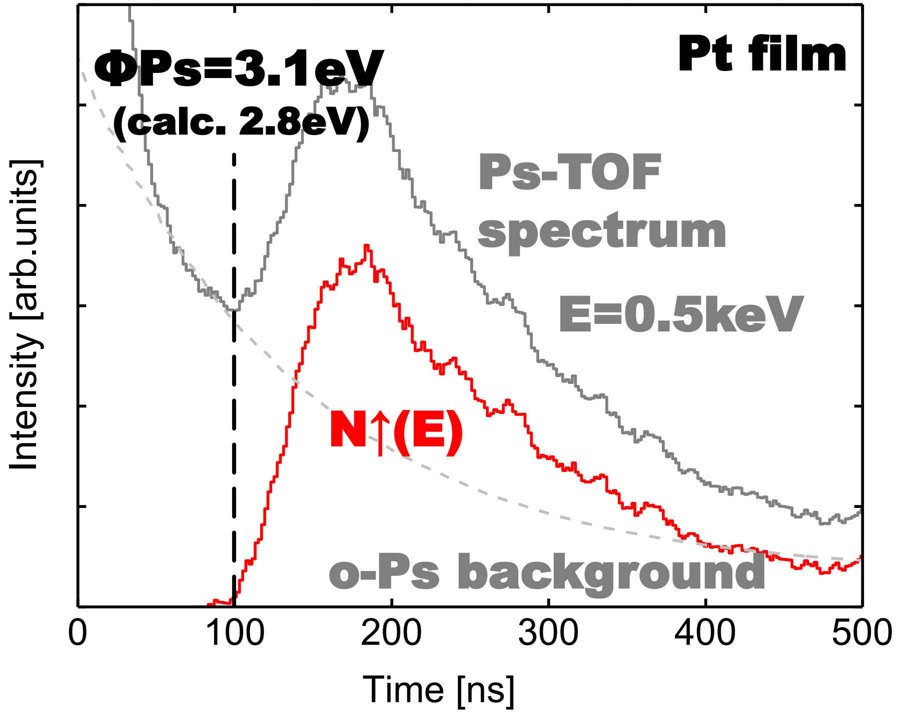
Asymmetricity $$A^{oPs} = \frac{I^{\uparrow}(E) - I^{\downarrow}(E)}{I^{\uparrow}(E) + I^{\downarrow}(E)} \propto P_{-}(E)$$



Cancellation of position dependency

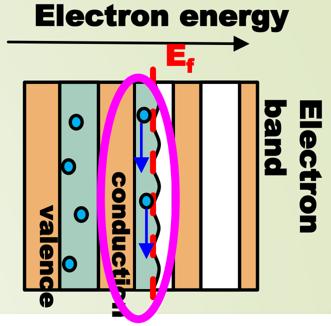
SP-PsTOF result（Pt film: non-magnetic）

- ✓ **The differential Ps-TOF spectrum was measured for the non-magnetic Pt thin film.**

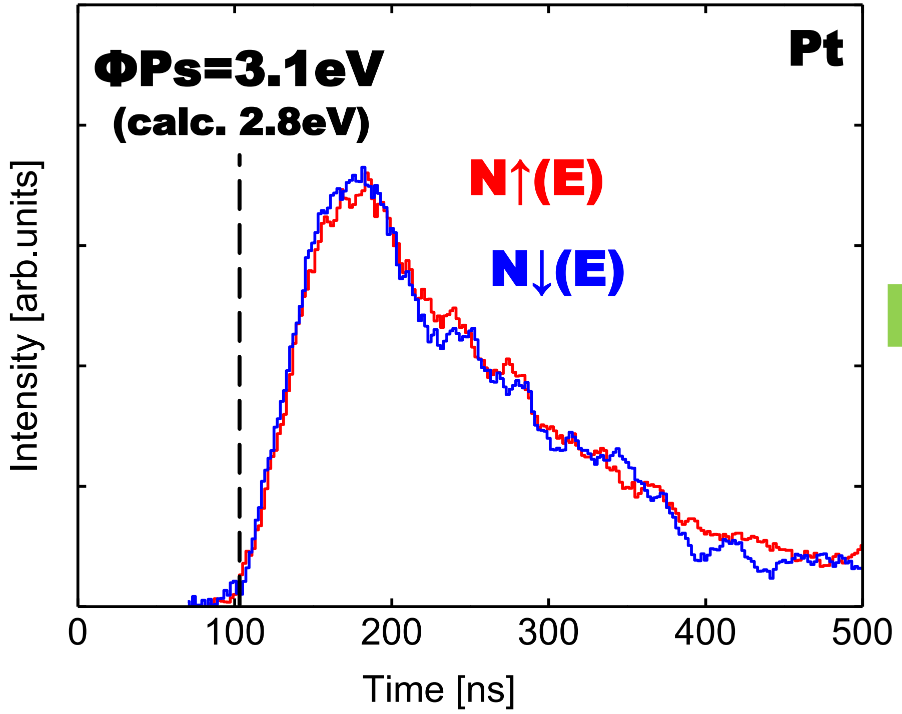


TOF time spectrum

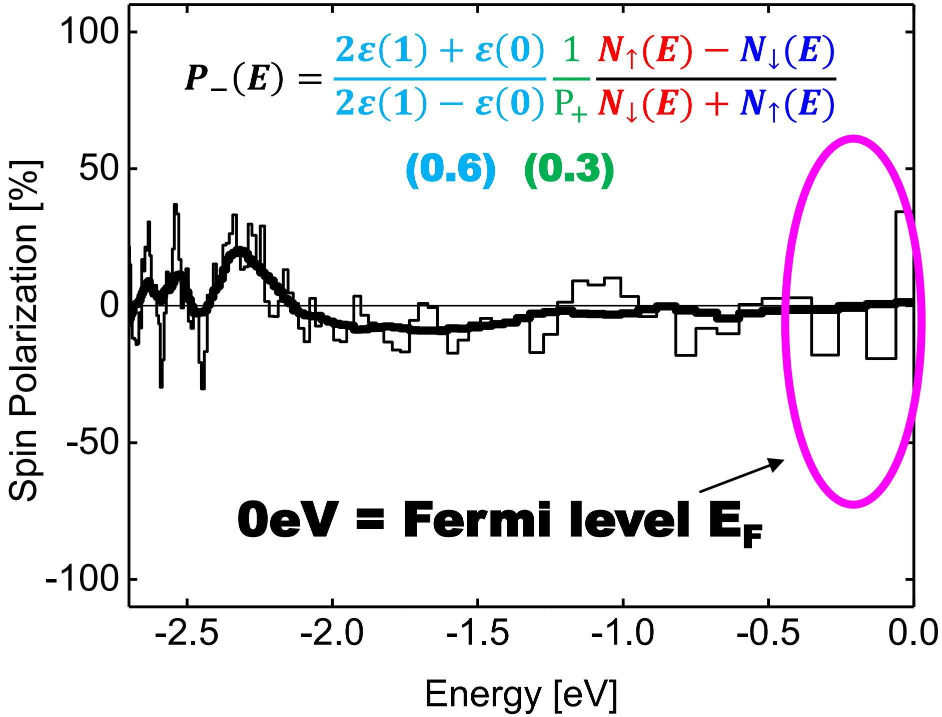
SP-PsTOF result（Pt film: non-magnetic）



- ✓ **Difference PsTOF by alternating magnetization.**
  TOF → Ps energy (adjusted to Ef=0)
  Intensity asymmetricity → electron spin polarization
- ✓ **Spin polarization ～0% (non-magnetic)**



PsTOF time spectrum



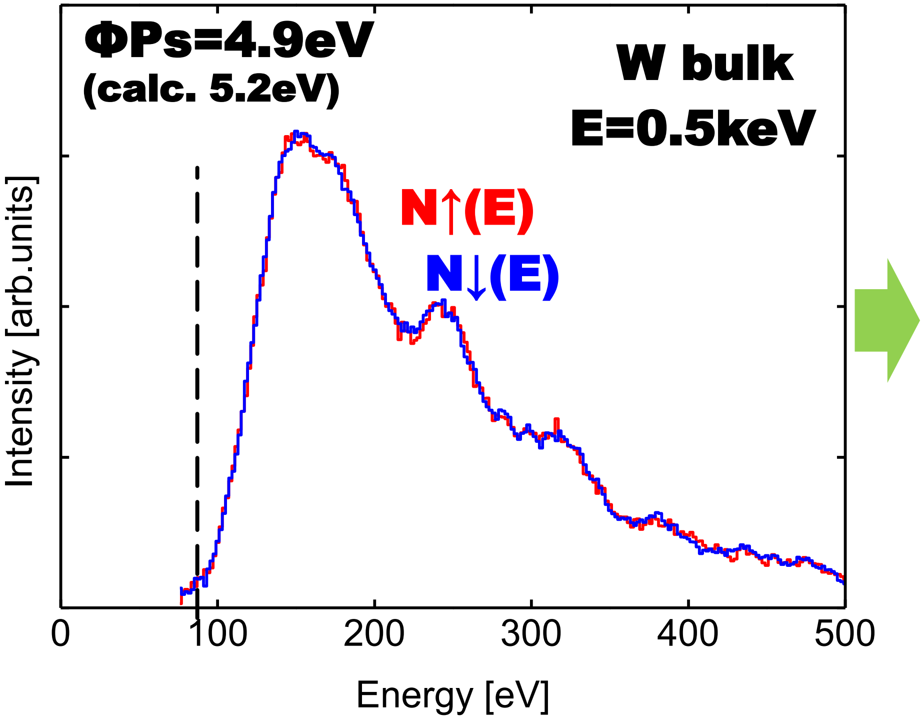
$$P_{-}(E) = \frac{2\varepsilon(1)+\varepsilon(0)}{2\varepsilon(1)-\varepsilon(0)} \frac{1}{P_{+}} \frac{N_{\uparrow}(E)-N_{\downarrow}(E)}{N_{\downarrow}(E)+N_{\uparrow}(E)}$$

(0.6) (0.3)

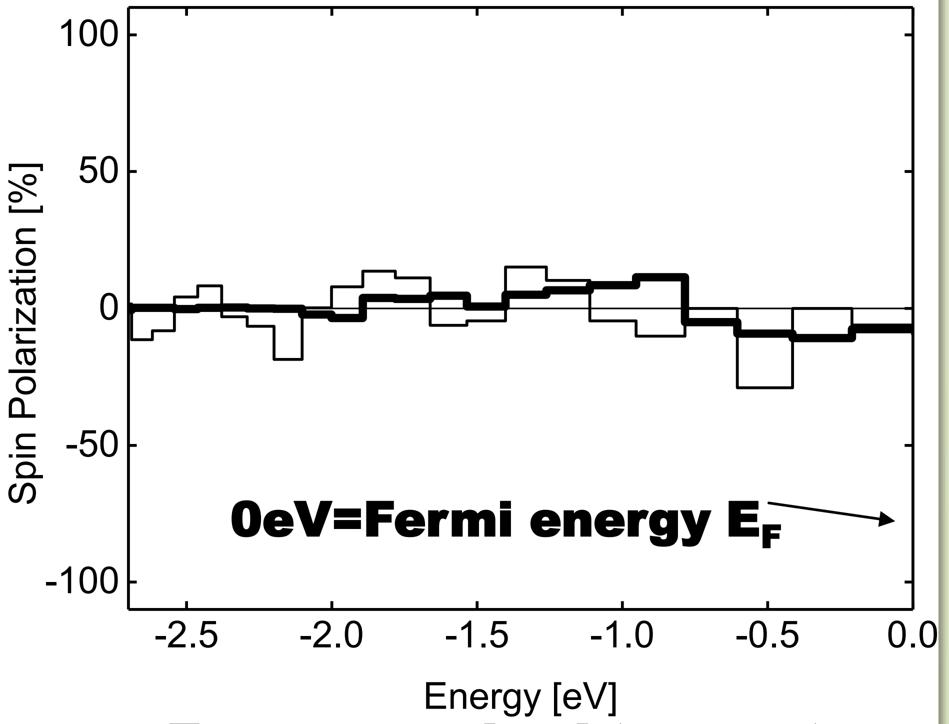
Energy-resolved topmost electron spin polarization

SP-PsTOF result（W bulk: non-magnetic）

- ✓ **The differential Ps-TOF spectrum was measured for the non-magnetic W bulk crystal.**
- ✓ **Spin polarization ～0% (non-magnetic)**



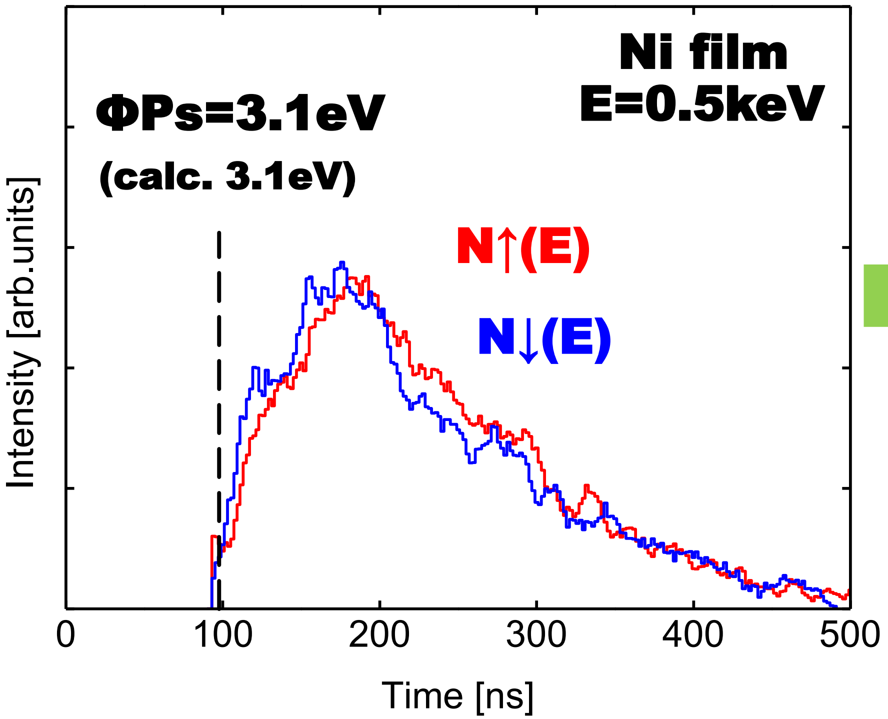
PsTOF time spectrum



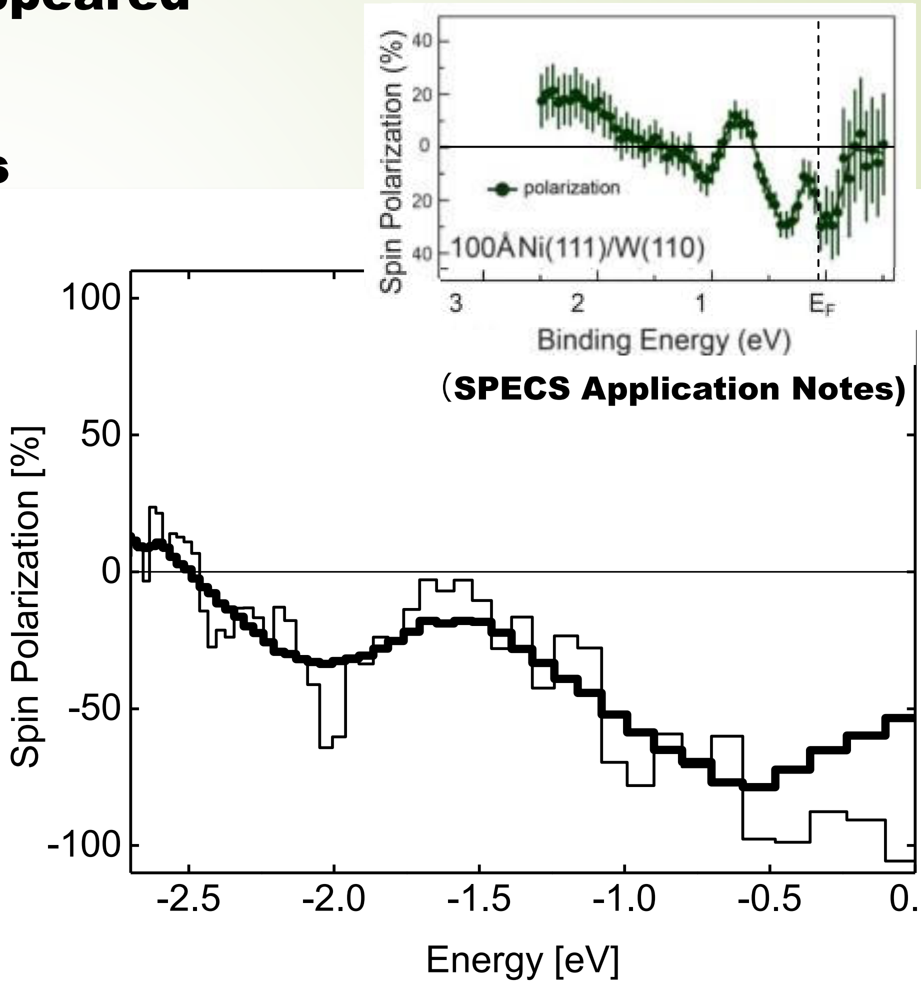
Energy-resolved topmost electron spin polarization

SP-PsTOF result（Ni film: magnetic）

- ✓ **Clear megative polarization is appeared near the Fermi level.**
- ✓ **Same tendency as other methods**



PsTOF time spectrum

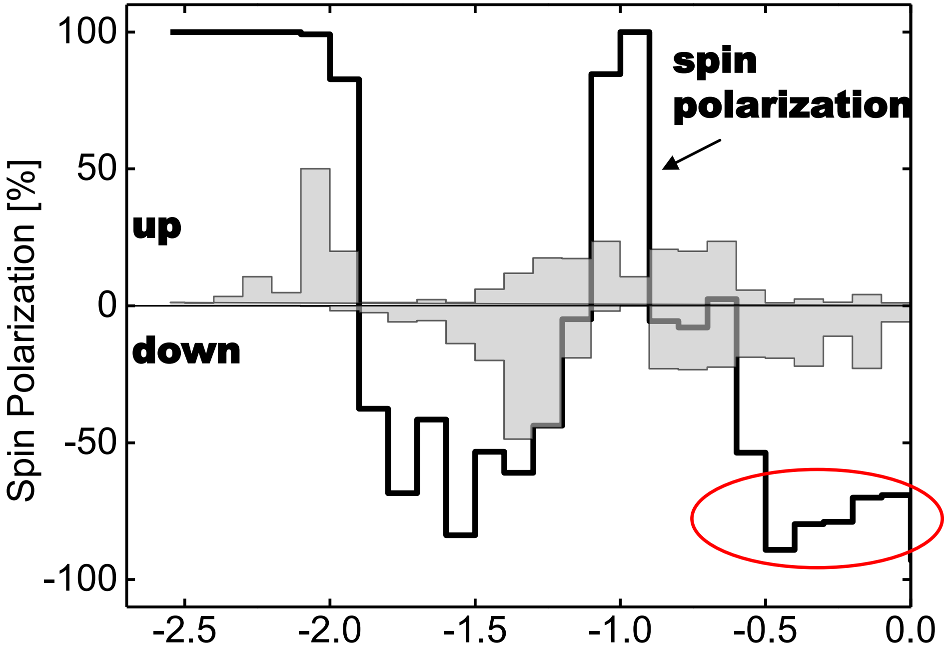


（SPECS Application Notes）

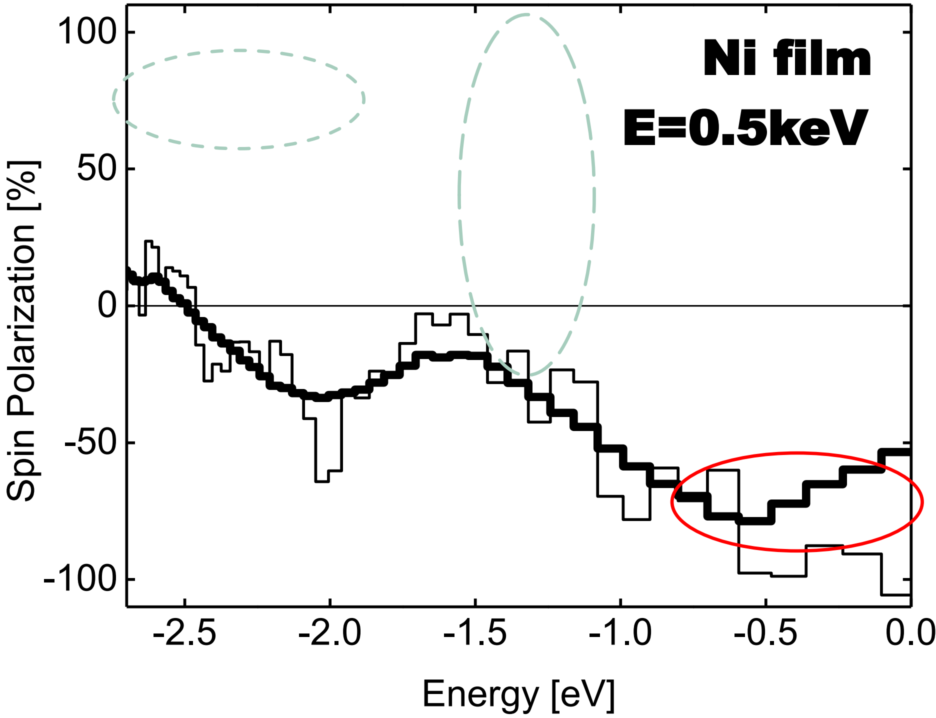
Energy-resolved topmost electron spin polarization

SP-PsTOF result（Ni film: magnetic）

- ✓ **Comparison with first-principles calculation.**
- ✓ **Experimental and calculation show basically the same tendency, but they seem not to be exactly the same.**



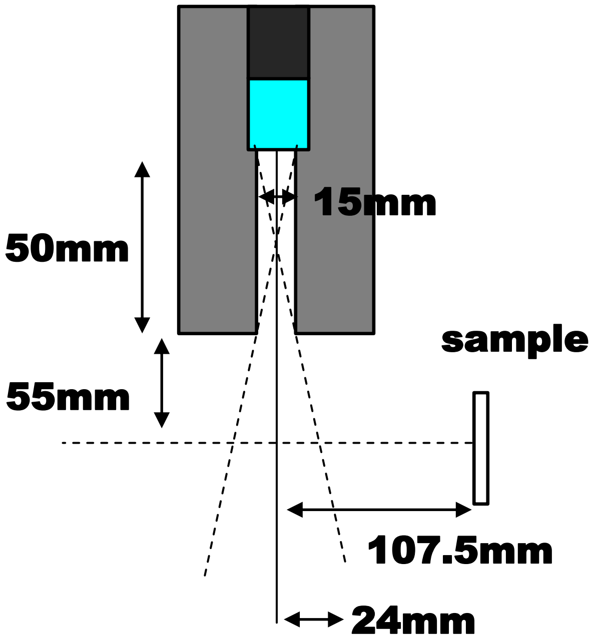
Calculated spin DOS



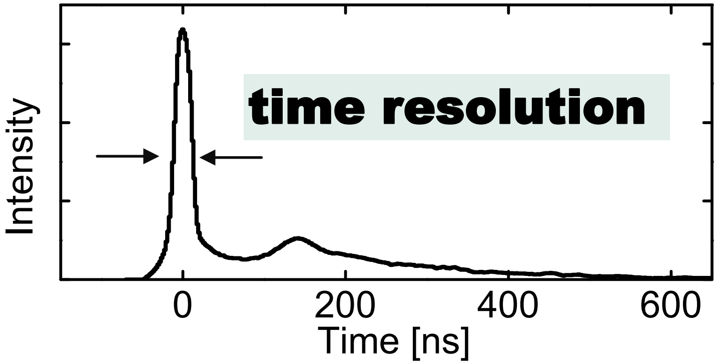
Experimental result

Convolution of energy resolution

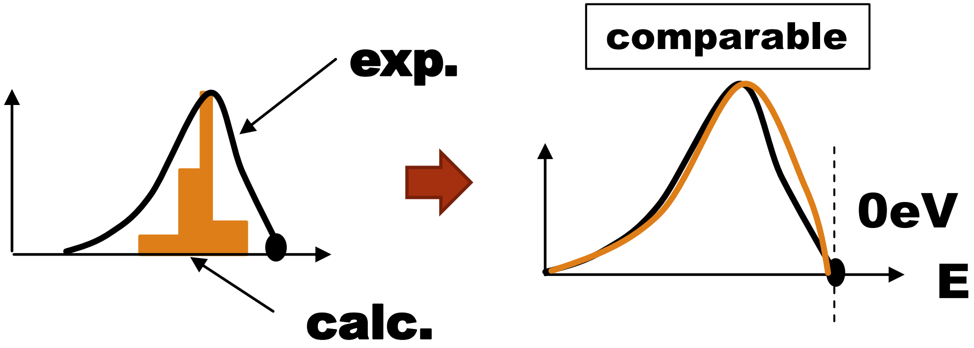
- ✓ Computational DOS had to be convolved at the resolution of the experimental setup.



special resolution

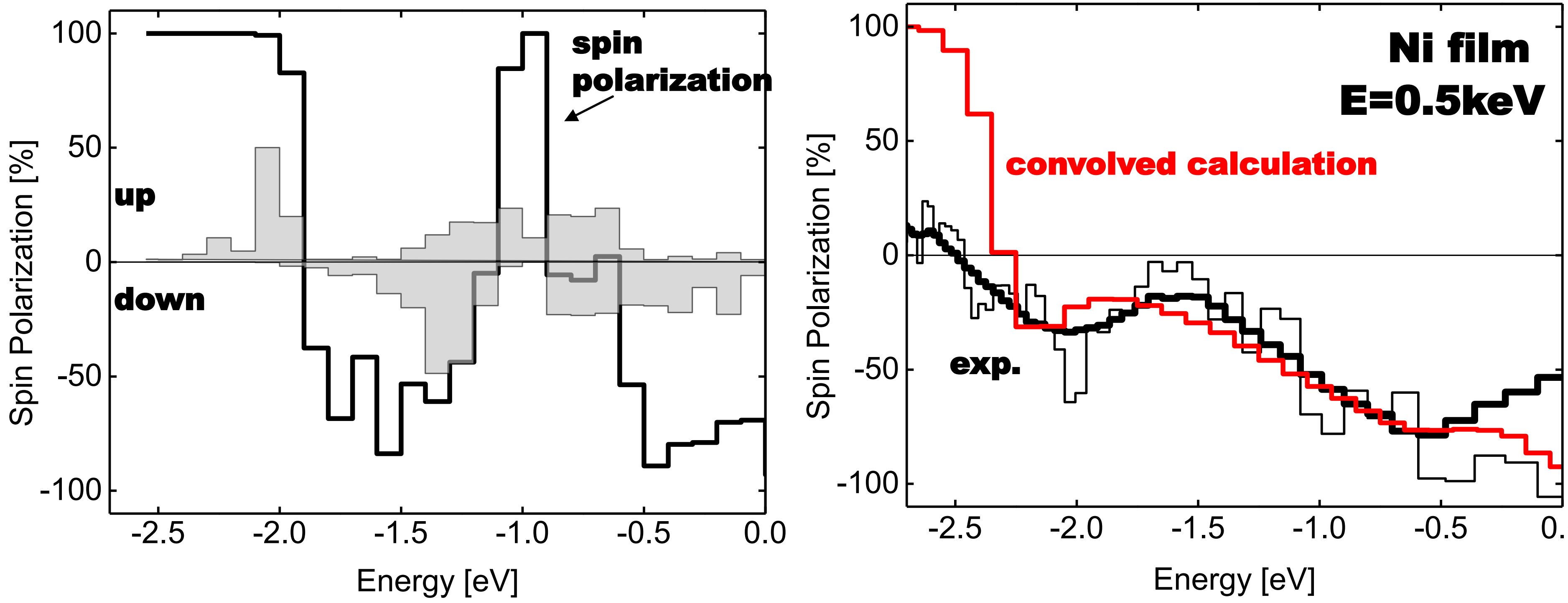


convolution of energy resolution



SP-PsTOF result（Ni film: magnetic）

- ✓ **For the Ni thin film, a negative polarization was detected near the Fermi surface.**
- ✓ **well reproduced with experimental result**

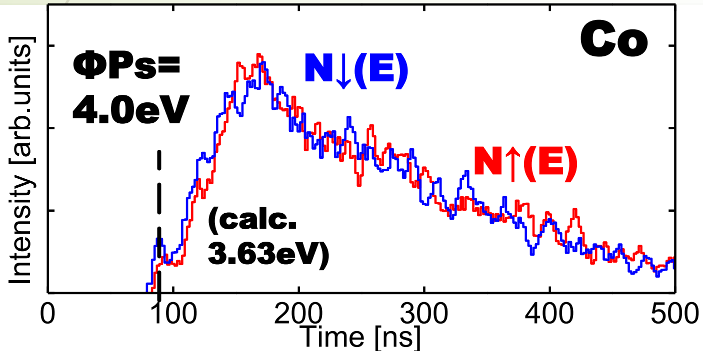


Calculated spin DOS **Experimental result**

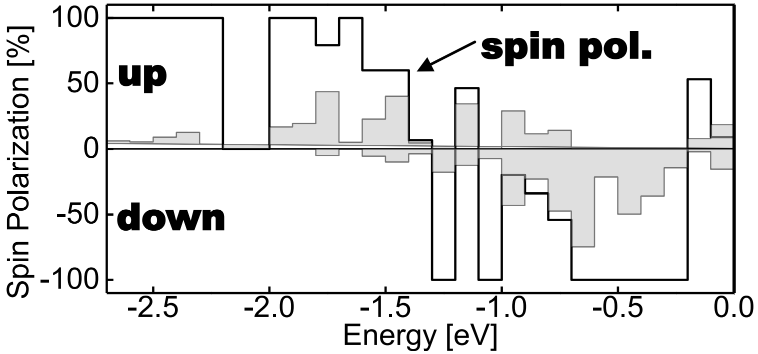
SP-PsTOF result（Co film: magnetic）

✓ **As another example of a ferromagnetic material, Co film was also measured.**

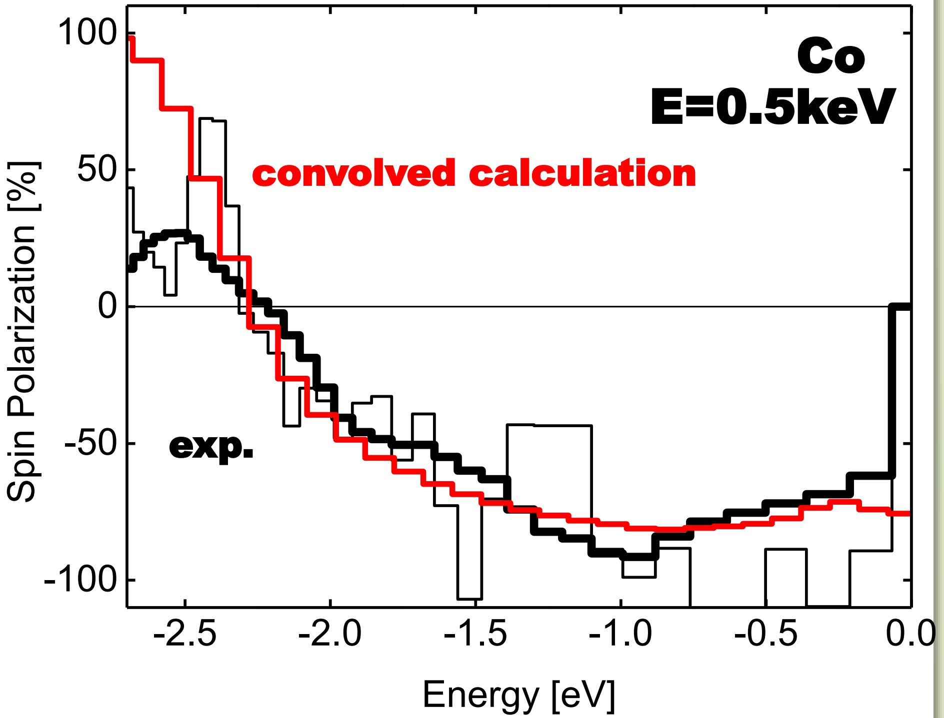
✓ **Like Ni sample, a negative polarization was observed.**



PsTOF time spectrum



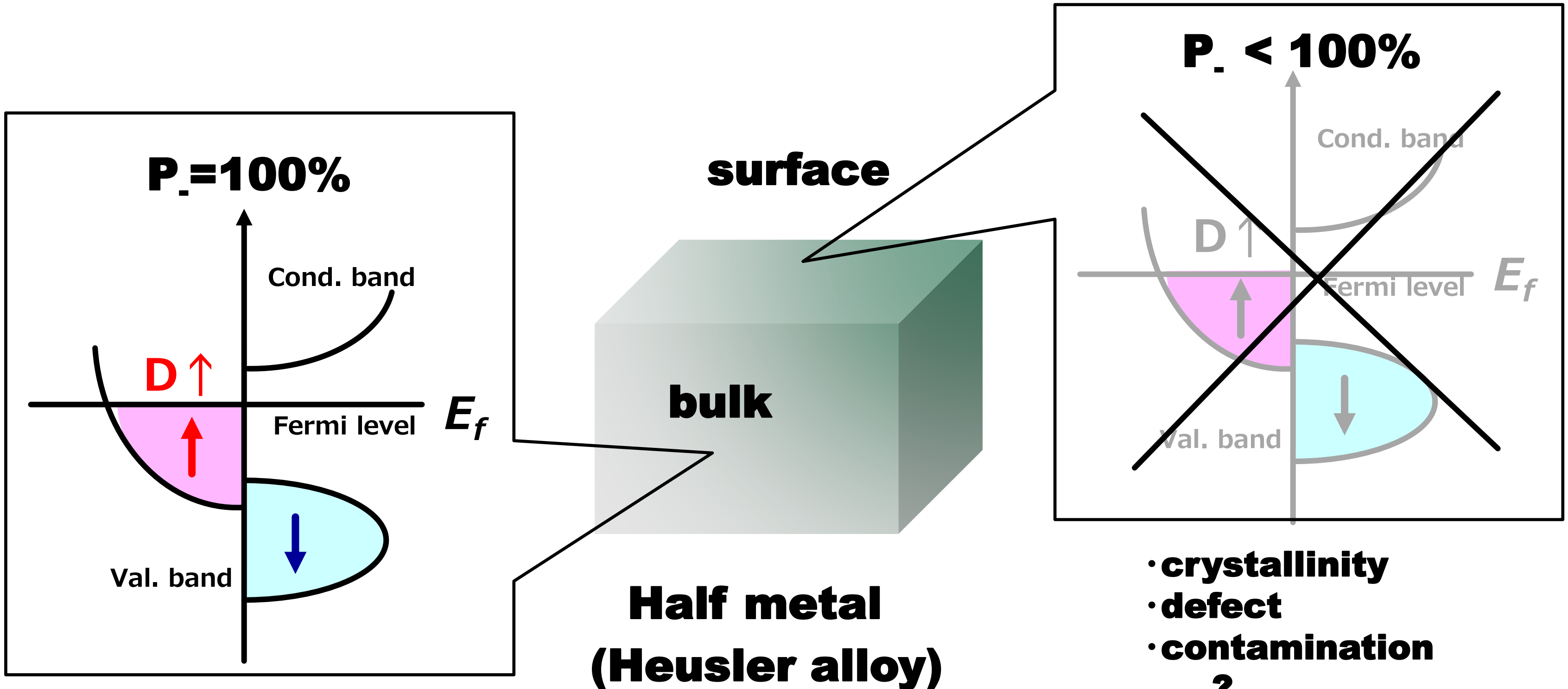
Calculated spin DOS



Energy-resolved topmost electron spin polarization

SP-PsTOF for Half metal

- ✓ Half metal has a 100% polarization near the Fermi level in bulk, but not on the surface. The reason is still unclear.
- ✓ The SP-PsTOF measurement is suitable for evaluating the surface spin polarization of half metal.



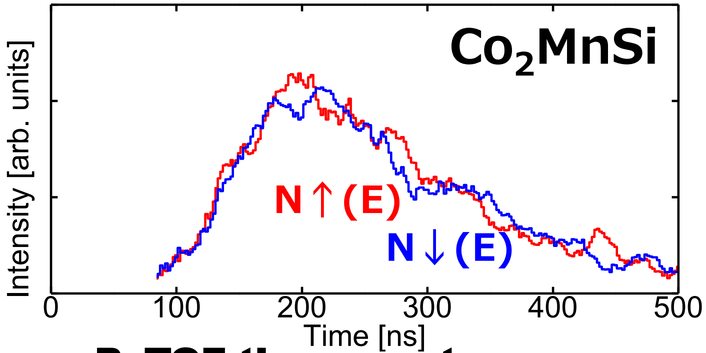
Half metal (Heusler alloy)

- ･crystallinity
- ･defect
- ･contamination

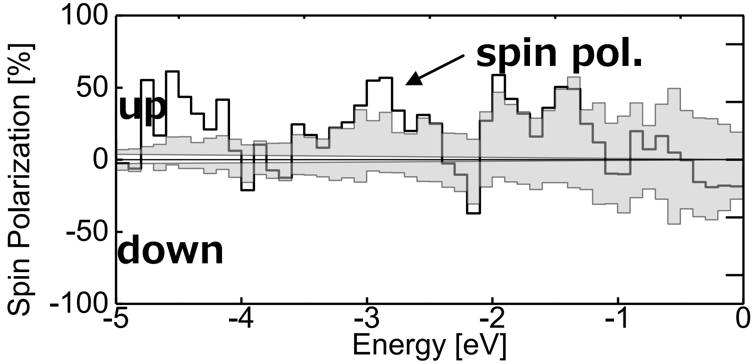
…?

SP-PsTOF result（Co_2MnSi film）

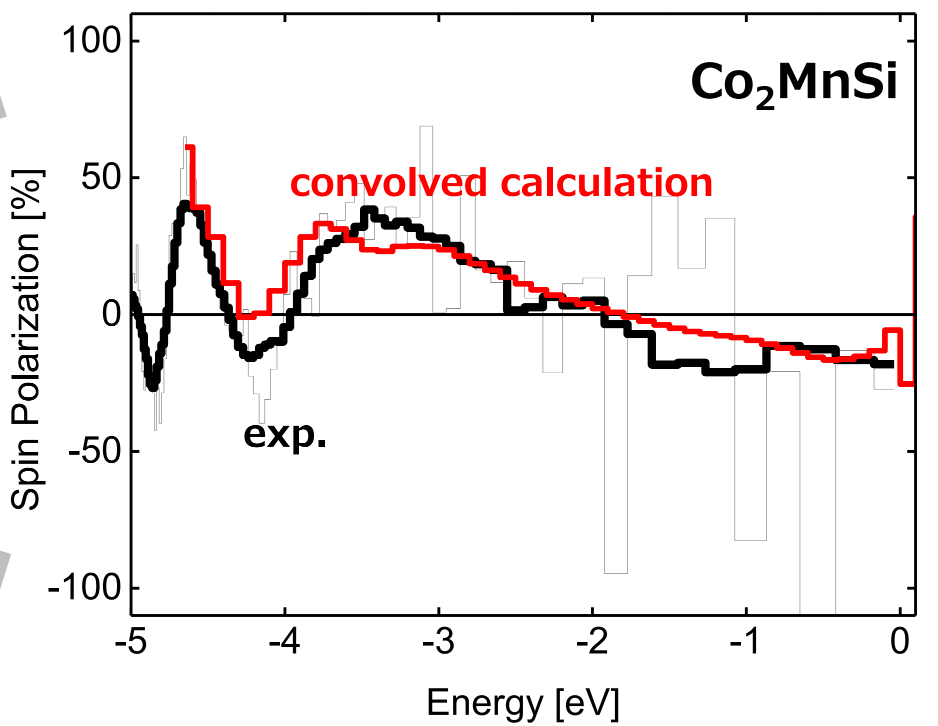
- ✓ Energy-resolved spin polarization of Co_2MnSi (CMS) Heusler alloy film was obtained.
- ✓ Spin polarization near the Fermi level is very weak.



PsTOF time spectrum



Calculated spin DOS

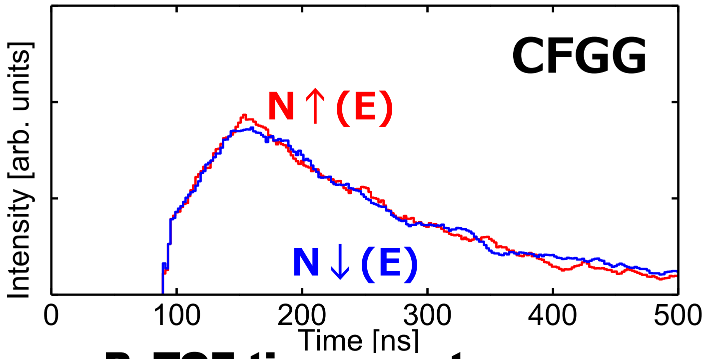


Energy-resolved topmost electron spin polarization

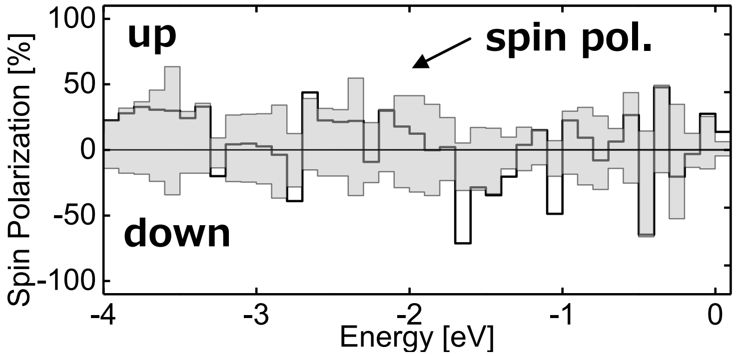
SP-PsTOF result（Co_2FeGa0.5Ge0.5 film）

✓ **Energy-resolved spin polarization of $Co_2FeGa_{0.5}Ge_{0.5}$ (CFGG) Heusler alloy film was obtained.**

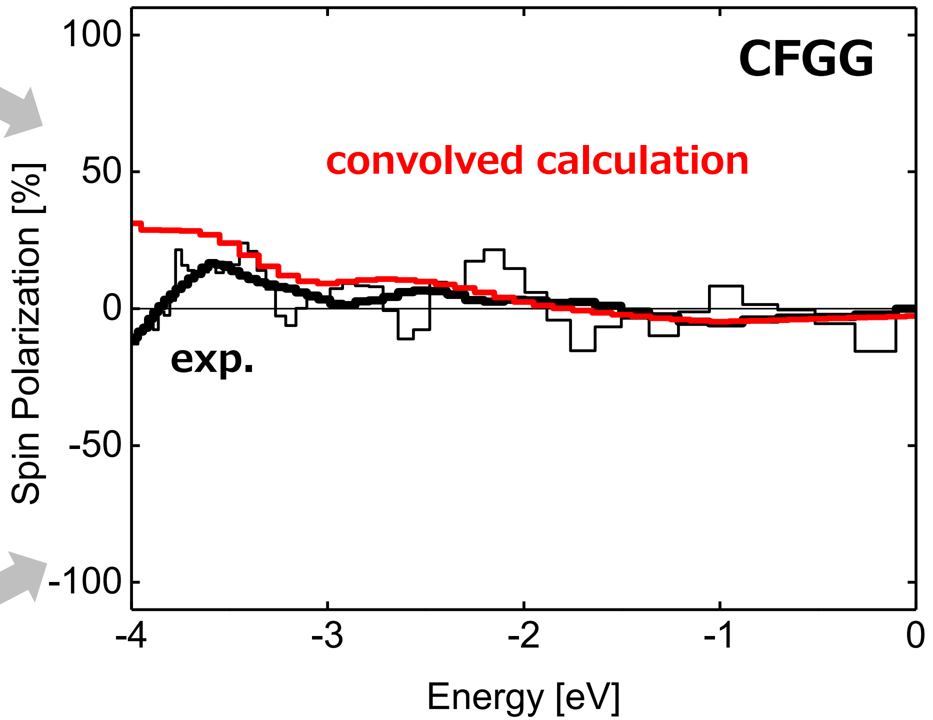
✓ **Spin polarization of surface electrons is very small.**



PsTOF time spectrum



Calculated spin DOS

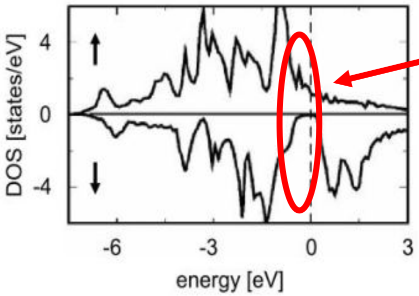


Energy-resolved topmost electron spin polarization

Improvement of SP-PsTOF apparatus

✓**For better measurements, improving the energy resolution is unavoidable.**

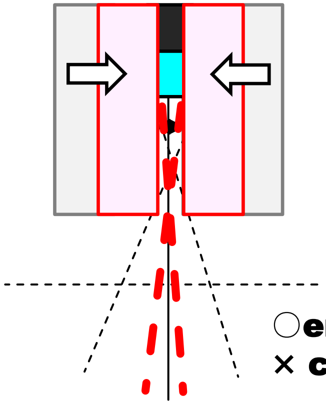
Co_2MnSi spin-polarized DOS



Below Fermi level ~ 0.5eV is polarized
→ Energy resolution of ~ 0.5eV is required.

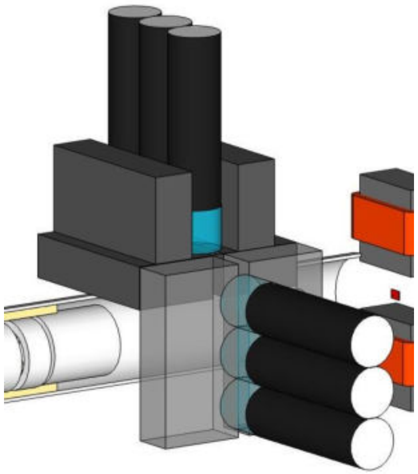
Present energy spread **1.4eV** @3eV

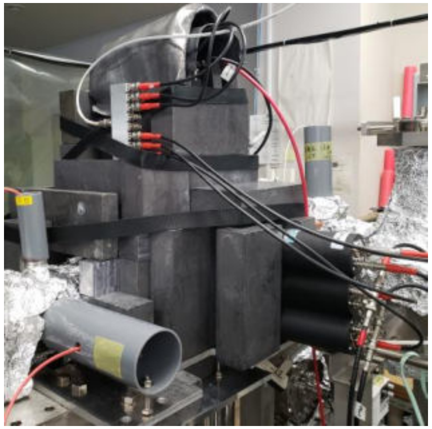
Limitation of lead slit width



○energy resolution
× counting rate

Additional detectors were installed

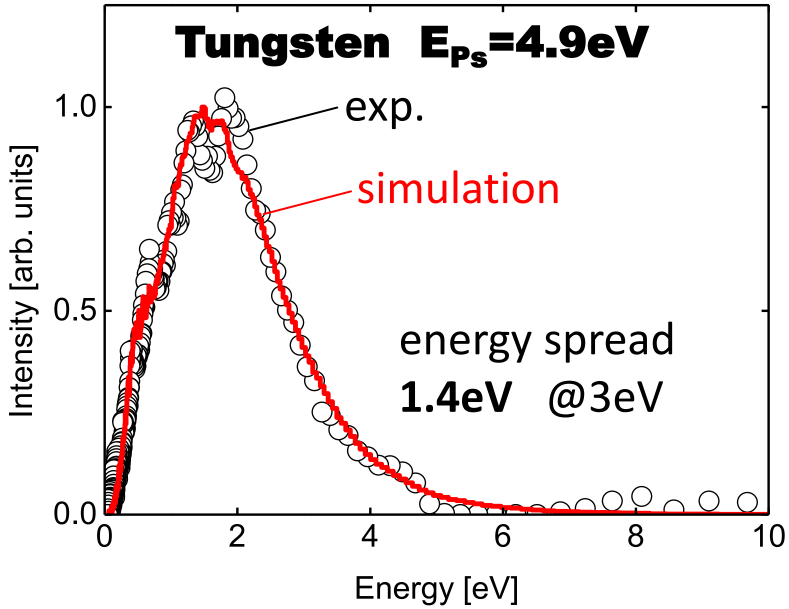




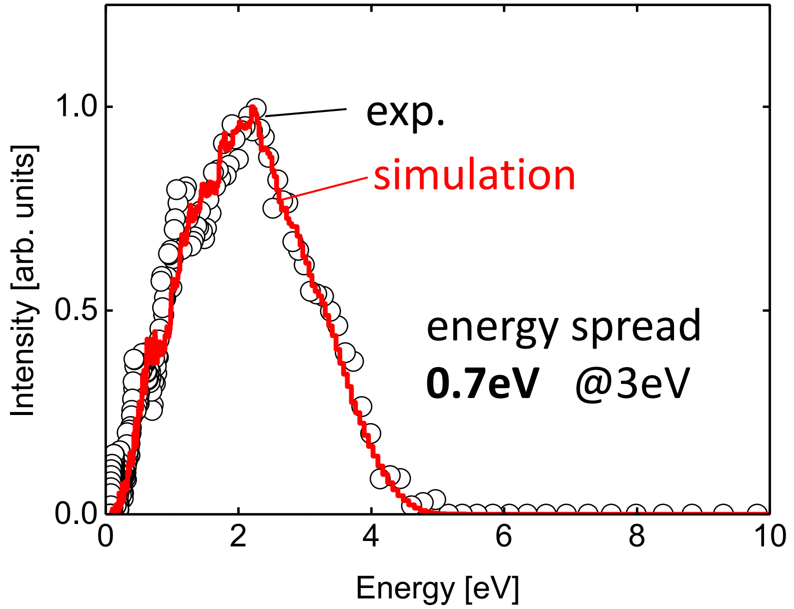
Improvement of PsTOF spectrum

- ✓ **We try to find the best balance between counting rate and energy resolution.**

slit width=15 mm
detector= 3 pcs.
Est. time resolution=30 ns
counting rate= 0.2-0.3 cps



slit width=8 mm
detector= 6 pcs.
Est. time resolution=5 ns
counting rate= 0.05-0.1 cps



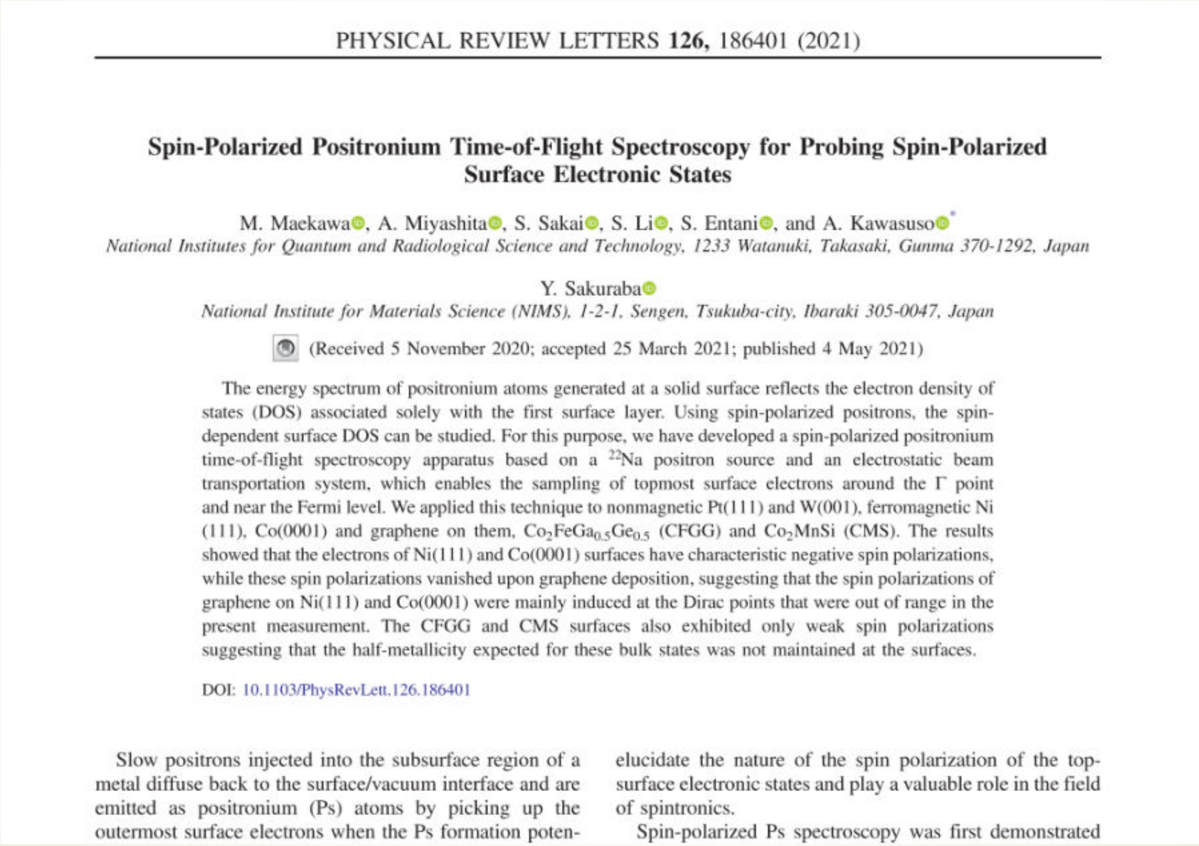
Summary

✓ Spin-polarized Ps TOF apparatus has been constructed.

✓ We succeeded in obtaining the first SP-PsTOF.
  - Clear asymmetry for ferromagnetic sample near E=ΦPs.

✓ In the Future, we have plan to measure the spintronics materials.

Thank you for your attention!



PHYSICAL REVIEW LETTERS **126,** 186401 (2021)

Spin-Polarized Positronium Time-of-Flight Spectroscopy for Probing Spin-Polarized Surface Electronic States

M. Maekawa, A. Miyashita, S. Sakai, S. Li, S. Entani, and A. Kawasuso*
National Institutes for Quantum and Radiological Science and Technology, 1233 Watanuki, Takasaki, Gunma 370-1292, Japan

Y. Sakuraba
National Institute for Materials Science (NIMS), 1-2-1, Sengen, Tsukuba-city, Ibaraki 305-0047, Japan

(Received 5 November 2020; accepted 25 March 2021; published 4 May 2021)

The energy spectrum of positronium atoms generated at a solid surface reflects the electron density of states (DOS) associated solely with the first surface layer. Using spin-polarized positrons, the spin-dependent surface DOS can be studied. For this purpose, we have developed a spin-polarized positronium time-of-flight spectroscopy apparatus based on a ^{22}Na positron source and an electrostatic beam transportation system, which enables the sampling of topmost surface electrons around the Γ point and near the Fermi level. We applied this technique to nonmagnetic Pt(111) and W(001), ferromagnetic Ni(111), Co(0001) and graphene on them, $Co_2FeGa_{0.5}Ge_{0.5}$ (CFGG) and Co_2MnSi (CMS). The results showed that the electrons of Ni(111) and Co(0001) surfaces have characteristic negative spin polarizations, while these spin polarizations vanished upon graphene deposition, suggesting that the spin polarizations of graphene on Ni(111) and Co(0001) were mainly induced at the Dirac points that were out of range in the present measurement. The CFGG and CMS surfaces also exhibited only weak spin polarizations suggesting that the half-metallicity expected for these bulk states was not maintained at the surfaces.

DOI: 10.1103/PhysRevLett.126.186401

Slow positrons injected into the subsurface region of a metal diffuse back to the surface/vacuum interface and are emitted as positronium (Ps) atoms by picking up the outermost surface electrons when the Ps formation poten-

elucidate the nature of the spin polarization of the top-surface electronic states and play a valuable role in the field of spintronics.

Spin-polarized Ps spectroscopy was first demonstrated

PHYSICAL REVIEW LETTERS 126, 186401 (2021)

Field asymmetry of o-Ps intensity

Number of positrons and electrons having up(↑) and down(↓) spins

$$n^{\uparrow} = \frac{1+P_{+}}{2} \qquad n^{\downarrow} = \frac{1-P_{+}}{2}$$

$$n^{\uparrow} = D_{maj}(E) \qquad n^{\downarrow} = D_{min}(E)$$

P_+ : Positron spin polarization

D(E) : electron density of state

Number of o-Ps

magnetic field	|11>	|1-1>	|10>
negative	$n^{\uparrow}n^{\uparrow}$	$n^{\downarrow}n^{\downarrow}$	$1/2(n^{\uparrow}n^{\downarrow}+n^{\downarrow}n^{\uparrow})$
positive	$n^{\uparrow}n^{\downarrow}$	$n^{\downarrow}n^{\uparrow}$	$1/2(n^{\uparrow}n^{\uparrow}+n^{\downarrow}n^{\downarrow})$

Fraction of o-Ps

$$F^{oPs} = \varepsilon_1\left(F_{|11>} + F_{|1-1>}\right) + \varepsilon_0 F_{|10>}$$

(ε_1,ε_0 : detection efficiency)

Spectrum asymmetricity

$$A^{oPs} = \frac{F^{oPs}_{Neg.field} - F^{oPs}_{Pos.field}}{F^{oPs}_{Neg.field} + F^{oPs}_{Pos.field}} = \frac{P_{+}(D_{maj}(E) - D_{min}(E)) \times \left(\varepsilon_1 - \frac{\varepsilon_0}{2}\right)}{(D_{maj}(E) + D_{min}(E)) \times \left(\varepsilon_1 + \frac{\varepsilon_0}{2}\right)}$$

$$= \frac{(2\varepsilon_1 - \varepsilon_0)}{(2\varepsilon_1 + \varepsilon_0)} P_{+} \times P_{-}\,(E)$$

$$P_{-}(E) = \frac{D_{maj}(E) - D_{min}(E)}{D_{maj}(E) + D_{min}(E)}$$

P_-(E) : spin-polarized electron density of state

$$P_{-}(E) \propto A^{oPs}$$

actually,

$$P_{-}(E) \propto \frac{1}{f_{Ps}(E)} A^{oPs}(E)$$

f_{Ps}:Level dependence of Ps formation probability