# Development and Application of Spin-Polarized Positron Beam with Radioisotopes

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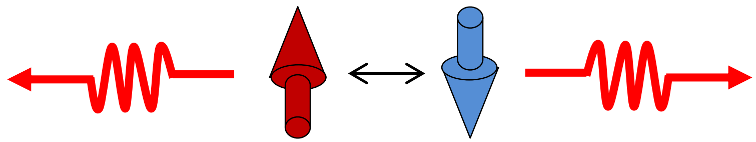
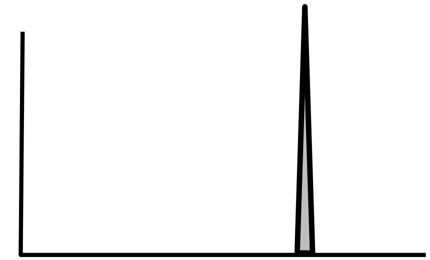
*Financial support : JSPS KAKENHI under Grant No. 24310072,15K14135,17K19061.*

# Contents:

# Spin dependent annihilation

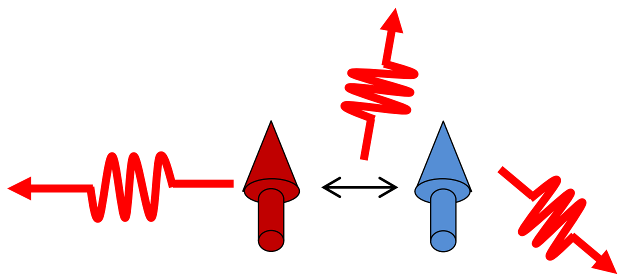
**Total spin <u>S=0</u> : Two-photon emission**

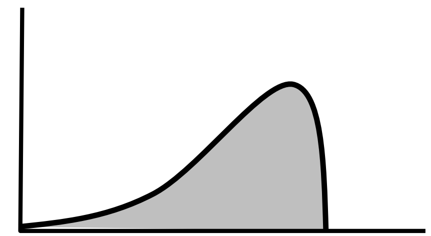




$m_0c^2$=511keV±ΔE

**Total spin <u>S=1</u> : Three-photon emission**





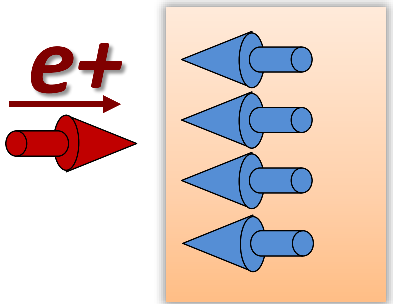
0～511keV continuous

## Electron spins are detected via

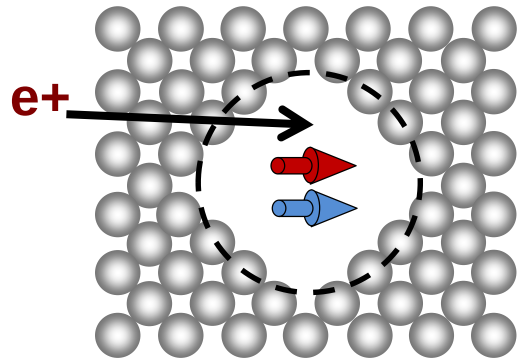
**Case A : 2γ-annihilation of *e*⁺ with unpaired electrons**

**Case B : 3γ-annihilation of Positronium (Ps)**

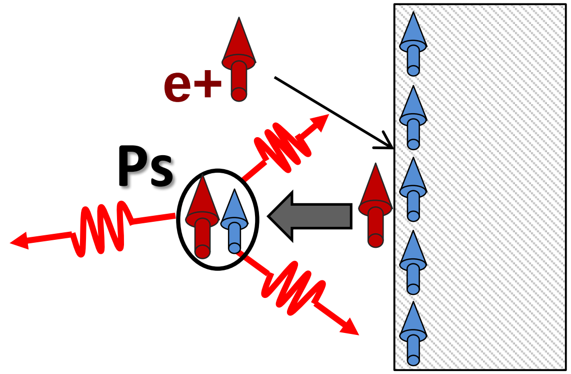
## Ferromagnetic band structure




## Vacancy-induced magnetism



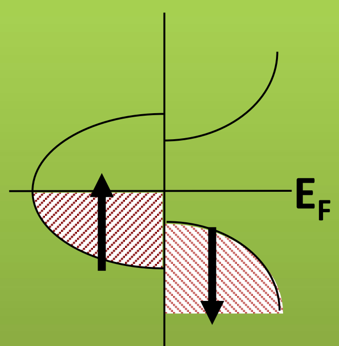

## Surface spin polarization




Maybe More…

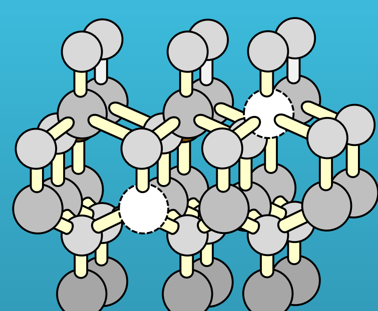
## Several potential applications

### Half-Metals



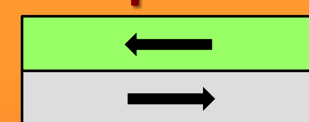

Heusler alloys
Metal oxide

### Magnetic semiconductor

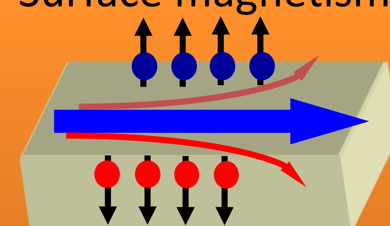


GaN, InN, $SnO_2$, $CeO_2$…

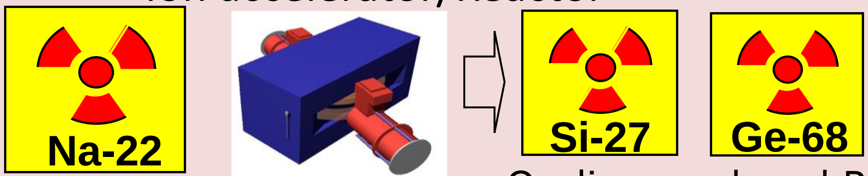
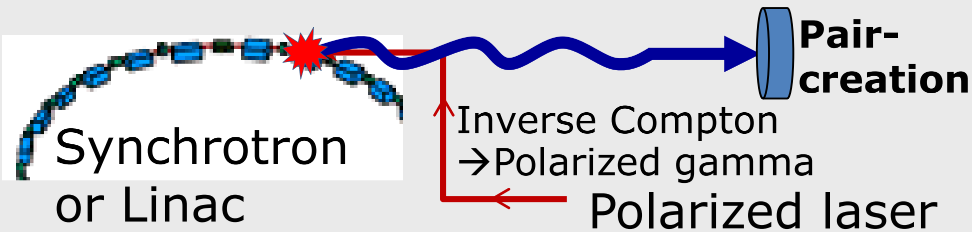
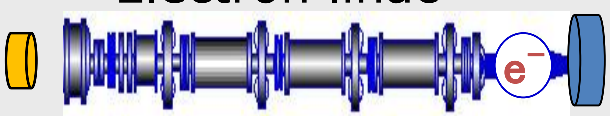
### Surface phenomena

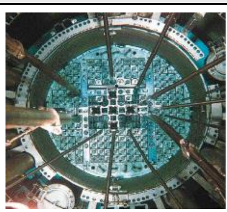


Surface magnetism

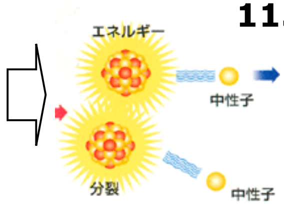


Spin Hall effect, Rashba effect
Topological Insulator

| Method | Intensity/Pol. | Facility |
|---|---|---|
| Ion accelerator/Reactor   Commercial RI / On-line produced RI | $10^3$-$10^6 e^+$/sec<br>30-50% | Possible anywhere with radioisotopes |
|   | $10^5$-$10^7 e^+$/sec<br>~50% | KEK<br>ELI-NP<br>(Extreme Light Infrastructure) |
| Electron linac   Photocathode<br>Polarized electron → Polarized gamma | $10^5$-$10^7 e^+$/sec<br>~50%<br>@PC current of 0.1~1mA | Jefferson Lab |



Reactor

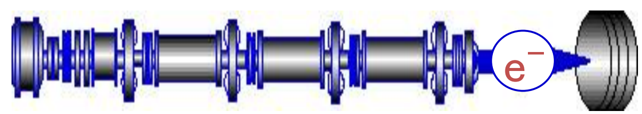



**$^{113}Cd(n,\gamma)^{114}Cd$**

Pair-creation

**$10^7$-$10^9 e^+$/sec**

**TUMunic, Delft Univ.**
**North Carolina, McMaster Univ.**
**Kyoto Univ.…**




Electron Linac

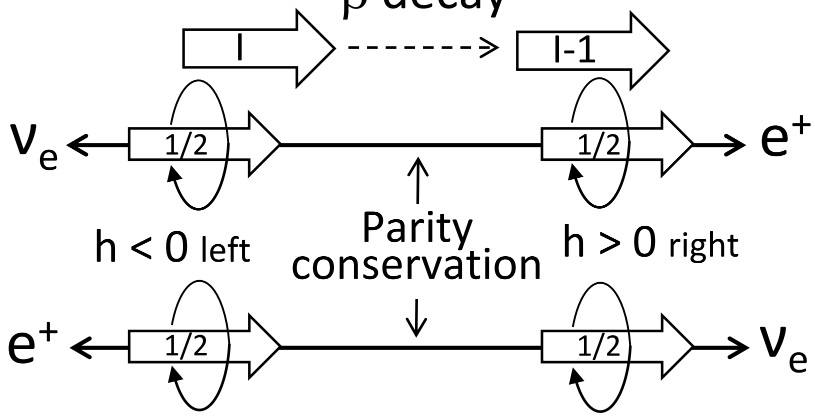
Pair-creation

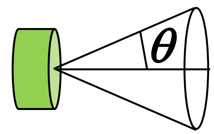
**$10^7$-$10^9 e^+$/sec**

**KEK, AIST**
**Rossendorf, Saclay, BeijingIHEP….**

**Unpolarized ⇨ Polarizer needed**

β decay



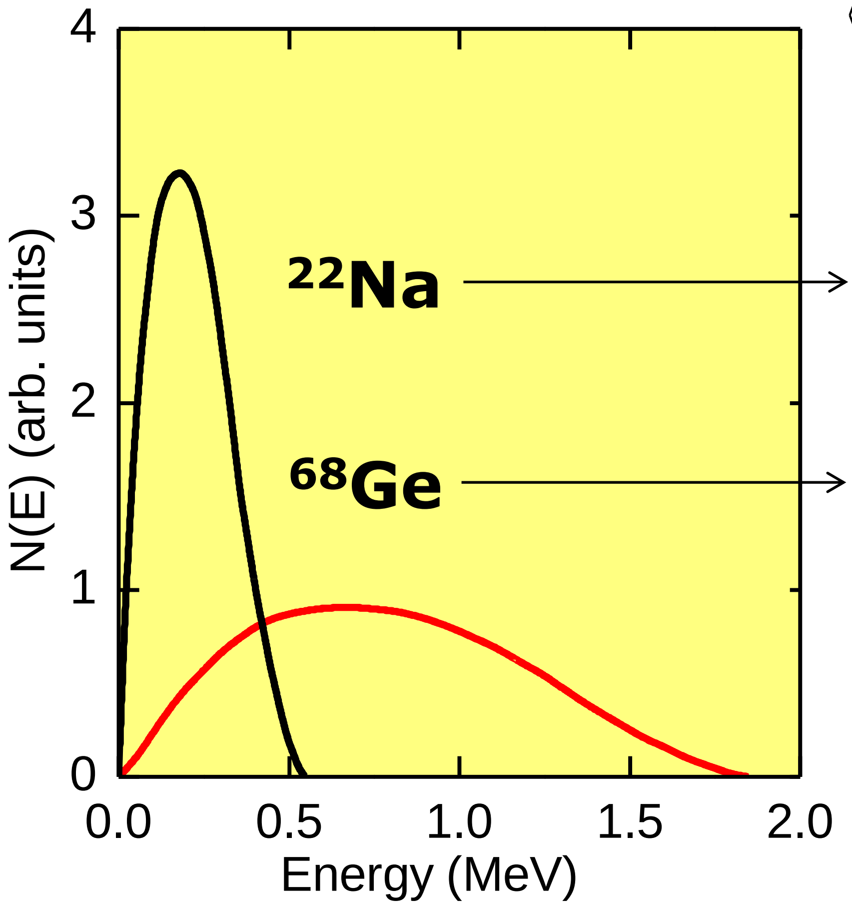

$$P_+ = \frac{v}{c}\frac{1+\cos\theta}{2}$$

(θ averaged)

Mean polarization for an energy window $E_1$ to $E_2$

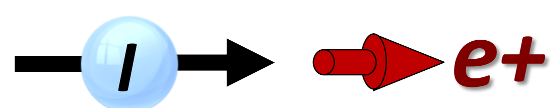
$$\langle P_+ \rangle = \int_{E_1}^{E_2} \sqrt{1 - \frac{1}{[1 + E/(mc^2)]^2}}\, N(E)\,dE\, \frac{1+\cos\theta}{2}$$

N(E) : $\beta^+$ energy distribution

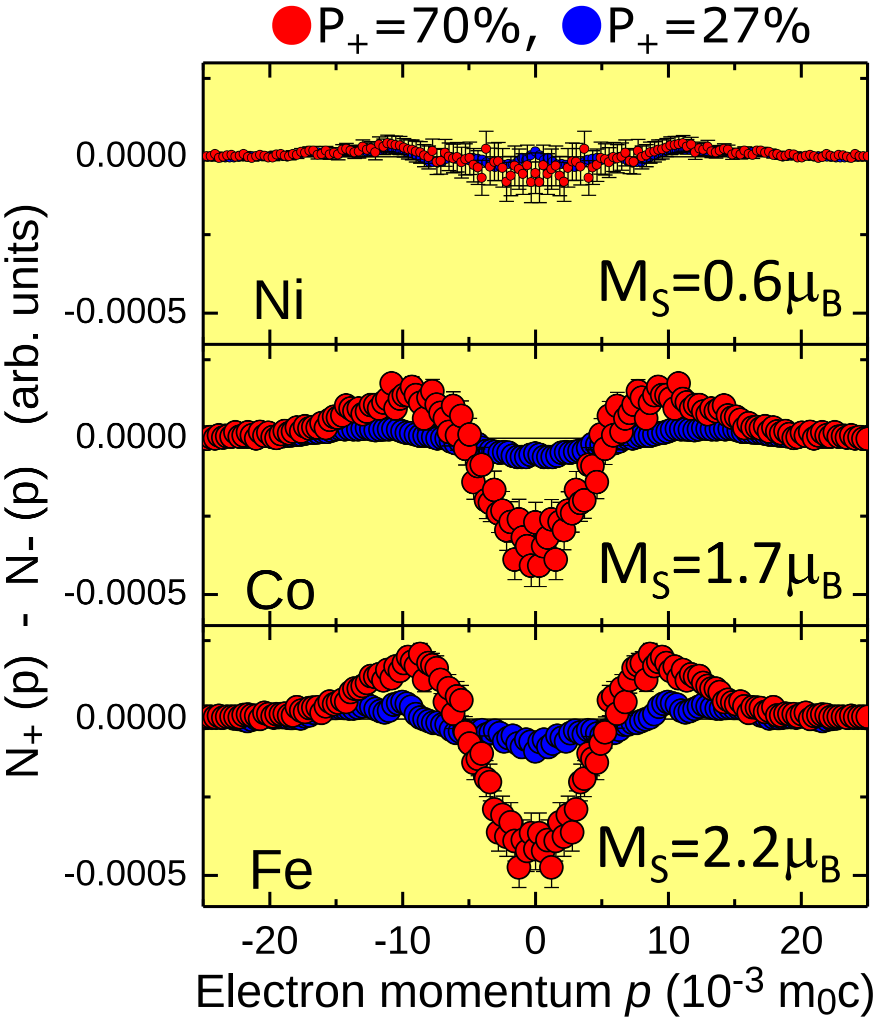


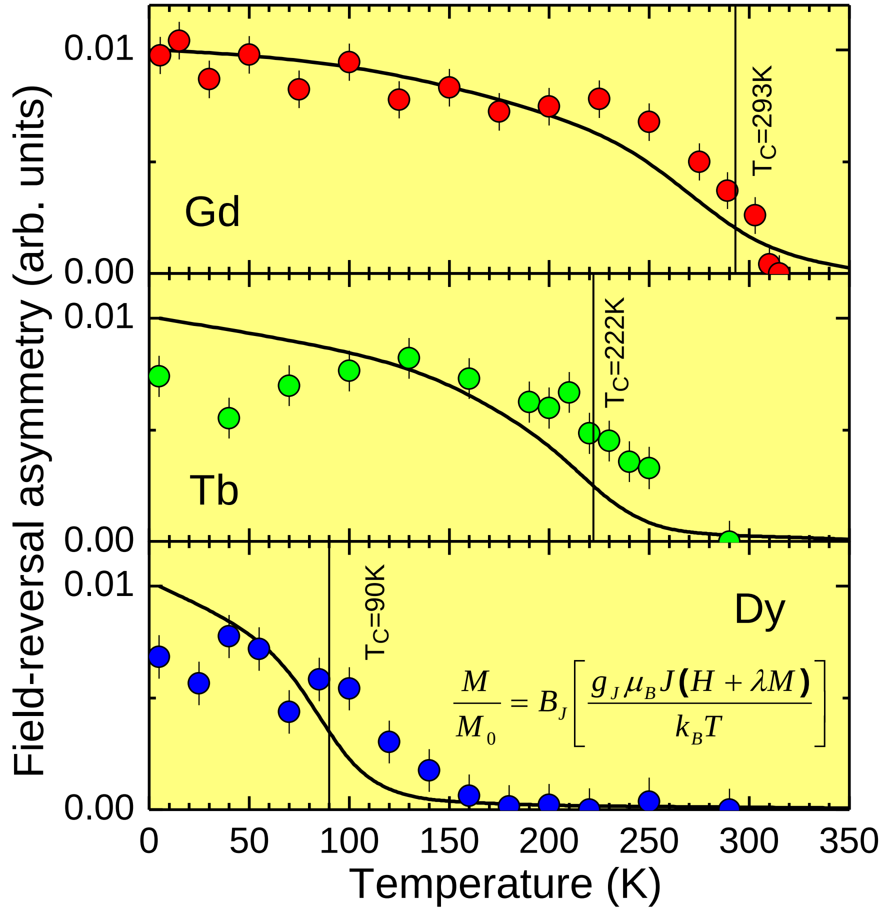

| Energy averaging | | 2π averaging |
|---|---|---|
| $\langle P_+ \rangle$=70% | ⇨ | 35% |
| $\langle P_+ \rangle$=94% | ⇨ | 47% |

**Higher energy endpoint**
**Higher energy $e^+$ in a direction**
**Align nuclear spins**



# Higher $e^+$ polarization gives better results



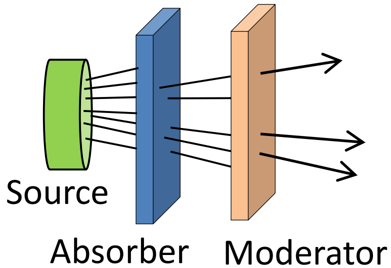

$$\frac{M}{M_0} = B_J\left[\frac{g_J \mu_B J(H + \lambda M)}{k_B T}\right]$$

PRB83(2011)100406(R).
PRB85（2012）024417.

# Expected Beam Polarization

- **Energy selection effect**

$$P_+(E) = \sqrt{1 - \frac{1}{[1+E/(mc^2)]^2}}\,\frac{1+\cos\theta}{2}$$




**1. Energy distribution after transmitting source with $d_S$ thick**

$$N_S(E) = \frac{1}{2}\int_0^{d_S} N_0(E)[A(z)/A_0]T_S(E,z)dz$$

Intrinsic distribution — Activity distribution — Transmittance

**2. Energy distribution after absorber with $d_A$ thick**

$$N_A(E) = N_S(E)T_A(E,d_A)$$

**3. Energy distribution after moderator with $d_M$ thick**

$$N(E) = N_S(E)T_A(E,d_A)\varepsilon_M(E,d_M)$$

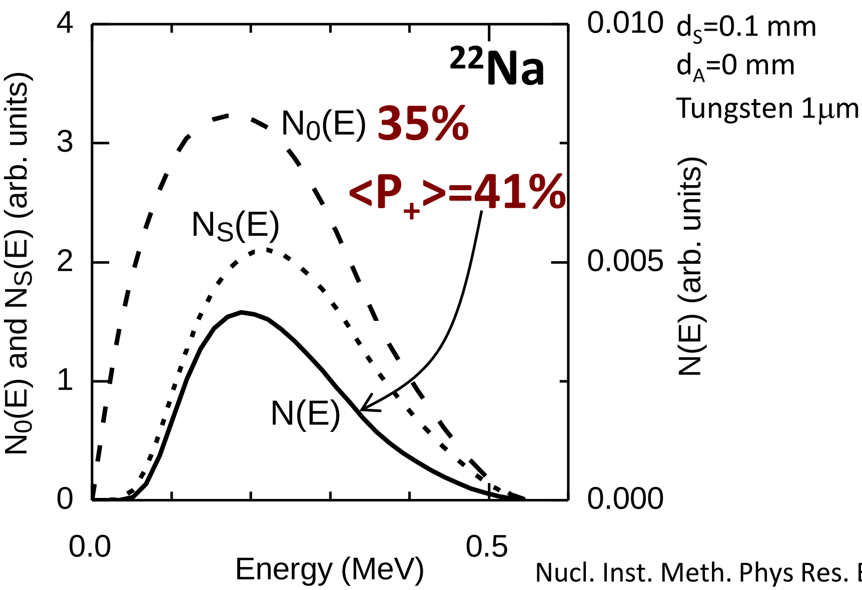
$$T(E,z) = \exp\left[-(z/z_0)^m\right]$$

$$z_0 = aE^n/\left[\rho\Gamma(1+1/m)\right]$$

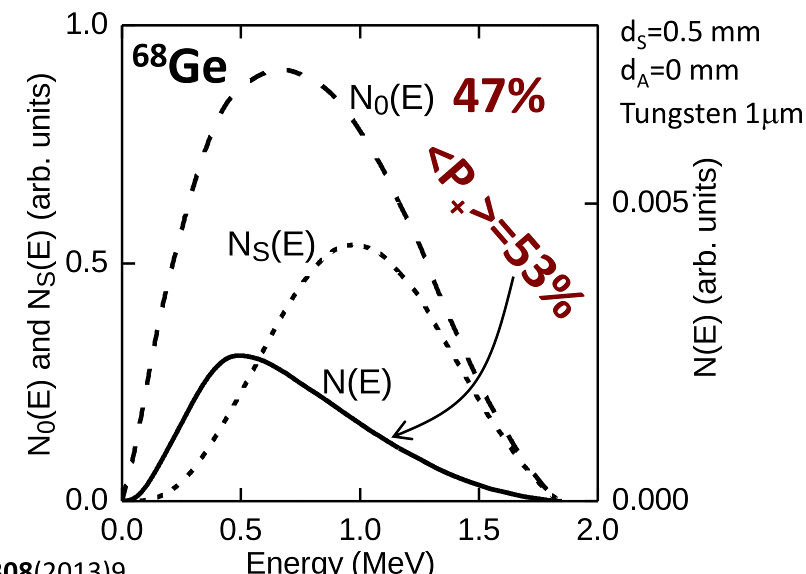
m, n: material parameter

A: 4 μg cm$^{-2}$keV$^{-n}$

Transmission $\varepsilon_M(E,d_M) = P_{em}\int_0^{d_M} p(E,z)\sinh(z/L)\sinh(d_M/L)dz$  Reflection $P_{em}\int_0^{d_M} p(E,z)\exp(-z/L)dz$

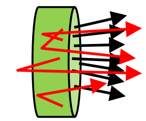



Nucl. Inst. Meth. Phys Res. B**308**(2013)9.



# Expected Beam Polarization (continued)

## ● Depolarization due to back-reflected positrons



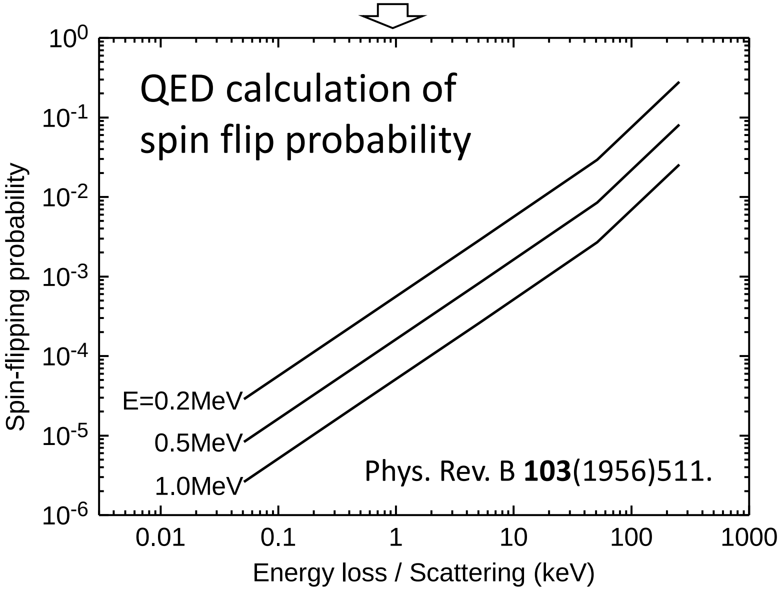
$$R = 0.342 \log Z - 0.146$$ Phys. Rev. A**7**(1973)135.

Positrons with all possible $\theta$ and E are back-reflected with this probability ⇨ $\Delta P_+ = 1 - R \sim 90\%$ for Carbon substrate

## ● Depolarization due to spin flip in moderator

| Inelastic processes | | Elastic processes |
|---|---|---|
| **Electromagnetic scattering (Bhabha, Bremsstrahlung)** | **~MeV** | **Mott scattering** |
| Dielectric scattering (electron-hole, Plasmon excitation) | ~keV | |
| Phonon scattering | ~eV | |

Stopping Power : **Bhabha** >> Bremsstralung (EZ/800)




Mean energy from $^{22}$Na ~0.2MeV

Energy transfer in W per collision ~500eV ⇨ $\Delta P_+ \sim 95\%$

→ ~400 collisions until zero energy

Calculated for electron

$$\Delta P = 1 - \left( \frac{1 - \exp(-\gamma d_M)}{\gamma d_M} \right)^2$$

$$\gamma = 4\sqrt{2}\pi N \left( \frac{e^2}{E + 2mc^2} \right) \left( \ln(2kb) - 0.577 + 2\xi S_1 + \xi^2 S_2 \right) \qquad b = \frac{\hbar}{me^2} Z^{-1/3}$$

$$\xi = \frac{e^2 Z}{\hbar c} \frac{E + mc^2}{E^{1/2}(E + mc^2)^{1/2}} \qquad S_1 = -\frac{\pi}{2} \sum_{l=1}^{\infty} \frac{H_1^{(1)}[i(l+1/2)/(kb)]}{kb(l+1/2)} \tan^{-1}[-(l+1/2)k/k_0]$$

$$S_2 = \sum_{l=1}^{\infty} [\tan^{-1}\{-(l+1/2)k/k_0\}]^2 (l+1/2)^{-3} \qquad k_0 = e^2 Z(E + mc^2)/(\hbar c)^2$$

Phys. Rev. **55**(1938)277.

No difference in charge in Coulomb scattering,

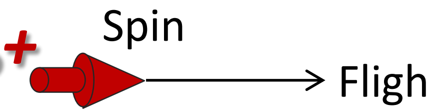
For $^{22}$Na ~ 0.2MeV and 1 $\mu$m W moderator,

⇨ $\Delta P_+ \sim 95\%$

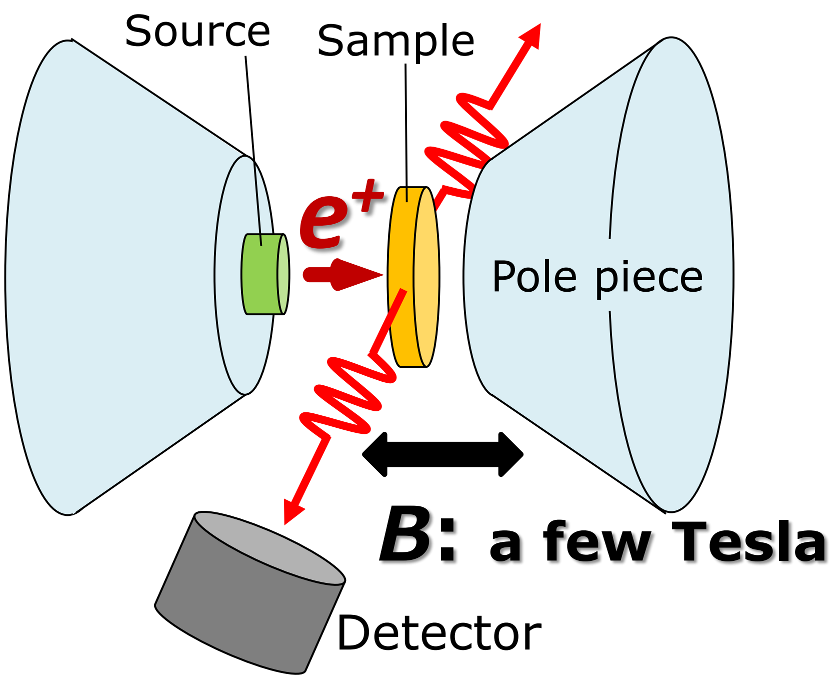
**Final beam polarization**

**$P_+ > 33\%$** $^{22}$Na **$P_+ > 45\%$** $^{68}$Ge

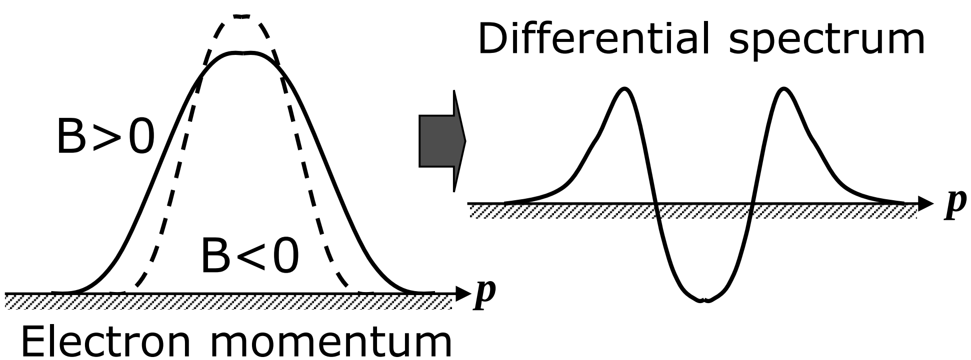
# Longitudinal spin polarization: $e^+$




## Non-moderated positron beam ($^{68}$Ge-$^{68}$Ga)

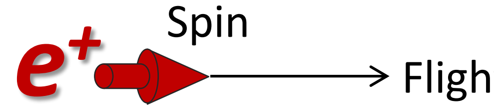



**Flux ~$10^6$ e$^+$/sec**
**$^{68}$Ge-$^{68}$Ga P~70%**

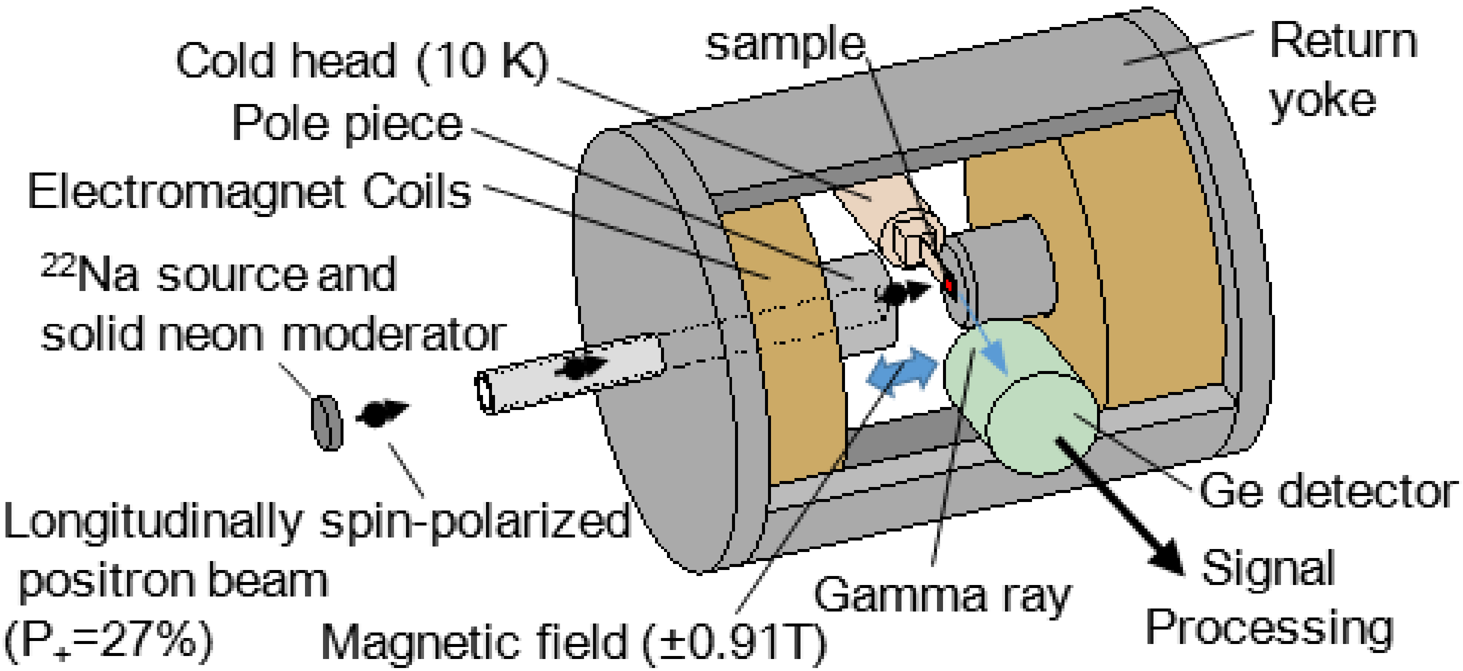


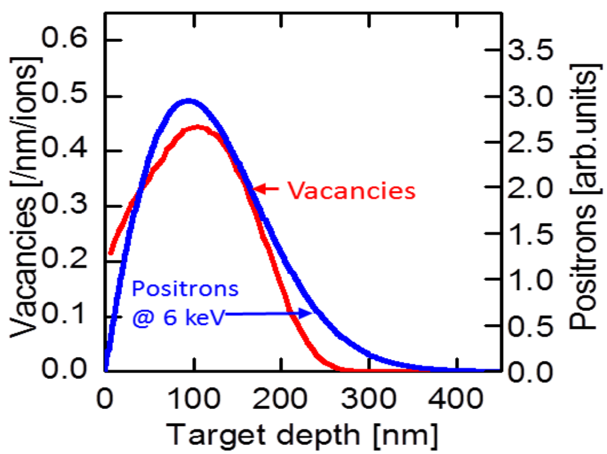

$$N^{maj}(p) - N^{\min}(p)$$

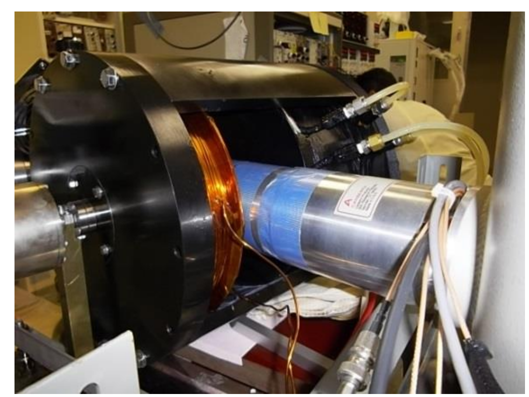
# Longitudinal spin polarization:




## Magnetically transported positron beam ($^{22}$Na)

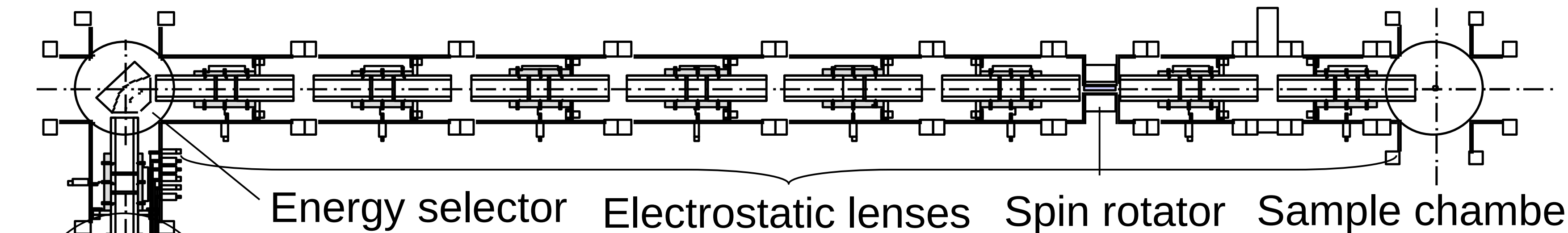


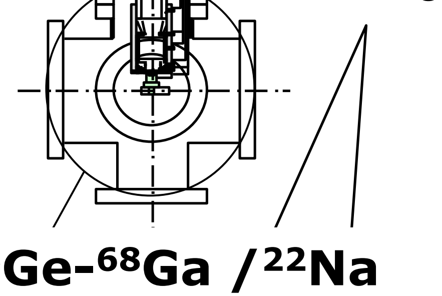



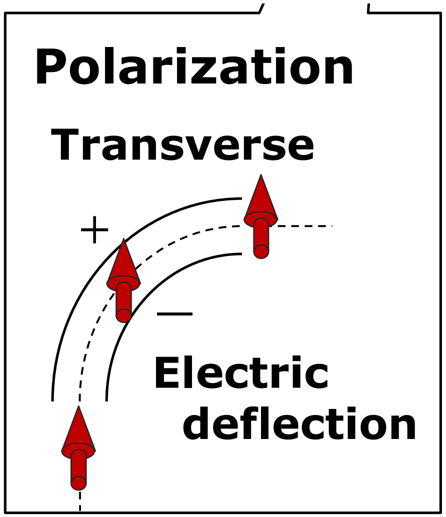



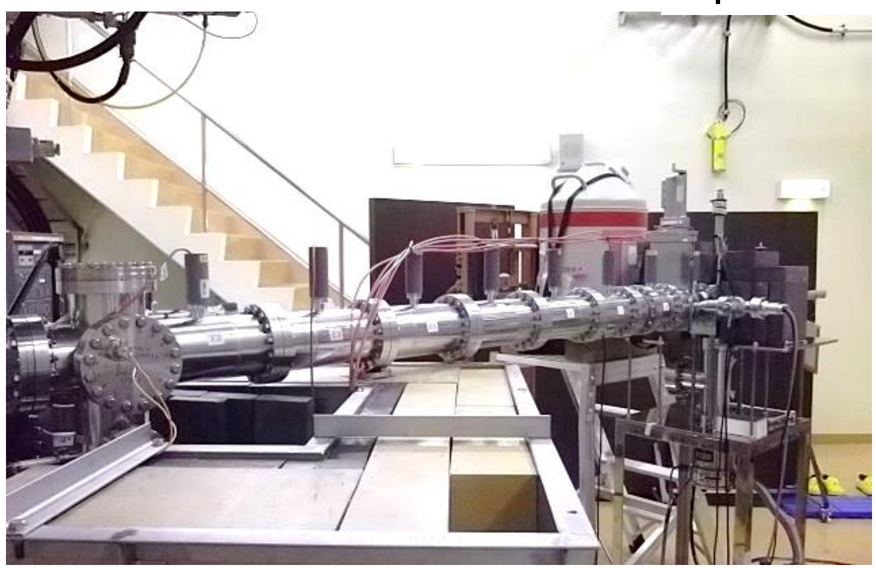
# Transverse spin polarization:

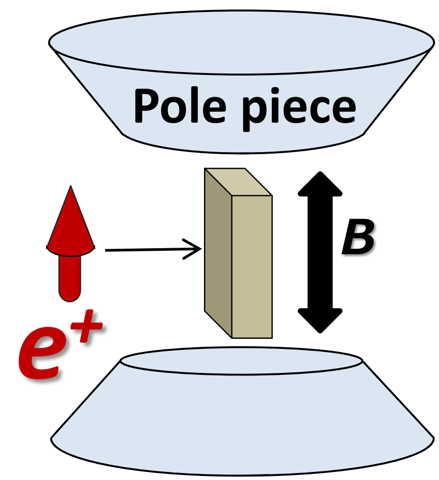

## Electrostatic positron beam #1 ($^{22}$Na, $^{68}$Ge-$^{68}$Ga)






**Flux ~$10^4$ $e^+$/sec**

**$^{22}$Na P=30%**

**$^{68}$Ge-$^{68}$Ga P=47%**

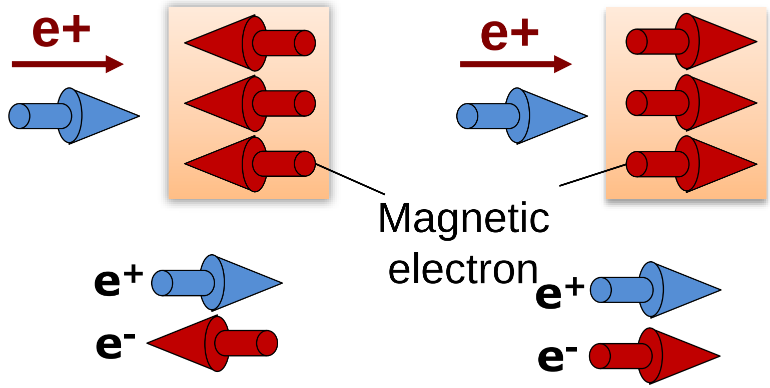
# Contents:

1. Introduction to Positron Spectroscopy
   - ✓ General aspects
   - ✓ Spin-Polarized Positron Spectroscopy

2. **Applications of Polarized Positron Spectroscopy**
   - ✓ **Classical to Heusler ferromagnets**
   - ✓ Vacancy-induced ferromagnetism
   - ✓ Surface spin polarization

3. Summary & Future Prospects
   - ✓ Further Research & Development

# Spin-polarized positron spectroscopy




Annihilation rate

**High** **Low**

Differential spectrum in ±B field

$$N_+(p_z) - N_-(p_z)$$

$$= \sum_{i=1}^{occ.} \left[ \frac{(1+P_+)N_i^{maj}(p_z)}{\lambda^{\Uparrow}} - \frac{(1-P_+)N_i^{\min}(p_z)}{\lambda^{\Downarrow}} \right]$$

Up and Down Spin positron annihilation rate

≠ **Difference between Majority and Minority spin bands**

Differential spectrum in majority and minority spin bands

$$N^{maj}(p_z) - N^{\min}(p_z) = \sum_{i=1}^{occ.} \left[ N_i^{maj}(p_z) - N_i^{\min}(p_z) \right]$$

Correction term

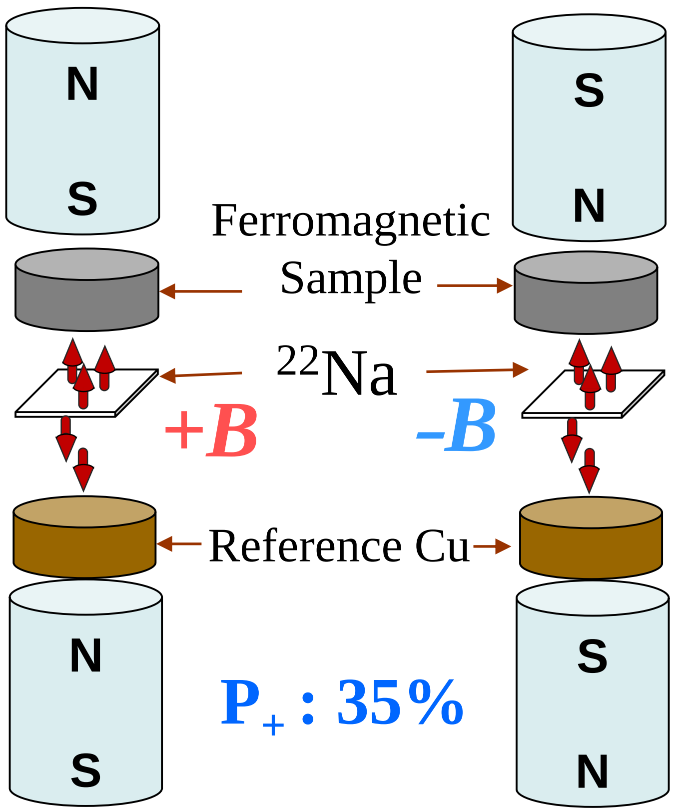
$$= [N_+(p_z) - N_-(p_z)] + P_+ \frac{\lambda^{\Uparrow} - \lambda^{\Downarrow}}{\lambda^{\Uparrow} + \lambda^{\Downarrow}} [N_+(p_z) + N_-(p_z)]$$

$$\lambda^{\Uparrow(\Downarrow)} = \lambda_{maj\,(\min)} + (\lambda_{maj\,(\min)} + 2\lambda_{\min(maj)})/1115 \qquad \lambda_{\pm} = (1 \pm P_+)\lambda_{maj}/2 + (1 \mp P_+)\lambda_{\min}/2$$

"Renormalization", ever pursued by Berko and Mills via 3γ observation

J. de Phys. 32(1971)C1-287.

# Spin-polarized positron spectroscopy



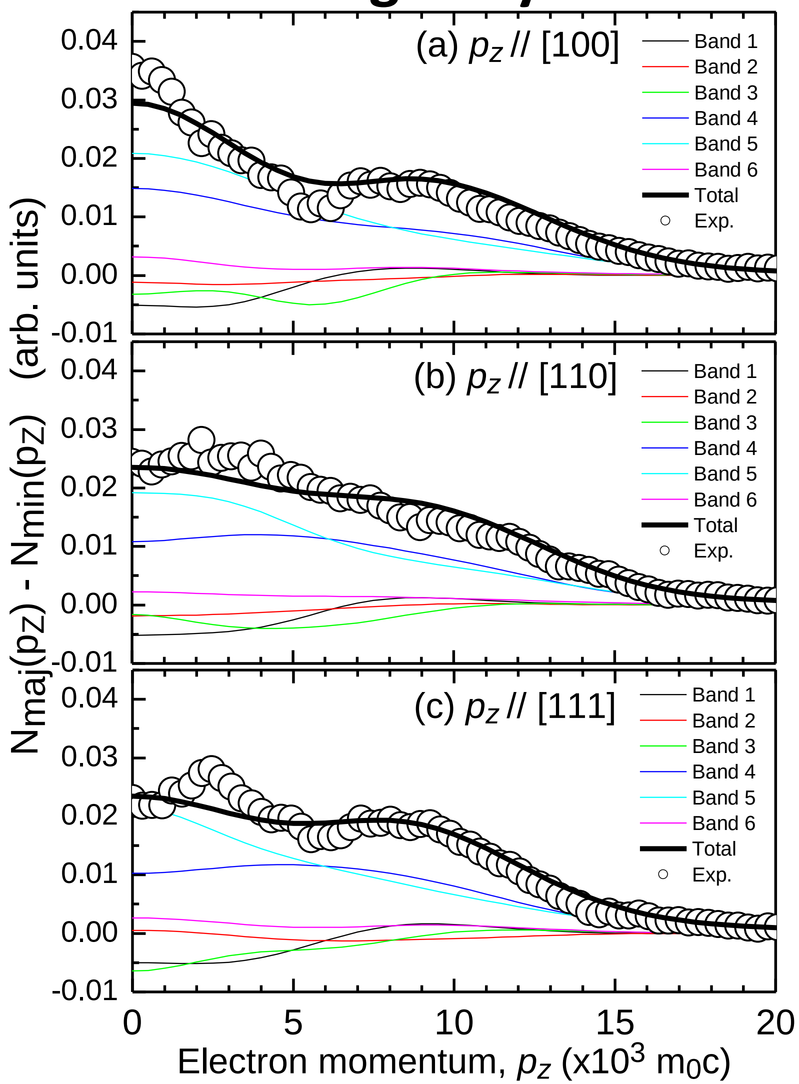

| | $\lambda^{\uparrow}$ (ns$^{-1}$) | $\lambda^{\downarrow}$ (ns$^{-1}$) |
|---|---|---|
| Fe | 9.35<br>10.6 | 8.47<br>10.1 |
| Co | 8.91<br>10.8 | 8.48<br>10.2 |
| Ni | 9.71<br>10.6 | 9.62<br>10.4 |
| Gd | 6.49<br>7.1 | 5.29<br>4.8 |
| $Co_2MnSi$ | 6.80<br>10.5 | 6.58<br>9.8 |
| $Co_2MnAl$ | 6.76<br>10.0 | 6.62<br>9.5 |
| NiMnSb | 5.88<br>7.0 | 5.53<br>6.2 |

Red: Calculation with BN enhancement factor

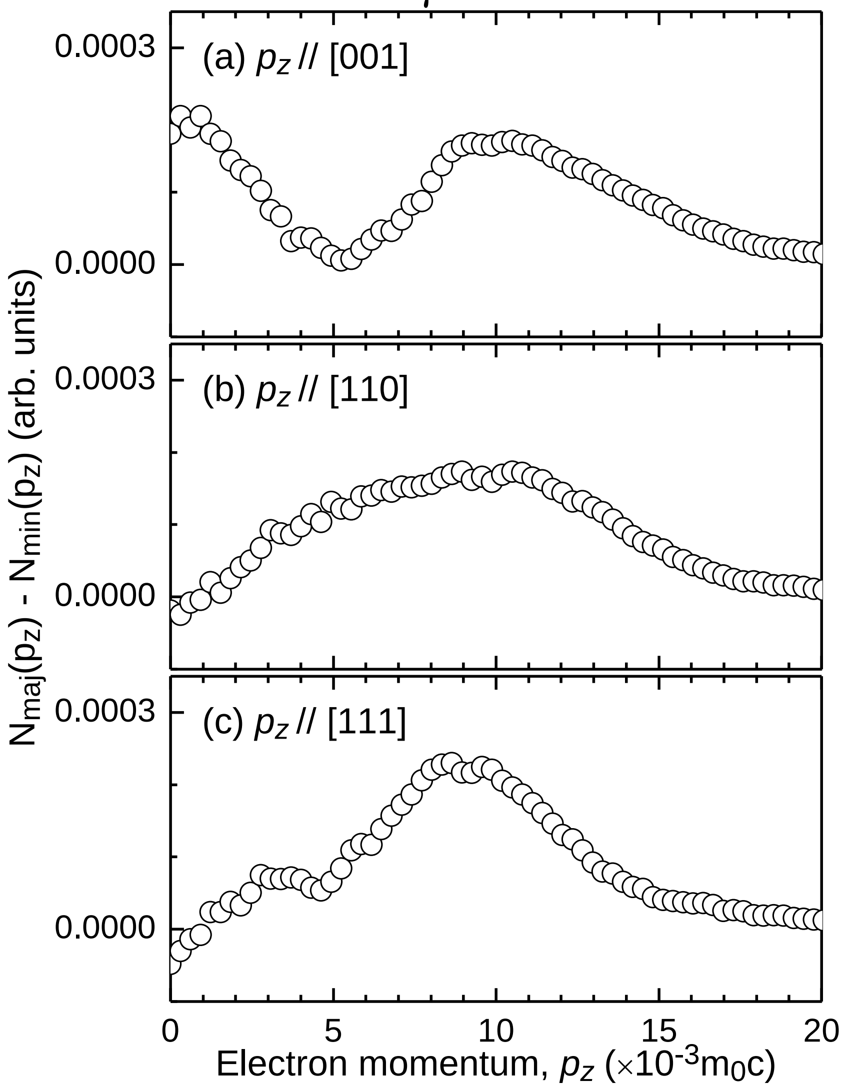
JPCM27(2015)246001.
Defect and Diffusion Forum, 373(2017)65-70.

# Spin-polarized positron spectroscopy

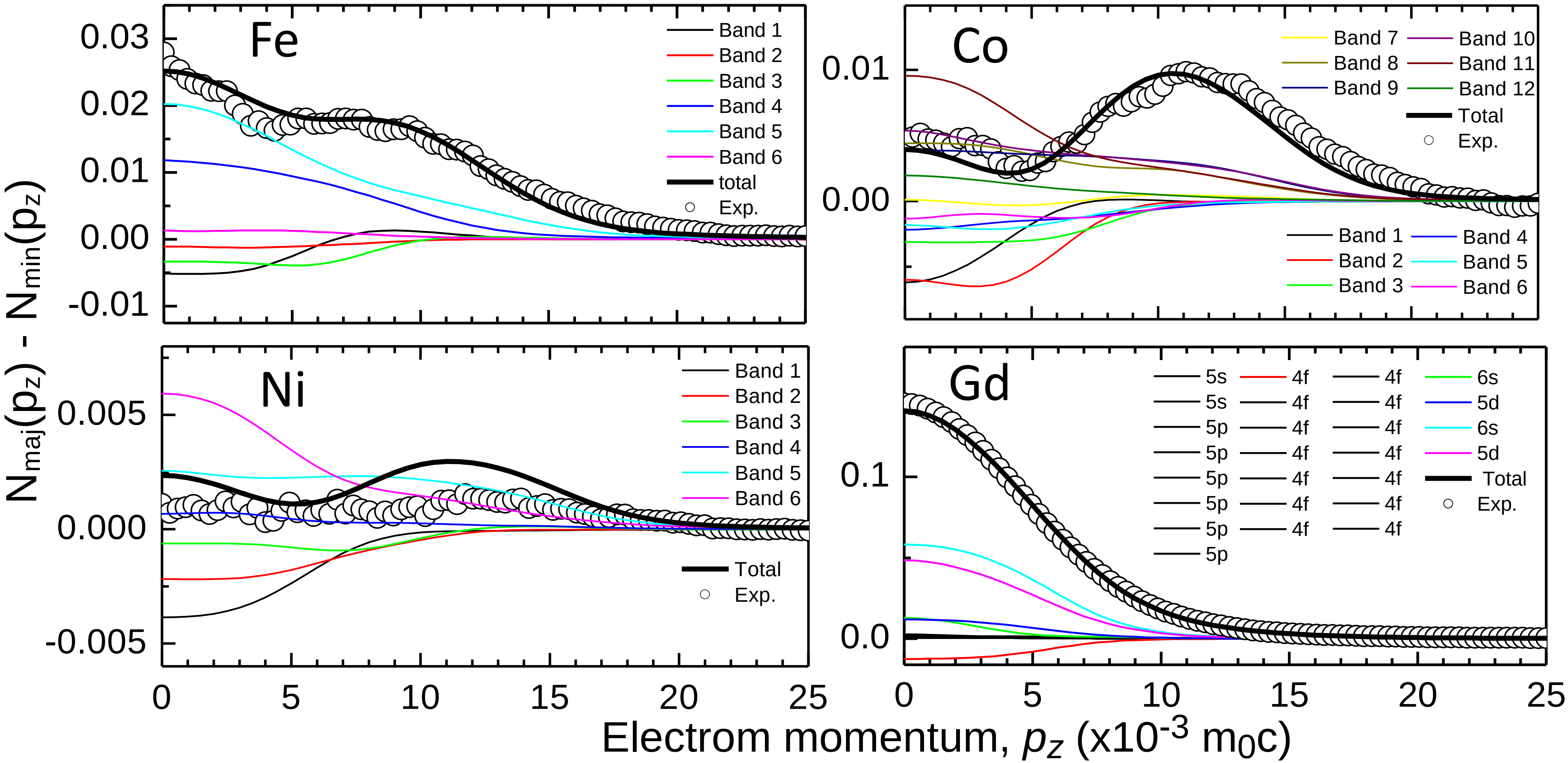
## Fe single crystal




## If BM's 3γ renormalization



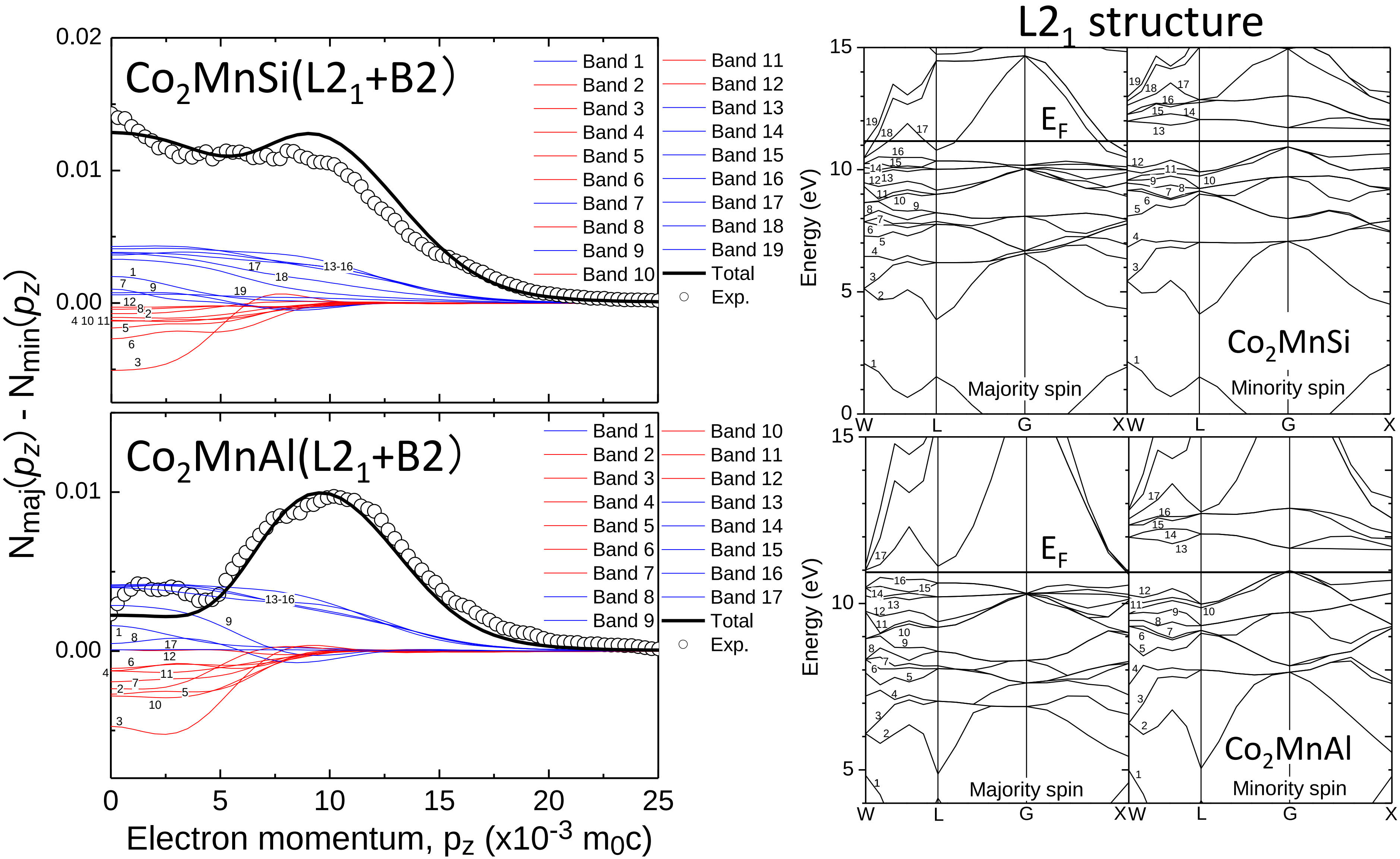
# Spin-polarized positron spectroscopy




**Agreement in experiment and calculation (ABINIT)**

➡ **Adequate precision of calculation**

PRB83(2011)100406(R). PRB85（2012）024417.
Defect and Diffusion Forum, 373(2017)65-70.




**Agreement in EXP. and CALC.**

- **Higher half-metallicity of $Co_2MnSi$ than $Co_2MnAl$**
- **Robust half-metallicity upon L2$_1$ to B2 disordering**

# Contents:

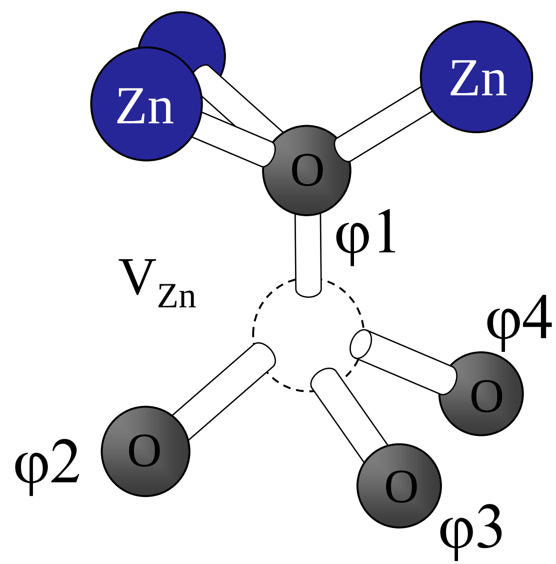
# Magnetic Semiconductor "Semiconductor spintronics"

*Carrier-mediated ferromagnetism, controlled by light and electric filed.*

**1990～**
- Synthesis of InMnAs and GaMnAs
- Many studies by doping of magnetic elements

**2004～**
- 「$d^0$」 ferromagnetism in Oxide, Nitride, Carbide
- Vacancies are proposed as the source of ferromagnetism

After Venkatesan et al. Nature430(2004)630.



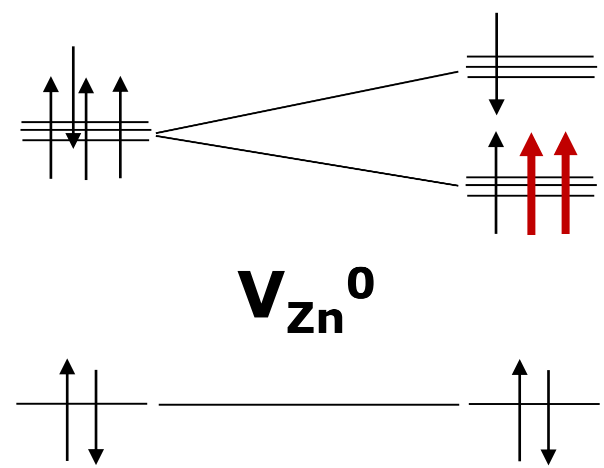

T2 orbital

$$\phi_1 + \phi_2 - \phi_3 - \phi_4$$
$$\phi_1 - \phi_2 + \phi_3 - \phi_4$$
$$\phi_1 - \phi_2 - \phi_3 + \phi_4$$

A1 orbital

$$\phi_1 + \phi_2 + \phi_3 + \phi_4$$

Exchange splitting



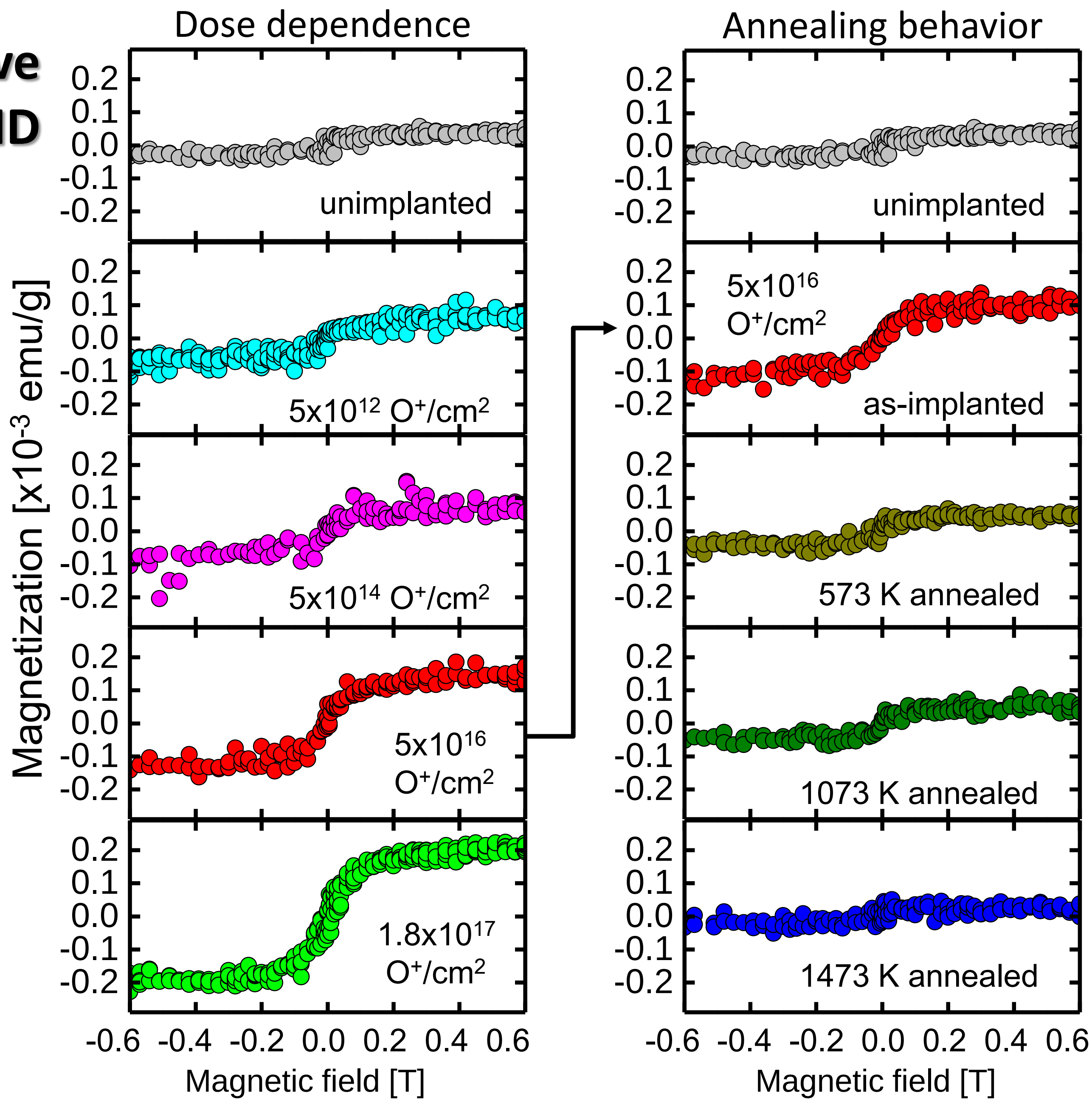
**$V_{Zn}{}^0$**

**S=1 → $2\mu_B$ per $V_{Zn}{}^0$**

**But, the direct evidences have not yet been found…**

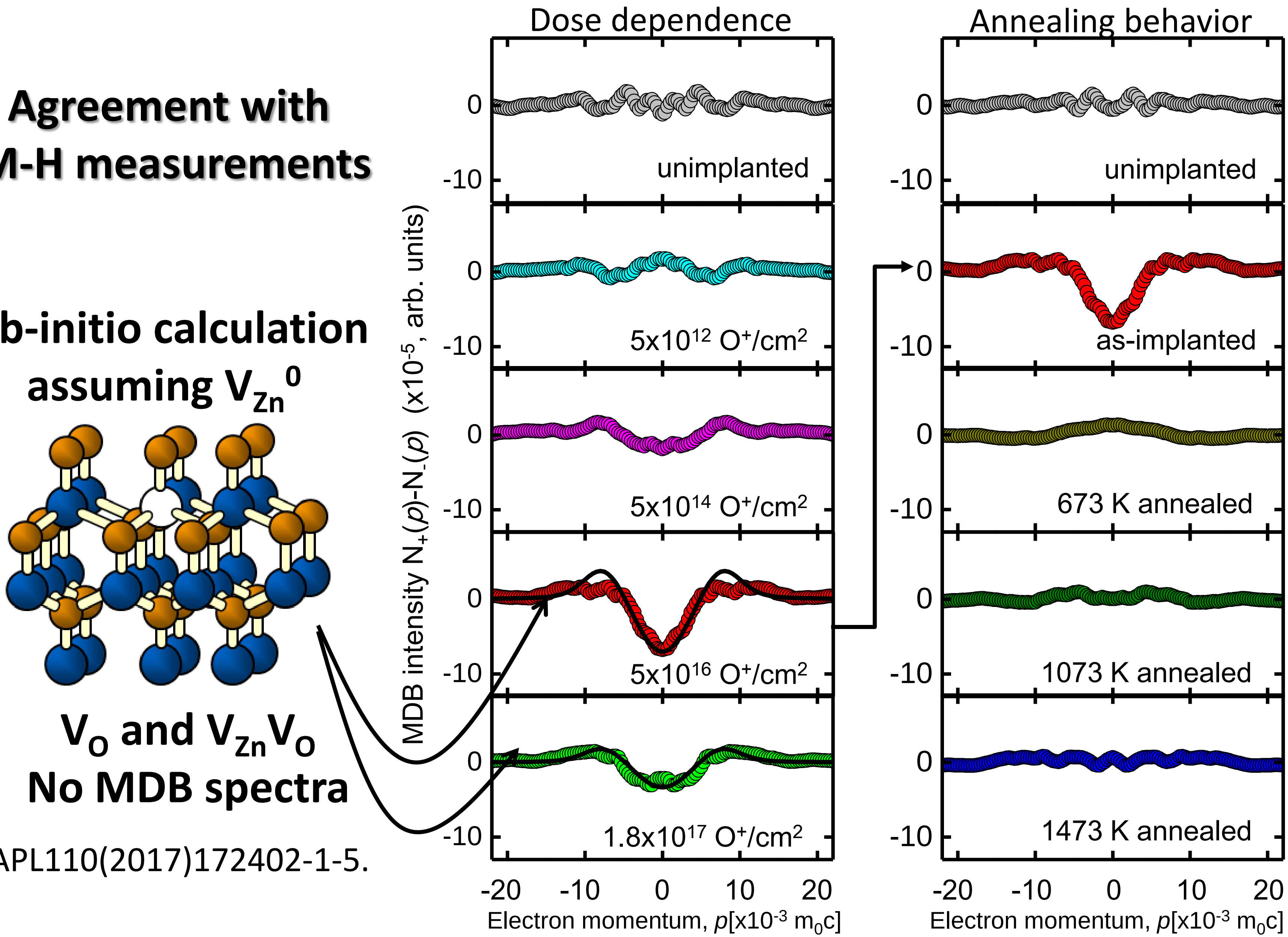
# M-H curve by SQUID

**100keV $O^+$ irradiated ZnO**



**Agreement with M-H measurements**

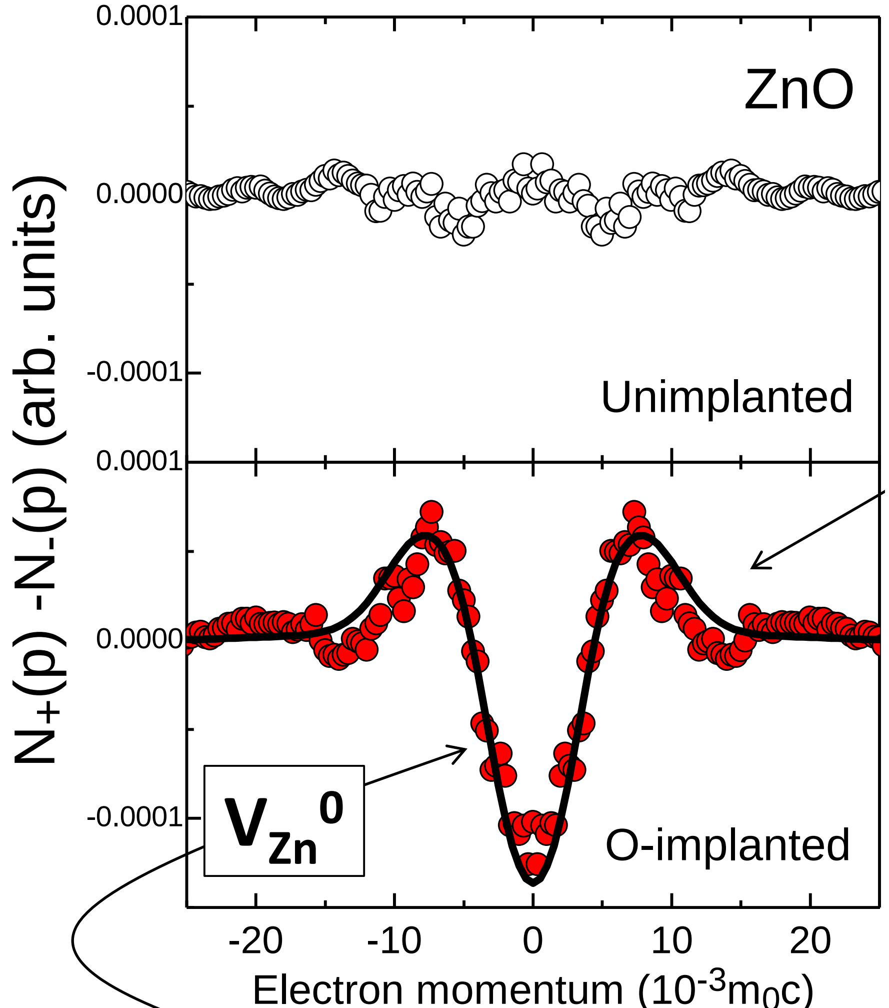
**Ab-initio calculation assuming $V_{Zn}^0$**

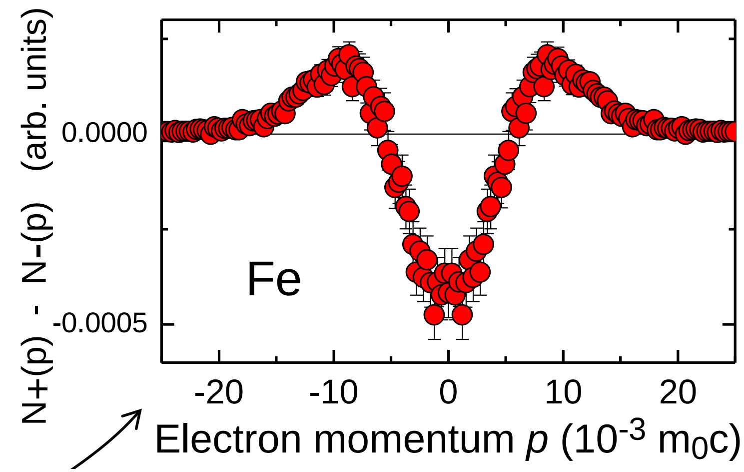



**$V_O$ and $V_{Zn}V_O$ No MDB spectra**

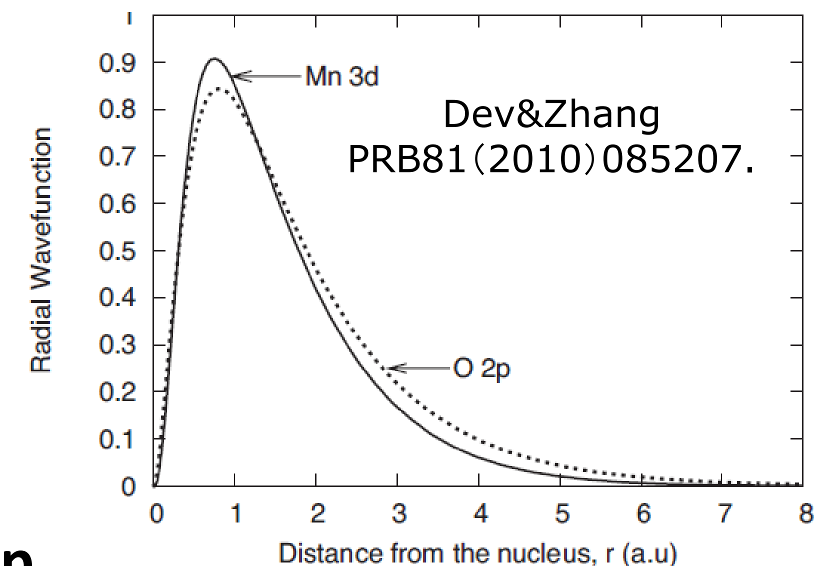
APL110(2017)172402-1-5.

# As for spectrum shape…



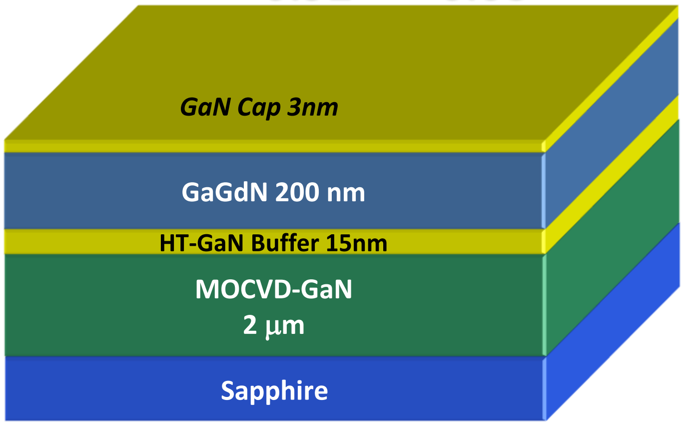

**Similar shape**



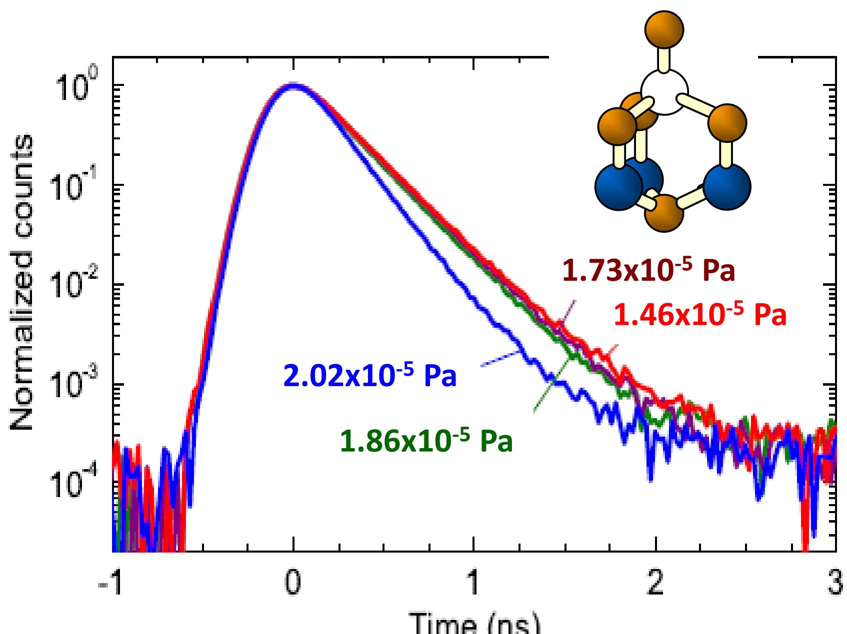

**Calculation reproduces exp.**

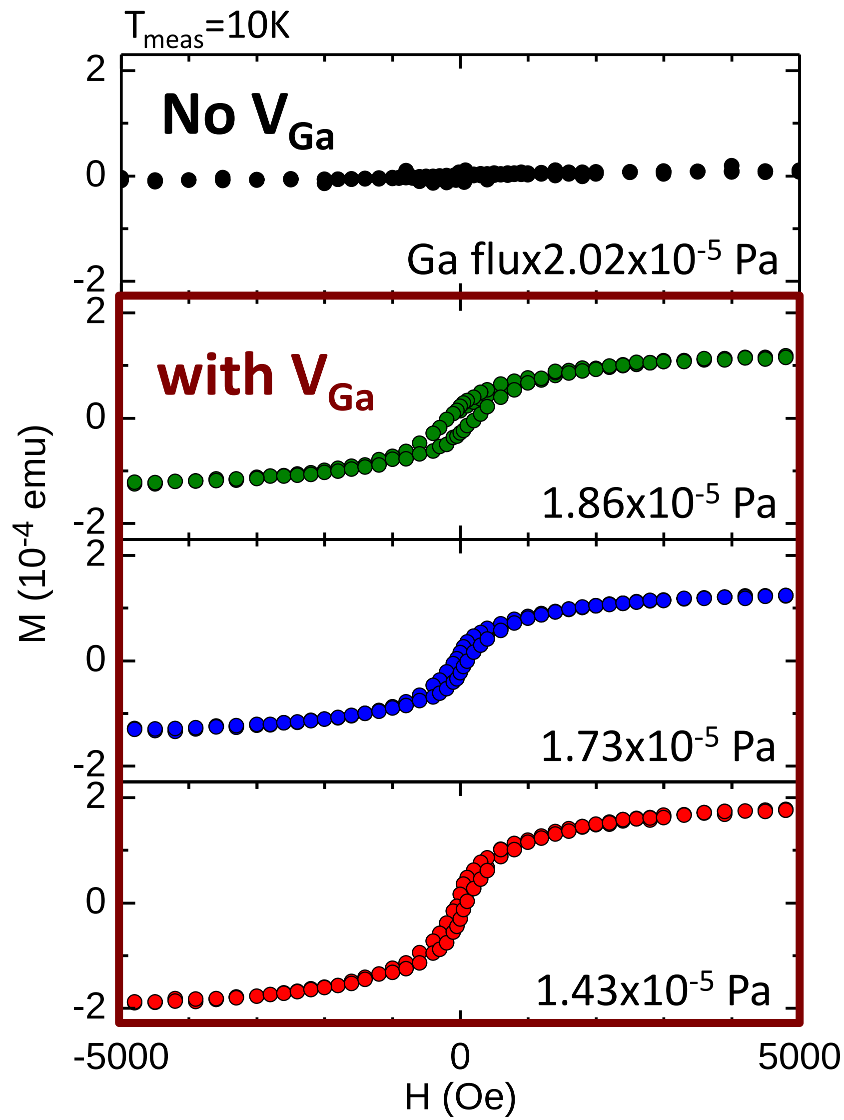
# Other on-going topics on Vacancy-induced FM

## $Ga_{0.92}Gd_{0.08}N$




**Ga flux change→ High density $V_{Ga}$**

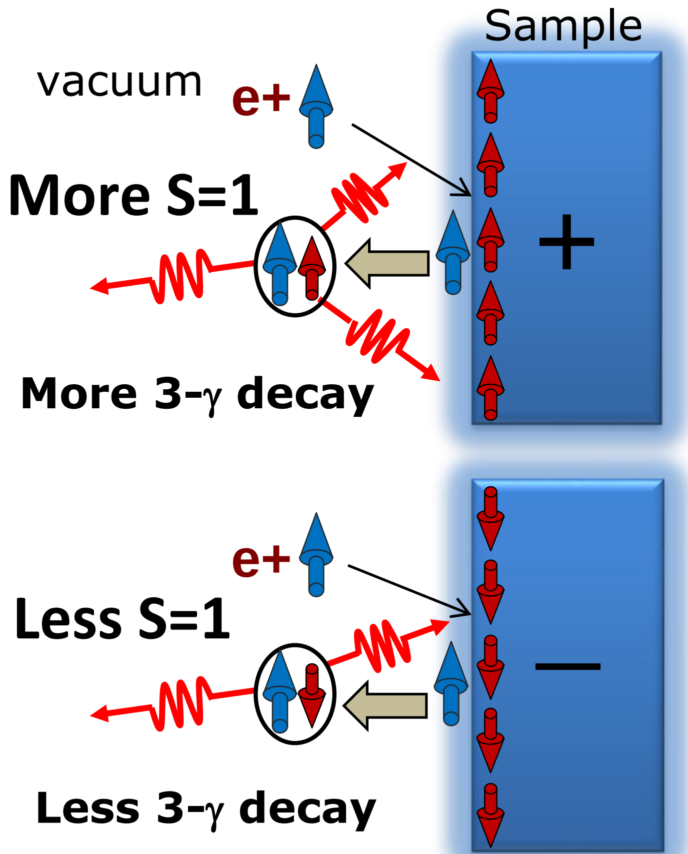


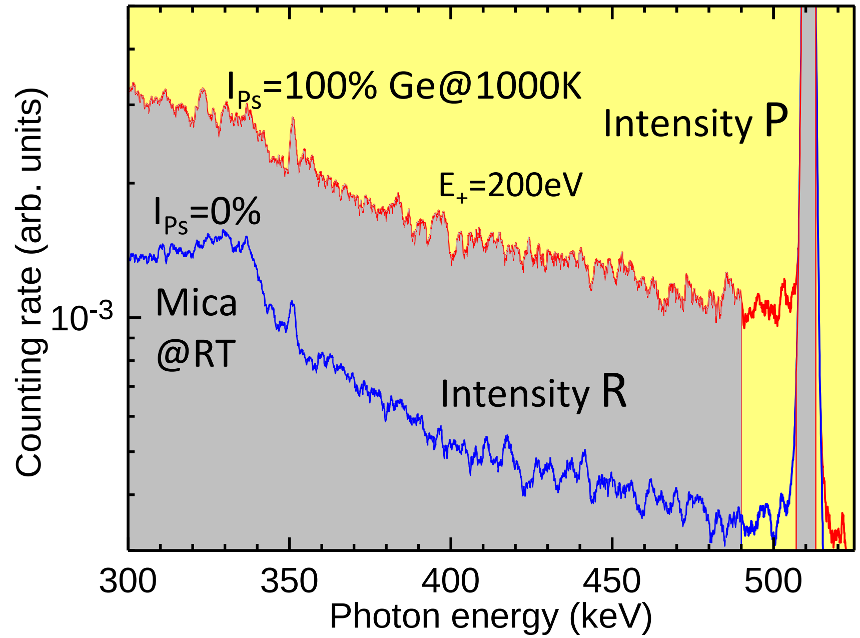




Yabuuchi et al. APL in press

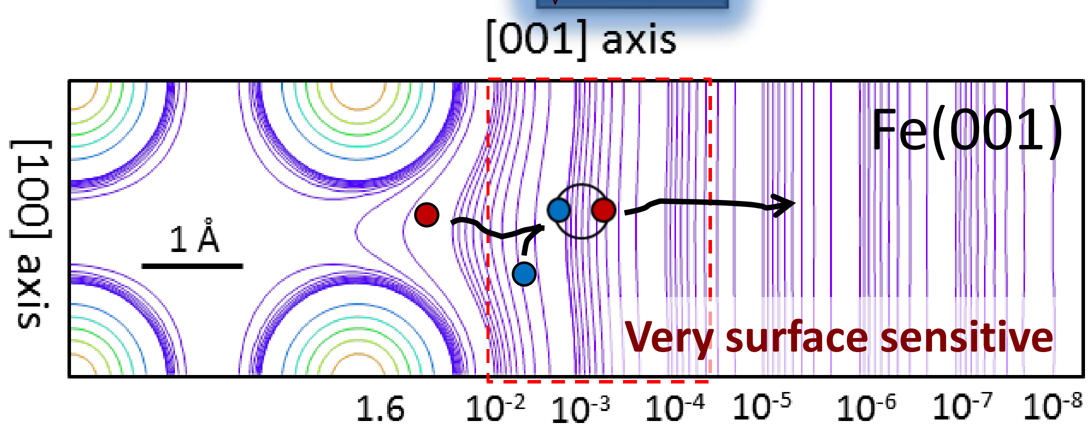
# Contents:

# Surface Spin-Polarization




$$I_{Ps}^{3\gamma} = \left[ 1 + \frac{P_{100\%}}{P_{0\%}} \frac{R_{100\%} - R}{R - R_{0\%}} \right]^{-1}$$




## Asymmetry of 3-gamma

$$\frac{I_{Ps}^{3\gamma}(+) - I_{Ps}^{3\gamma}(-)}{I_{Ps}^{3\gamma}(+) + I_{Ps}^{3\gamma}(-)} = \alpha P_+ P_-$$

# Surface Spin-Polarization

**Gidley et al. PRL49(1982)1779.**

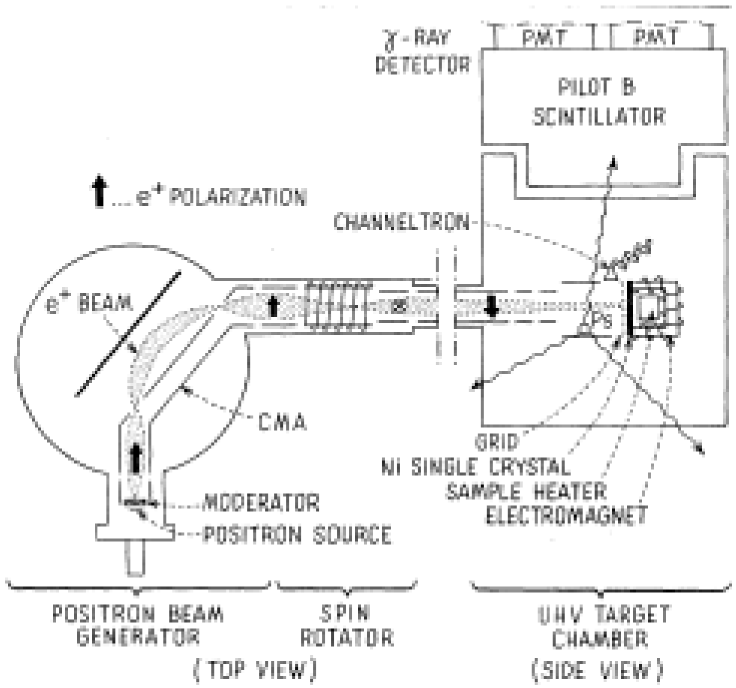
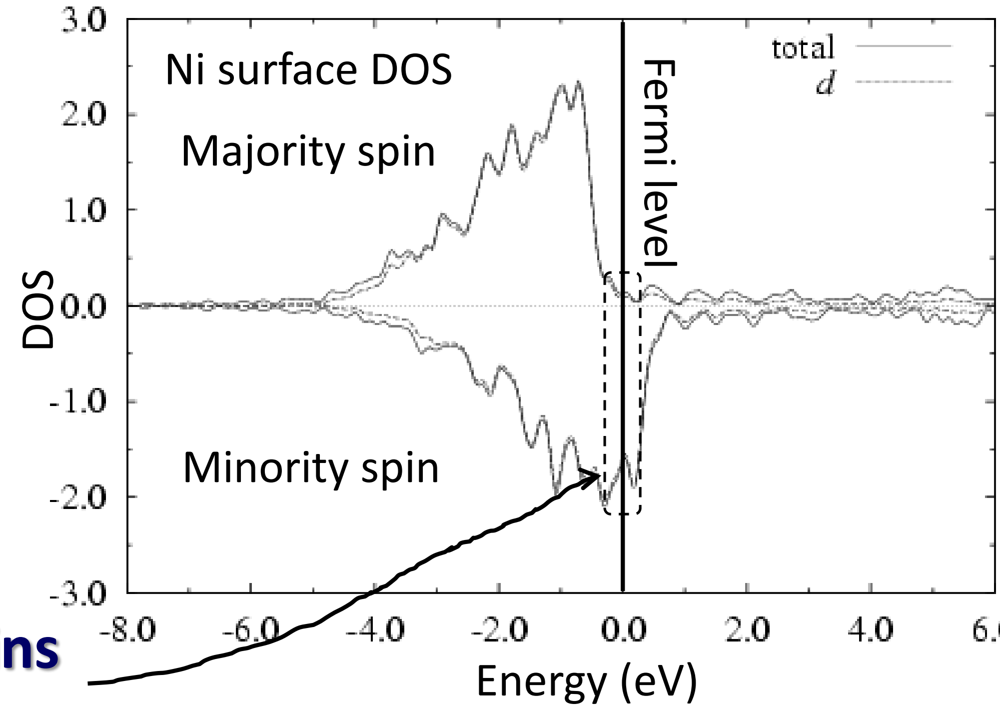



FIG. 1. The experimental arrangement. Details of the target chamber are rotated 90° for clarity.

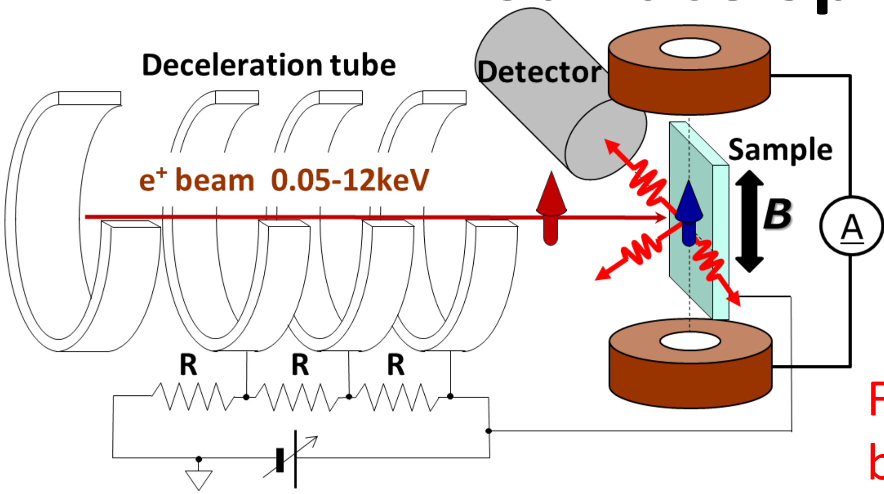
- **Ni surface spin polarization was determined to be –2.5%.**
- **Negative sign = Majority spins are detected.**




Park, et al. JKPS47(2005)655.

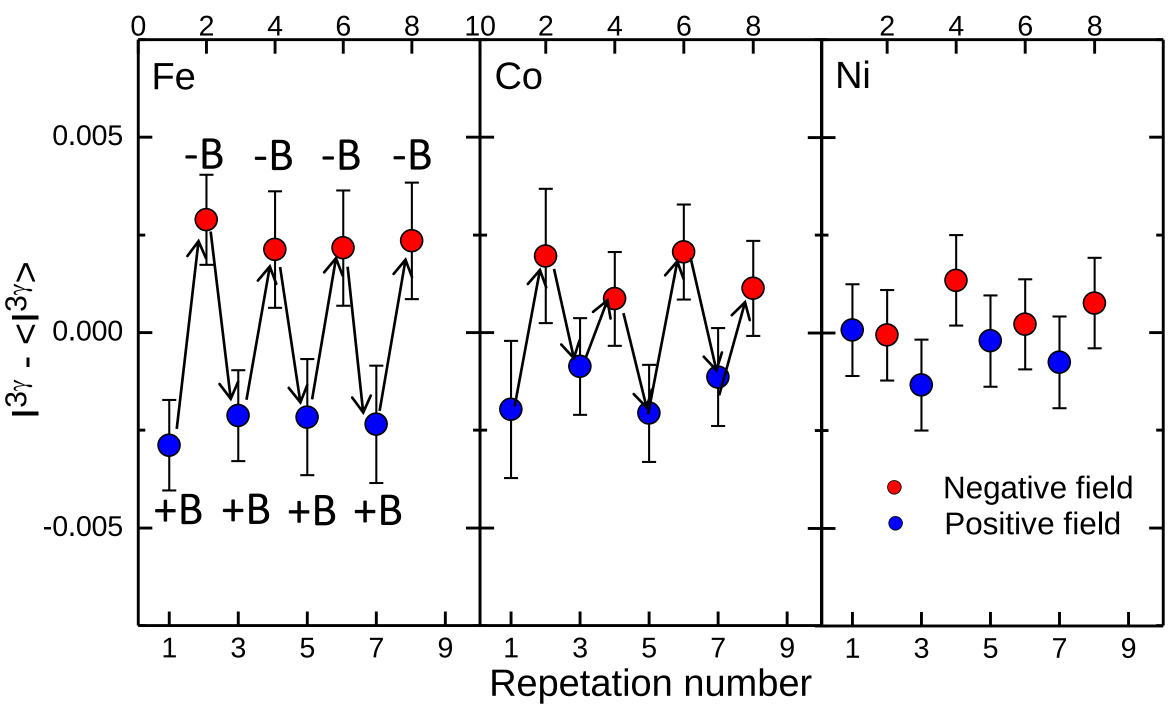
**At the Fermi level, minority spins are predominant and P=+85%.**

# Surface Spin-Polarization




**bcc Fe(001)/MgO(001)**
**fcc-Co(001)/MgO(001), hcp-Co(0001)/$Al_2O_3$**
**fcc-Ni(001)/MgO(001), Ni(111) /$Al_2O_3$**
**1.0kV $Ar^+$ sputtering＋800℃×1min.**
**Magnetic field 150~400 Gauss**

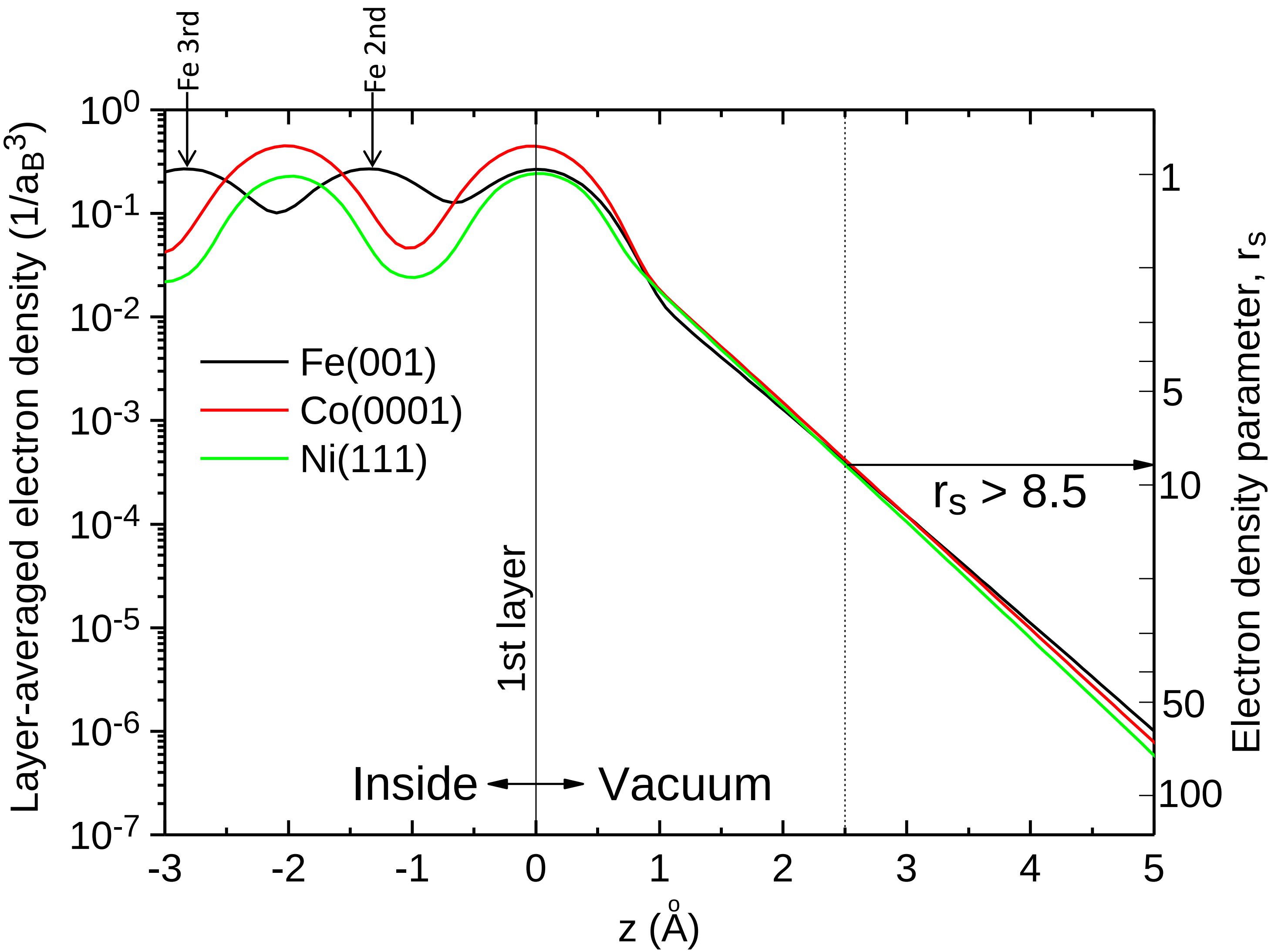
Fake polarization due to disturbance of beam by switching field direction is removed by rotating sample (explanation skipped)




| Fe | Co | Ni |
|---|---|---|
| (001)<br>–3.7% | (001)<br>–2.6% | (001)<br>– 0.4% |
| | (0001)<br>-2.9% | (111)<br>-1.4% |

Defect and Diffusion Forum, 373(2017)65-70.

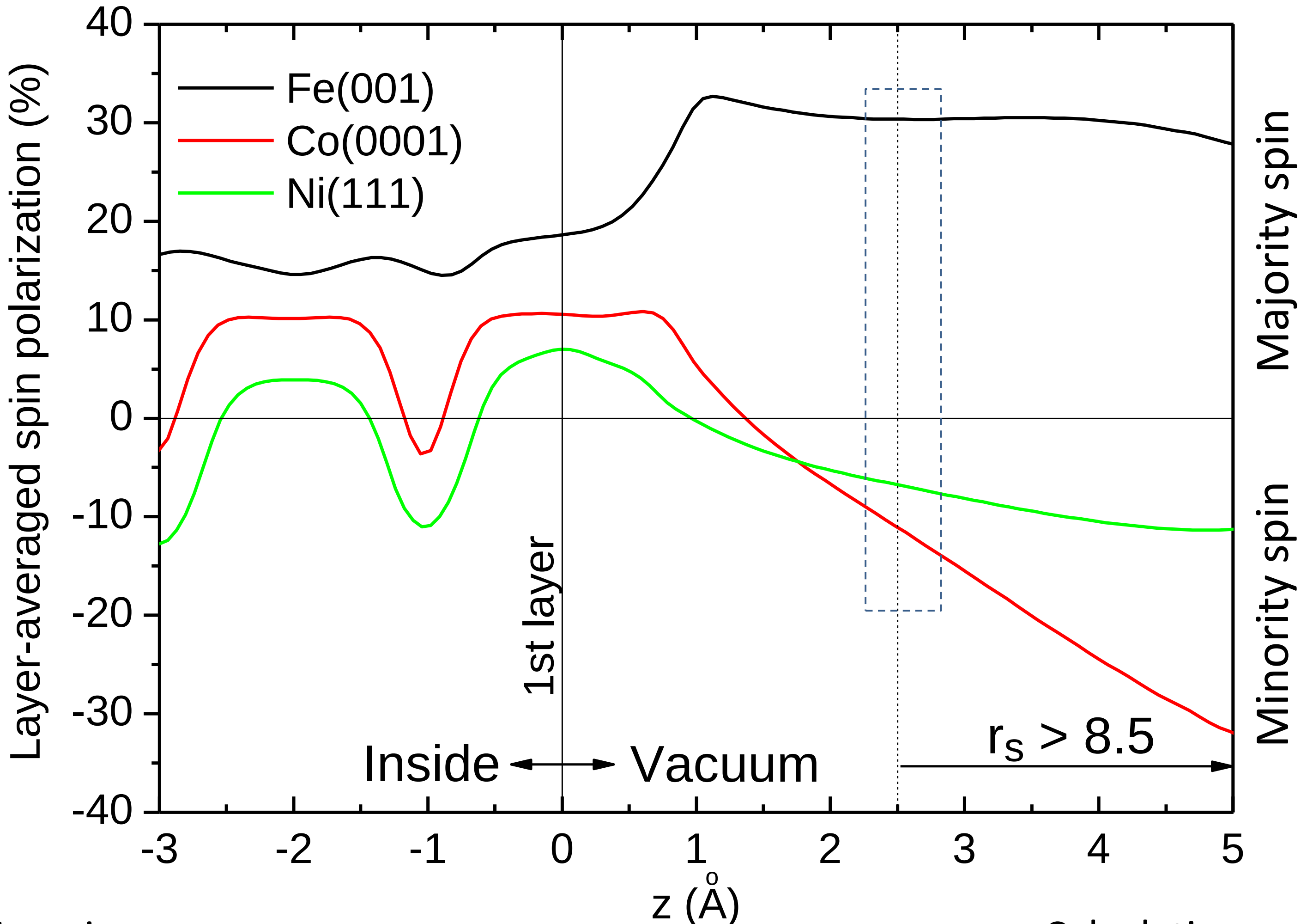
**Majority spins are detected**
**Rather small polarization**




**Ps may be formed in vacuum region. But, we do not know where of vacuum region……**

**In a static theory, Ps is possible only at low density limit, $r_s > 8.5$.**

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




Experiment

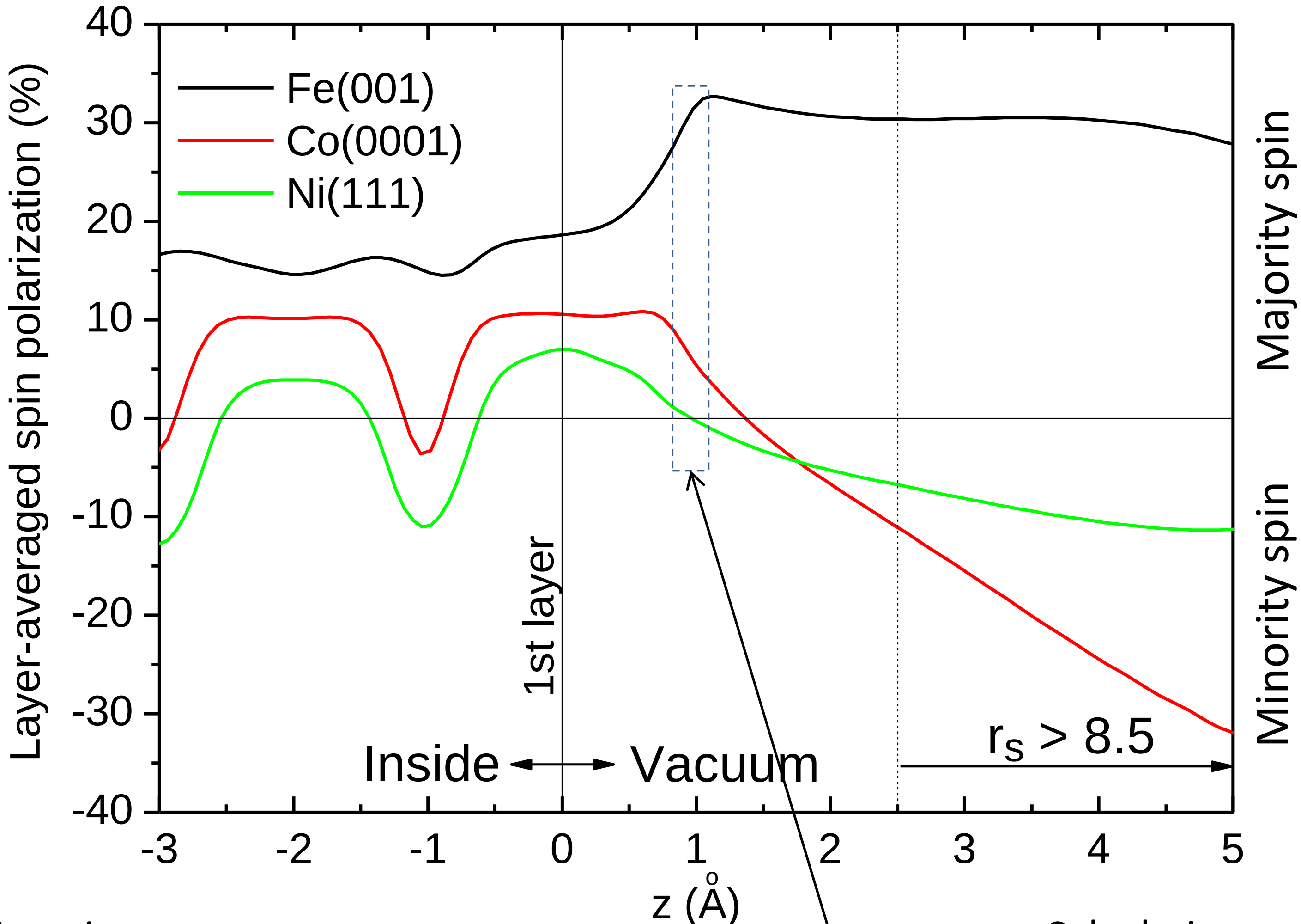
| Fe | Co | Ni |
|---|---|---|
| −3.7% | −2.6% | −0.4% |

Calculation

| Fe | Co | Ni |
|---|---|---|
| −30% | + 12 % | + 7% |

**@$r_s$ = 8.5**
**z >2.5Å**

**Experiment is not well-explained if Ps is formed at $r_s$ > 8.5**



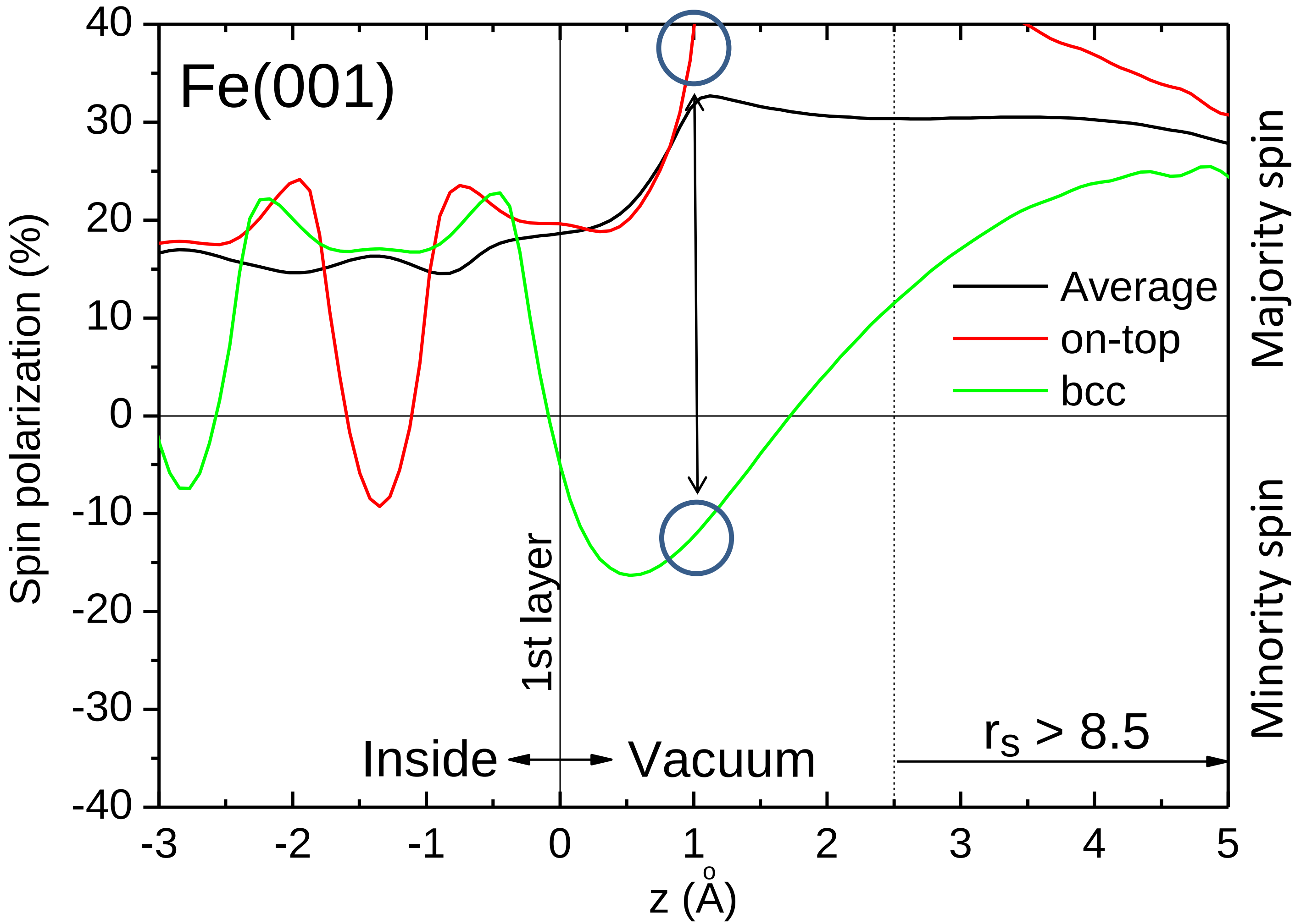

Experiment

| Fe | Co | Ni |
|---|---|---|
| −3.7% | −2.6% | −0.4% |

Calculation

| Fe | Co | Ni |
|---|---|---|
| −31% | −5 % | ~ 0 % |

**Experiment may be explained if Ps is formed at z ~ 1 Å**



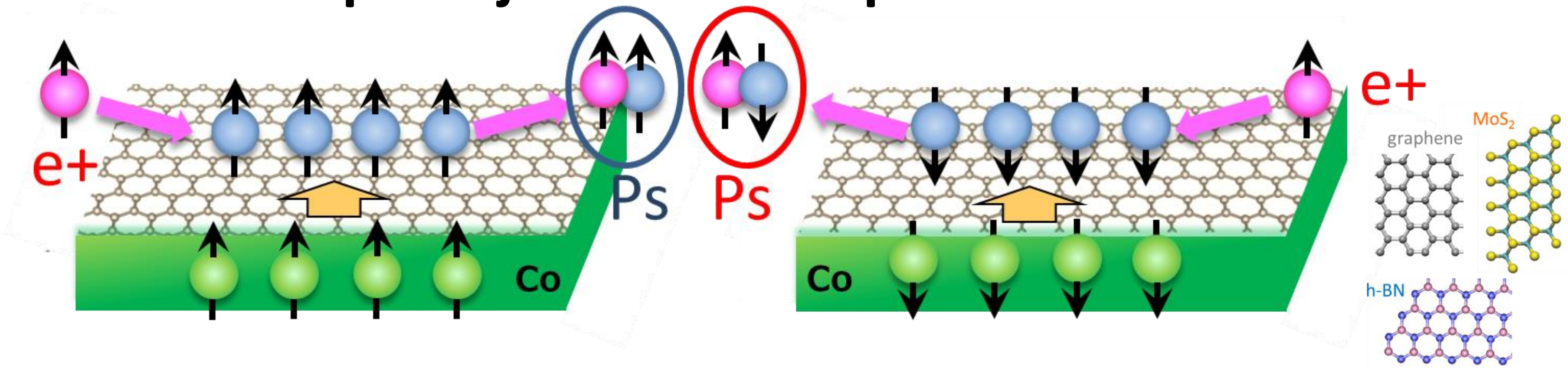

**Polarization is strongly site dependent**

**Suppressed polarization as observed for Fe may reflect spatial distribution of positron wavefunction**

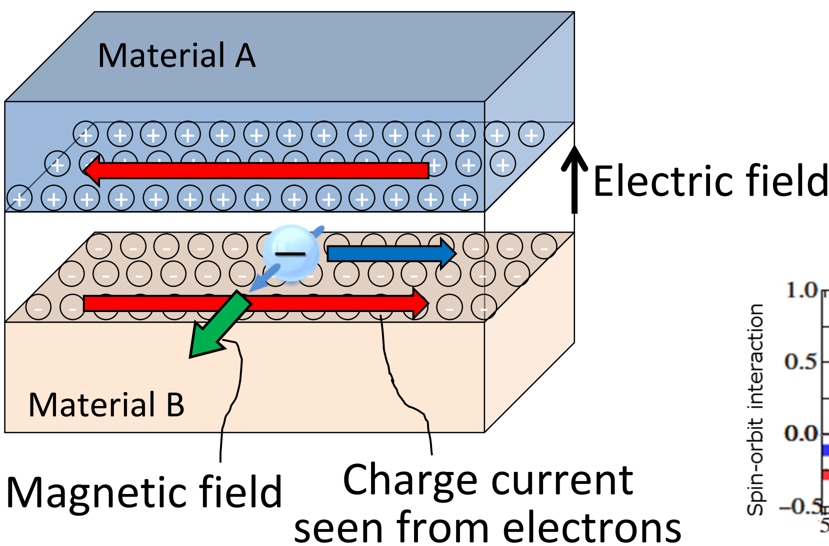
**Further studies about Ps formation is required !**

# Other on-going topics on surface polarization

## Spin-injection to Graphene-related materials

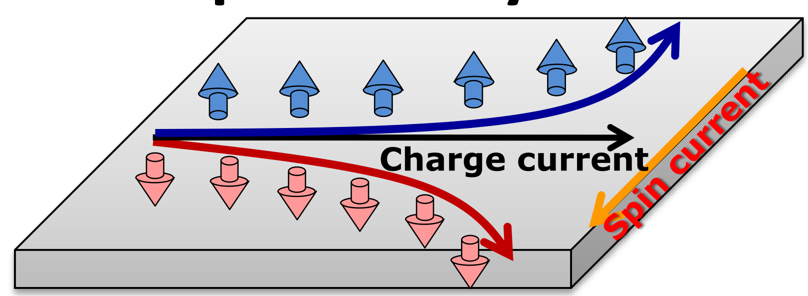


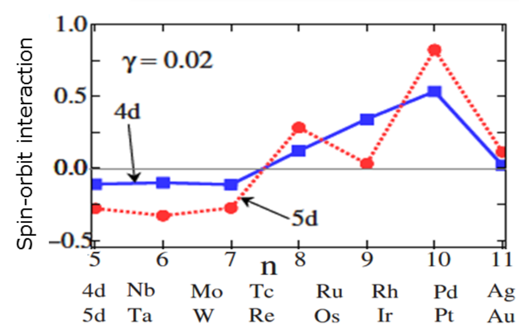

## Rashba systems (2DEG)




Phys. Rev. Lett. 114(2015)166602-1-5.

## Spin-Hall systems

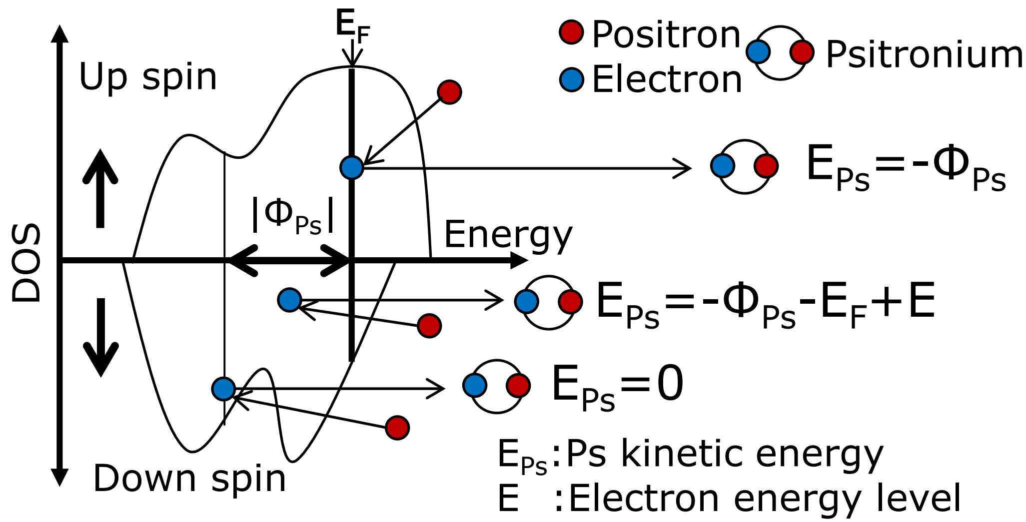


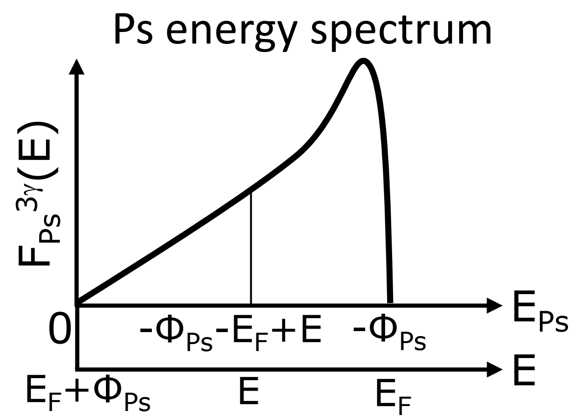




Scientific Reports 4(2014)04844.
J. Mag. Mag. Mater. **342**(2013)139-143.

# Surface Ps Spectroscopy

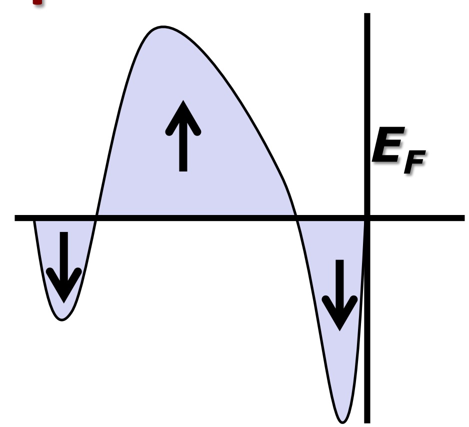
## Relationship between DOS and Ps TOF spectrum




## ● Spin-polarized Ps TOF → Spin-polarized DOS

$$P(E) = \frac{D^{\uparrow}(E) - D^{\downarrow}(E)}{D^{\uparrow}(E) + D^{\downarrow}(E)}$$




**Much detailed**
**surface polarized electronic states!**

# Summary & Outlook:

- Polarized positron beam
- Ferromagnetic band structure
- Vacancy-induced ferromagnetism
- Surface spin polarization

**To establish this method as a standard tool, further experimental and theoretical R&Ds are required.**

| | |
|---|---|
| **Positron annihilation**<br>Vacancy, Band, Precipitate | **SP-Positron annihilation**<br>Polarized band, Magnetic vacancy & precipitate |
| **Positronium Time of Flight**<br>Surface electronic state | **SP-Positronium Time of Flight**<br>Surface spin-polarized band |
| **Positron & Ps Diffraction**<br>Surface structure | **SP-Positron & Ps Diffraction**<br>Surface magnetic structure |
| **Positron-excited 2nd electron**<br>Electronic structure | **Positron-excited SP-2nd electron**<br>Surface spin-polarization, Magnetic domain |
| **Positron-induced Auger electron**<br>Elemental analysis | **Positron-induced SP-Auger electron**<br>Elementary spin analysis |

**"Intense and Highly Polarized Positron Source" will be a great contribution to the community.**