Study of Atomic Processes after Formation of Pionic Atoms

E546: Measurement of Electronic X Rays Correlated with Pionic X rays
E567: Precise Measurement of Electronic X Rays from Pionic Atoms

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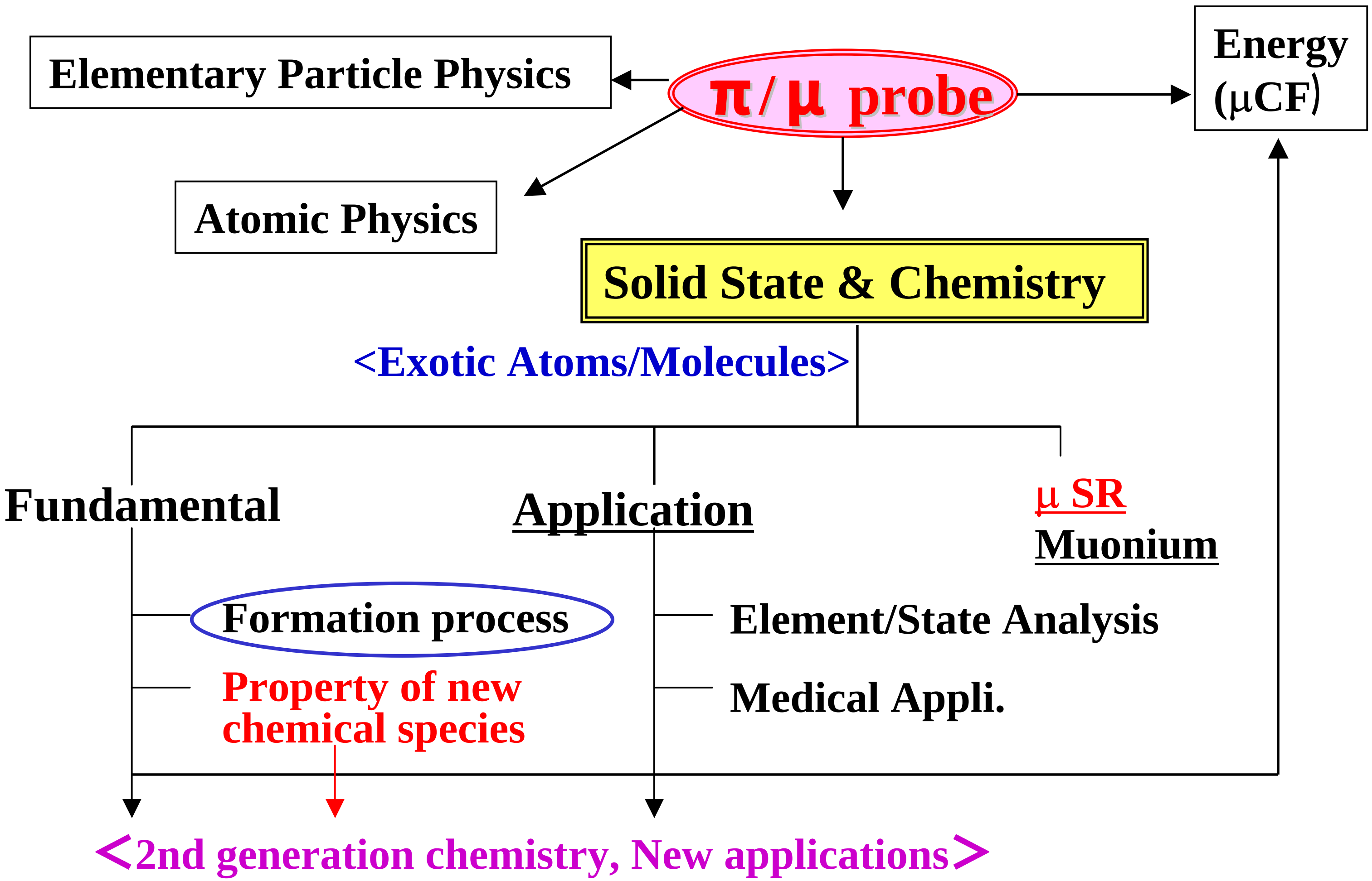
Summary: Negative pion capture process on molecules has been studied for the creation of new chemistry by the 2nd generation substance consisting of muonic and/or pionic atoms. In our previous project (E262 and E360), we studied the pion capture process on hydrogen containing molecules in liquid and gas phases. In this project, we have examined electronic X-ray emission rates correlating with each pionic X ray and measured precisely the energies of electronic X rays to understand electron rearrangement during the pionic cascade in pionic atoms.

Contents of the Presentation

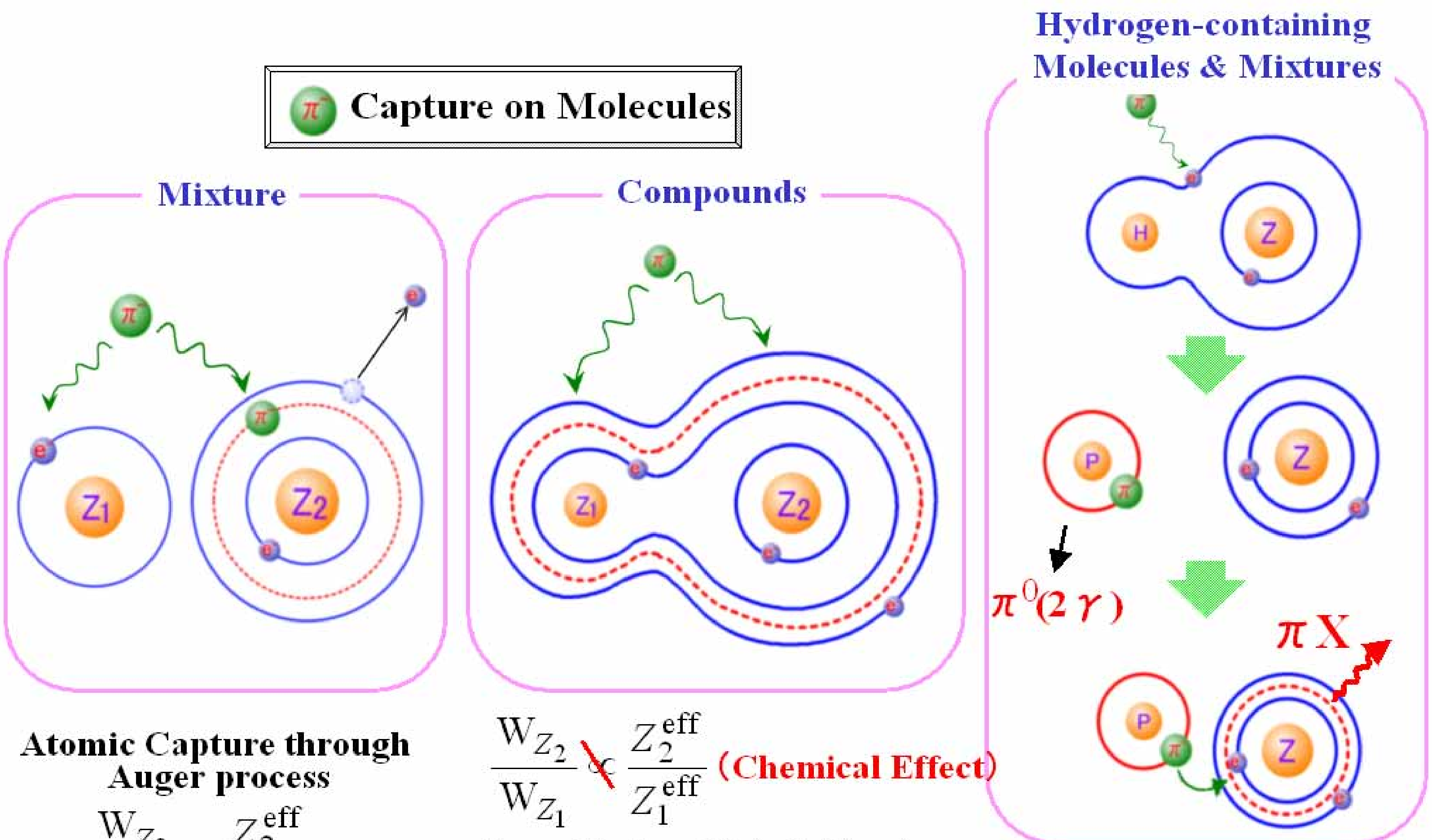
1. **Background of the Project (Exotic Atoms and its Chemistry, Previous Studies of Our Group (πAX))**
2. **Scientific Goal**
3. **Objectives of E546 & E567**
4. **Measurements**
5. **Results and Analysis**
6. **Discussion**
7. **Summary and perspectives**

1. Background of the Project

1-1. Formation of Exotic Atoms and its Chemistry



1-1. Formation of Exotic Atoms and its Chemistry
(Pion Capture Process on Molecules)



Atomic Capture through Auger process

$$\frac{W_{Z_2}}{W_{Z_1}} \propto \frac{Z_2^{eff}}{Z_1^{eff}}$$

(Z−law)

$$\frac{W_{Z_2}}{W_{Z_1}} \not\propto \frac{Z_2^{eff}}{Z_1^{eff}}$$ (Chemical Effect)

Form Electron Dist. & Structure

Capture on π^--LMM orbital

π^- Transfer { External / Internal

1-2. πAX Project (Previous works of our group)

Goal: Comprehensive understanding of the formation mechanism of pionic atoms and its behavior in material to construct the 2-nd generation chemistry and extend to new application of exotic atoms

Pion Capture Process in Molecules Large Mesomoleculer (LMM) Model

Chemical Effect →Mechanism of the Capture Process [E97-E163: 1982-1988]

＜Simple Molecules＞ → Modified LMM Model

Behavior of Pionic Hydrogen Atoms in Molecules

＜Hydrogen-containing Molecules＞

- Pion Transfer Process in Condensed Phases [E202/262: 1989-1995]

→ Chemical Effect in Transfer Process → Comprehensive understanding

Combined LMM model(LMM+Transfer)

- Pion Transfer Process in Gas Phase [E360: 1996-1999]

→ Pressure Dependence of Transfer Rates → Dynamic and Microscopic Model

Review of πAX: A. Shinohara, J. Nucl. Radiochem. Sic., 1(1), 33-37 (2000).

Comparison in π^-p behavior between R-OH and RCOOH

Selective measurement of W_H using the deuterated compounds

$\pi^- p \rightarrow \pi^0 n$

$\pi^- d \nrightarrow \pi^0 n n$

π^-p from C-H

polyethylene

π^-p from COOH/-OH 起源の

→ Different behavior

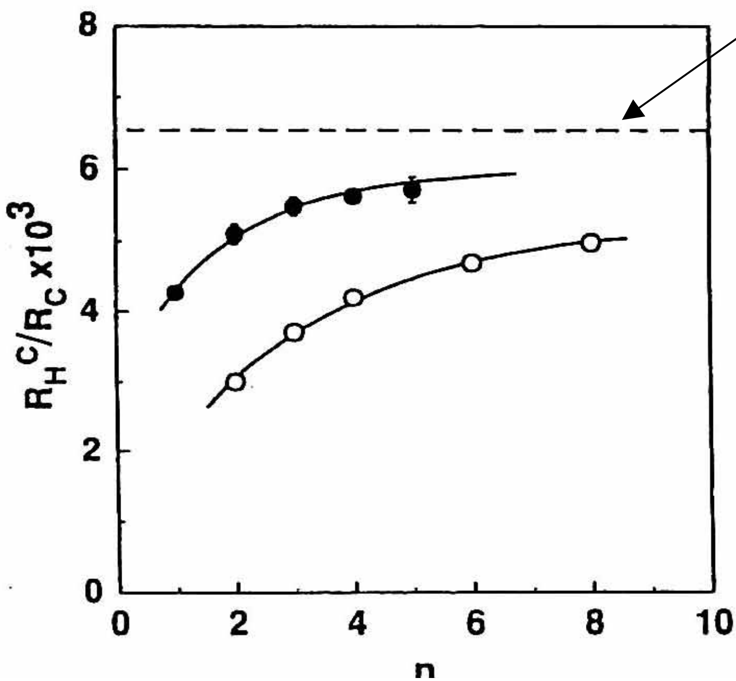


FIG. 1. Capture ratios per atom of the hydrogen bound to carbon relative to the carbon, R_H^C/R_C, as a function of the number (n) of carbon atoms for alcohols (●) and carboxylic acids (○). The dashed line represents the capture ratio for polyethylene. Solid curves are the model calculations with the obtained parameters (see text).

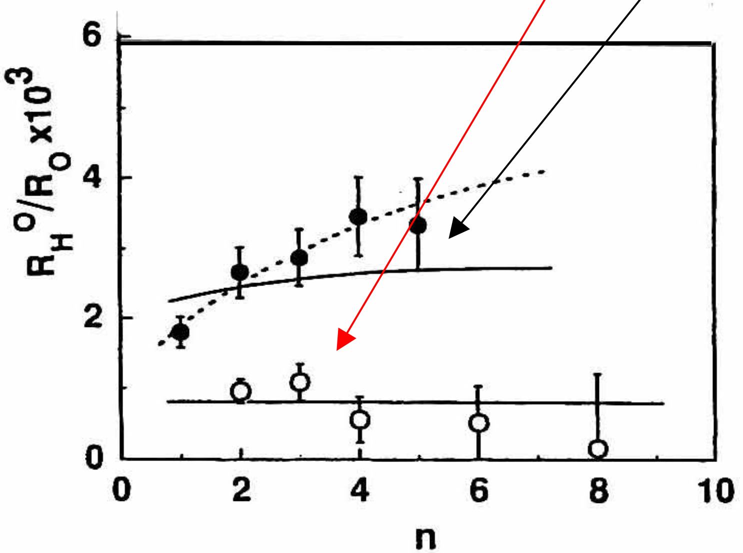
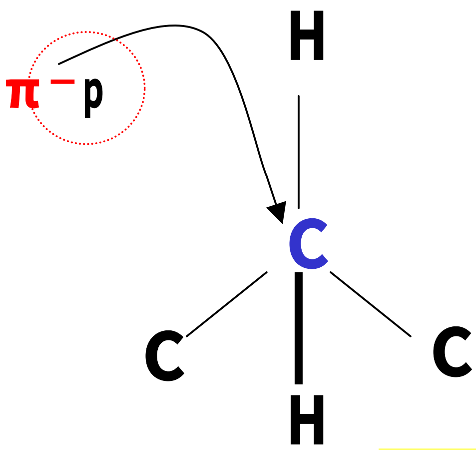
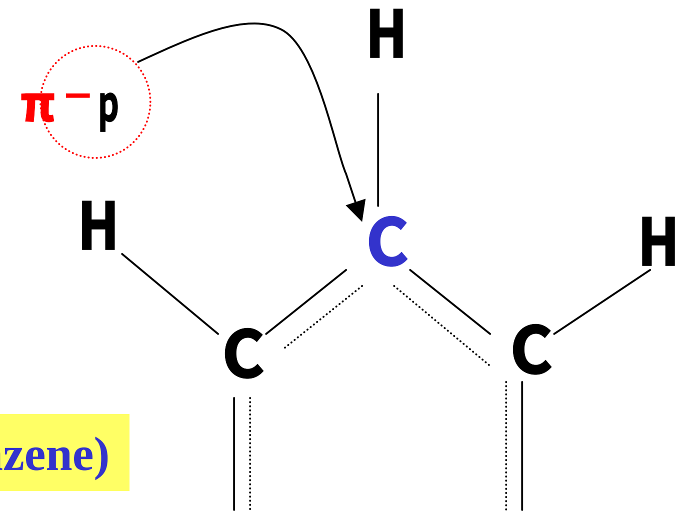


FIG. 2. Capture ratios per atom of the hydrogen bound to oxygen relative to the oxygen, R_H^O/R_O, as a function of the number (n) of carbon atoms for alcohols (●) and carboxylic acids (○). The solid curve represents the model calculations with $\Lambda_C = 1.7$ and $\Lambda_O = 4.1$, and the dotted curve represents those with $a'_\gamma = 0.14$ (see text) for alcohols. No external transfer was found for carboxylic acids.

Ref.: A. Shinohara, et al., Phys. Rev. Lett., 76(14) (1996) 2460-2463.

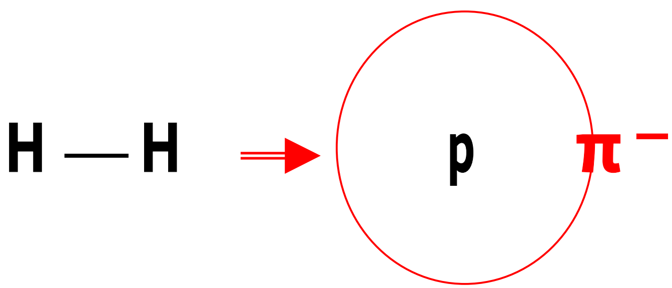
Chemical Effect in the Pion Transfer Process

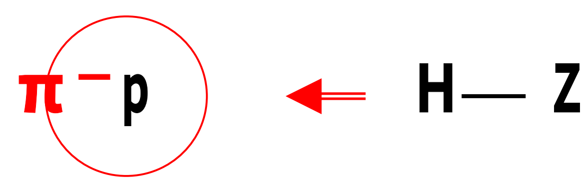




Λ_C(alkanes) < Λ_C(Benzene)

→Structure Effect





Λ(H-H) > Λ(H-Z)

→Atomic State Effect

Refs.: **A. Shinohara, et al.,** Phys. Rev. A, 53(1) (1996) 130-138.;
A. Shinohara, et al., J. Radioanal. Nucl. Chem. 239, 169-173 (1999).;
T. Muroyama, et al., ibd., 239, 159-163 (1999).

Concept of the capture process

(Formation Process of π/μ Atoms)

↓

Atomic Process after the Capture Stage

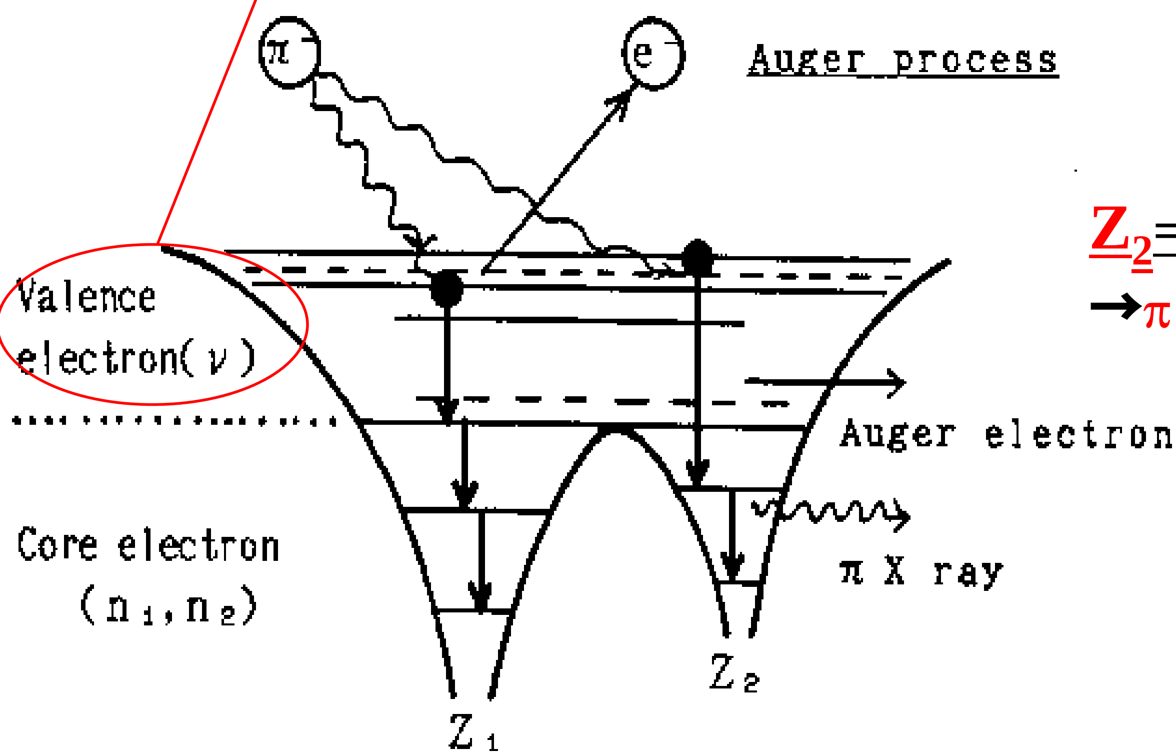
↑

μZ atom → Chemical Species ?

Initial Stage: Capture on a LMM orbital through Auger process

↑

Distribution of Valence Electrons
(Spatial dist. & Structure)



Z_2=1(Hydrogen)

→π^-p (Pseudo Neutron) in Material

LMM
+
Phenomenological Model of Transfer Process
↓
"Combined LMM Model"

2. Scientic Goal of the Project

Creation of the new chemical concept of the 2nd generation substances such as muonic atoms.

(I) Elucidation of a microscopic mechanism for the negative pion/muon capture process

(II) Understanding of the atomic process after formation of the exotic atoms

(III) Several approaches to examine the prospect of muonic atoms as new chemical species

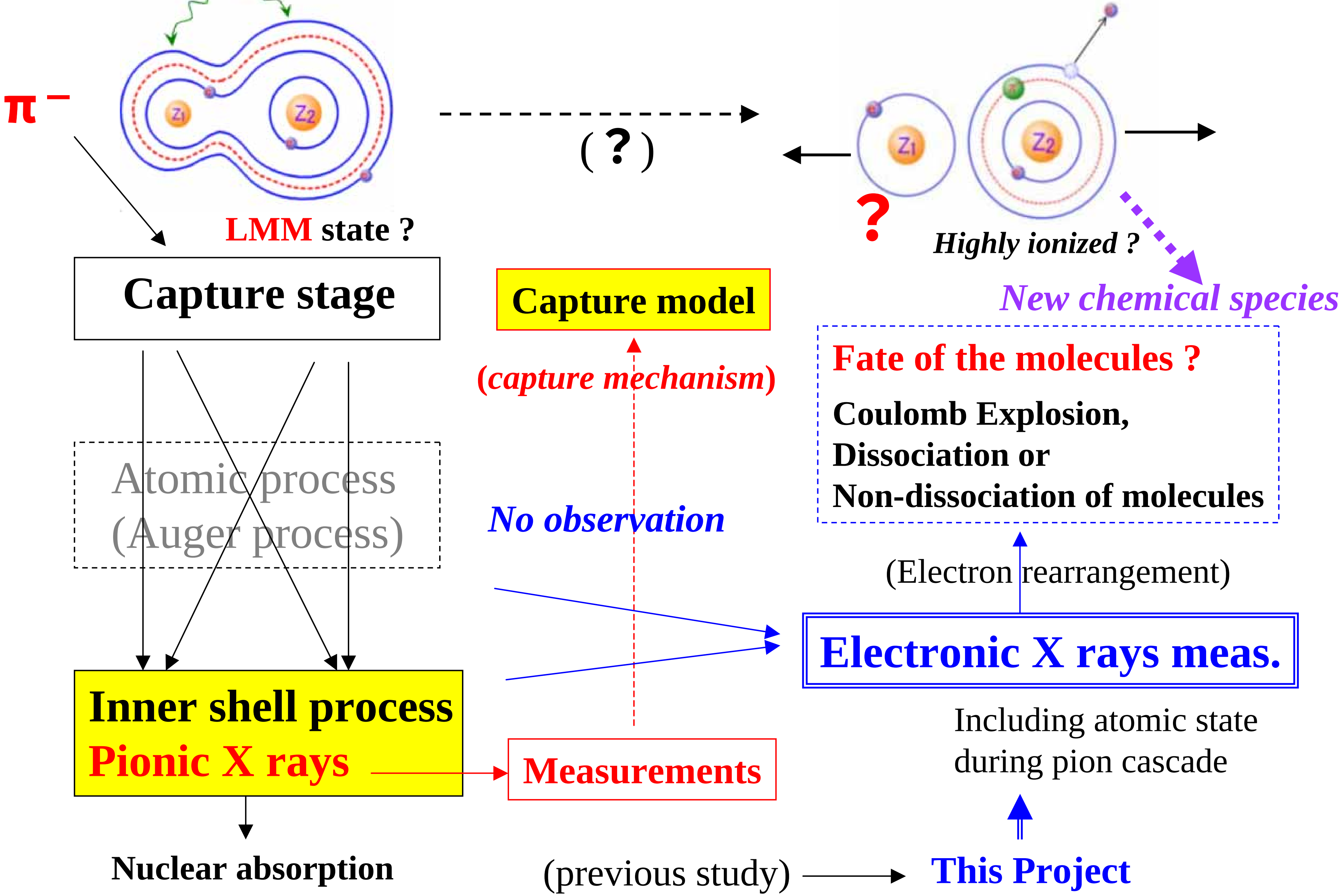
- **Generation of muonic atom beam**
- **Study on the chemical reaction of the muonic atoms by means of a molecular beam method**

Phase-1

Phase-2

Phase-3

Total Process of Pionic Atom Formation



3. Objectives of E546 & E567 (Phase-2)

(Study on the atomic inner shell processes after pion/muon capture)

The electronic X-ray energies of pionic atoms are influenced by the atomic state of the pion and the electron arrangements, especially that of the inner electrons. We have studied on the electron rearrangement process after pionic atom formation by measuring electronic X rays emitted from pionic atoms ranging from zinc to uranium.
[→Experiments at KEK-PS and KEK-MSL]

Exp-1: Dependence of the energy shifts of the electronic K X rays on the atomic number

Exp-2: Electronic X-ray intensity ratio (K_β/K_α) for several compounds

Atomic inner shell processes after pion/muon capture

Atomic Process: Electronic X rays (e X rays) and Auger process in the (Z-1)'atomic state during the pionic cascades in Z atom.

Exp-1: Energy shifts of the electronic K X rays

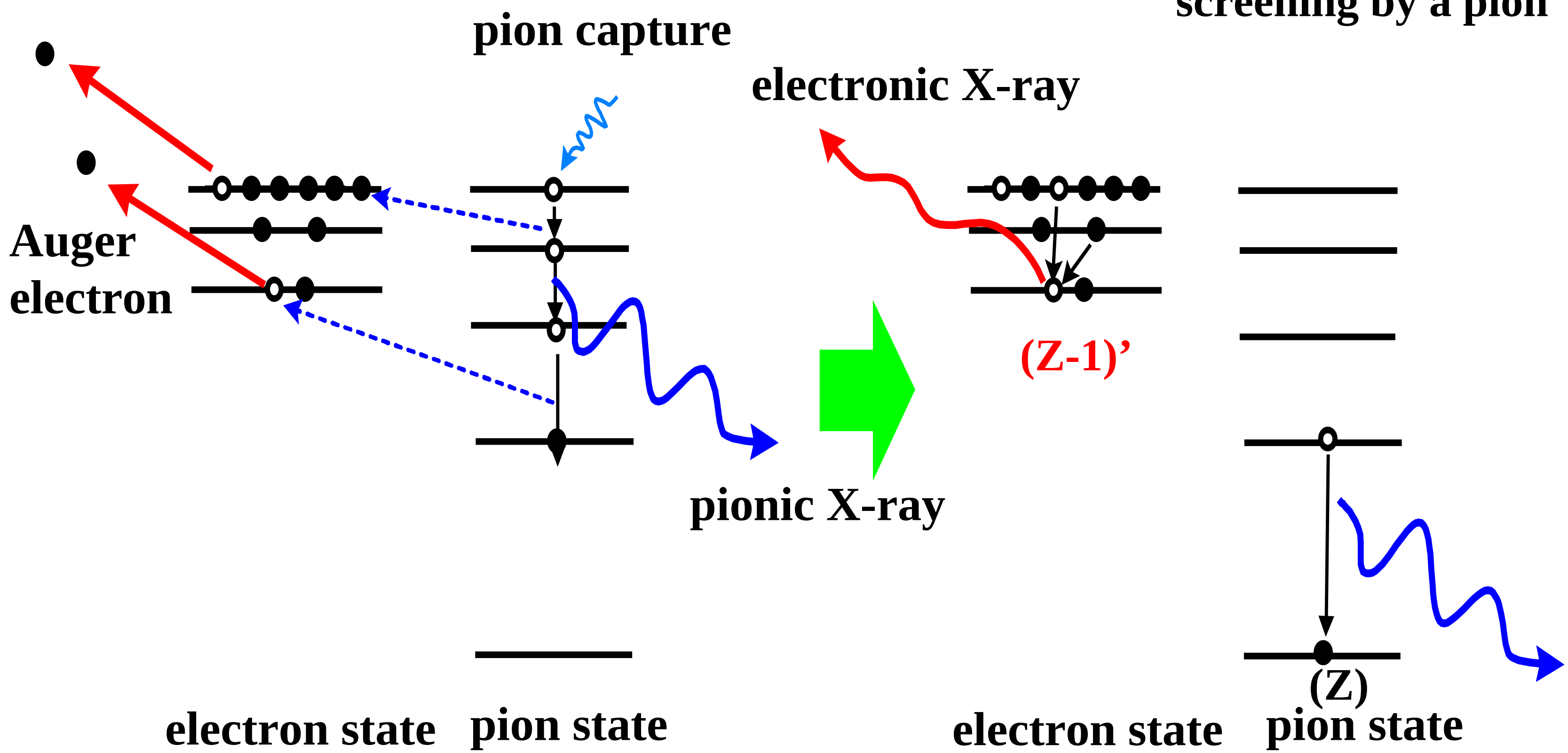
X-ray Energy: E_X

Energy Shift: $\triangle E = E_X(\mu Z) - E_X(Z-1)$

$E_X(\pi Z) > E_X(Z-1)$

because of incomplete screening by a pion

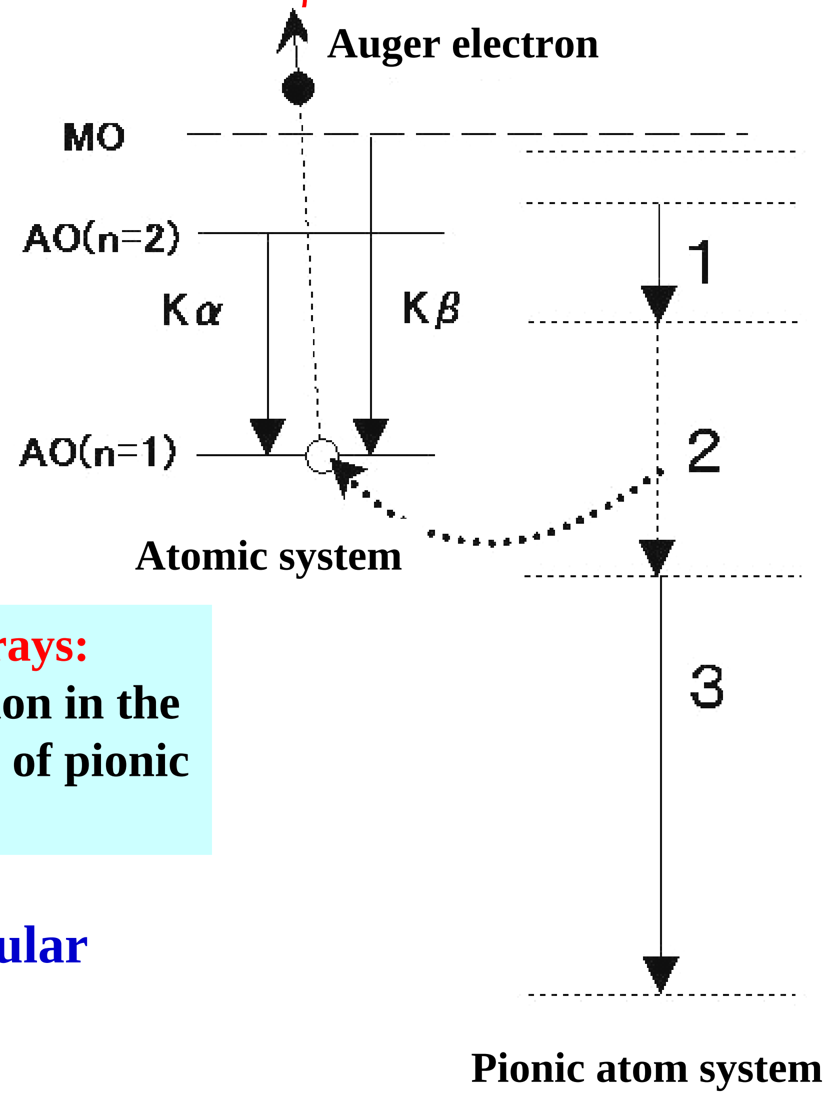
Electron Hole Creation



Exp-2: Electronic X-ray intensity ratio (K_α/K_β)

What will happen to the pion captured molecule ?

Dissociation or non-dissociation of molecule including a pionic/muonic atom



Concept of e and π X-rays correlation measurement

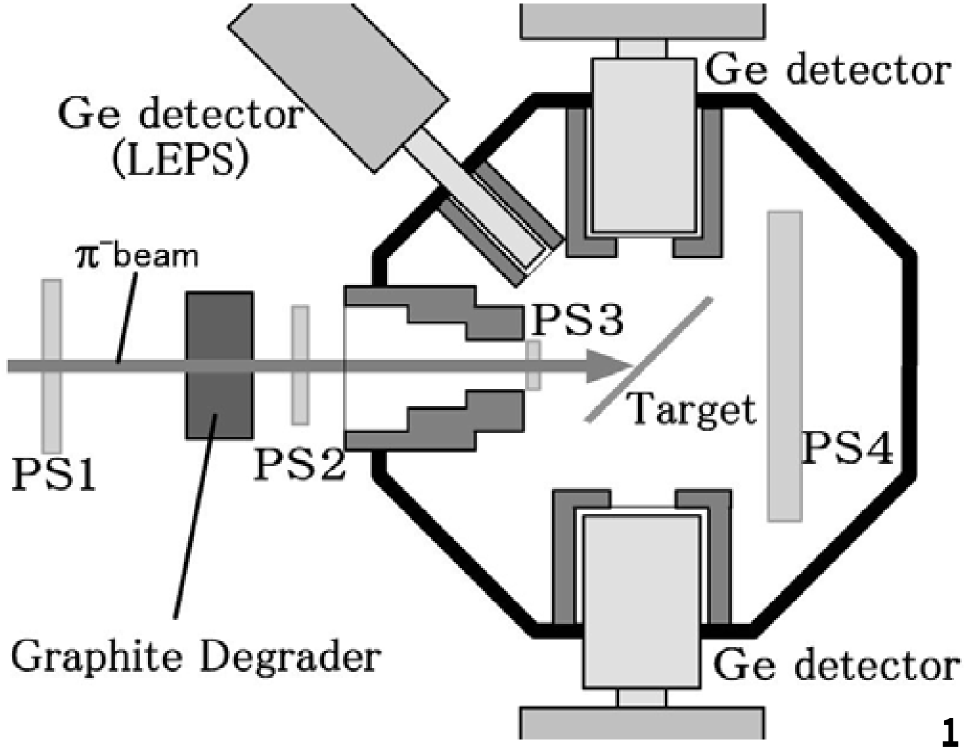
Electronic X-ray fine structure (Kβ/Kα)

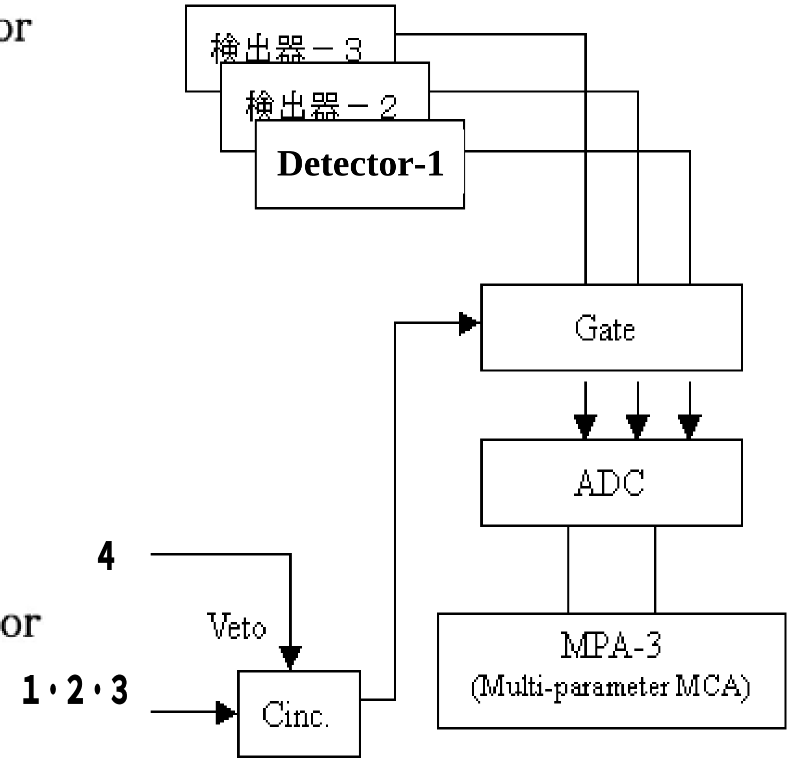
Pionic X rays:
Observation in the final state of pionic atoms

We can obtain information on the molecular state, if the valence electrons (MO) participate in the K_β X-ray emission.

4. Measurements

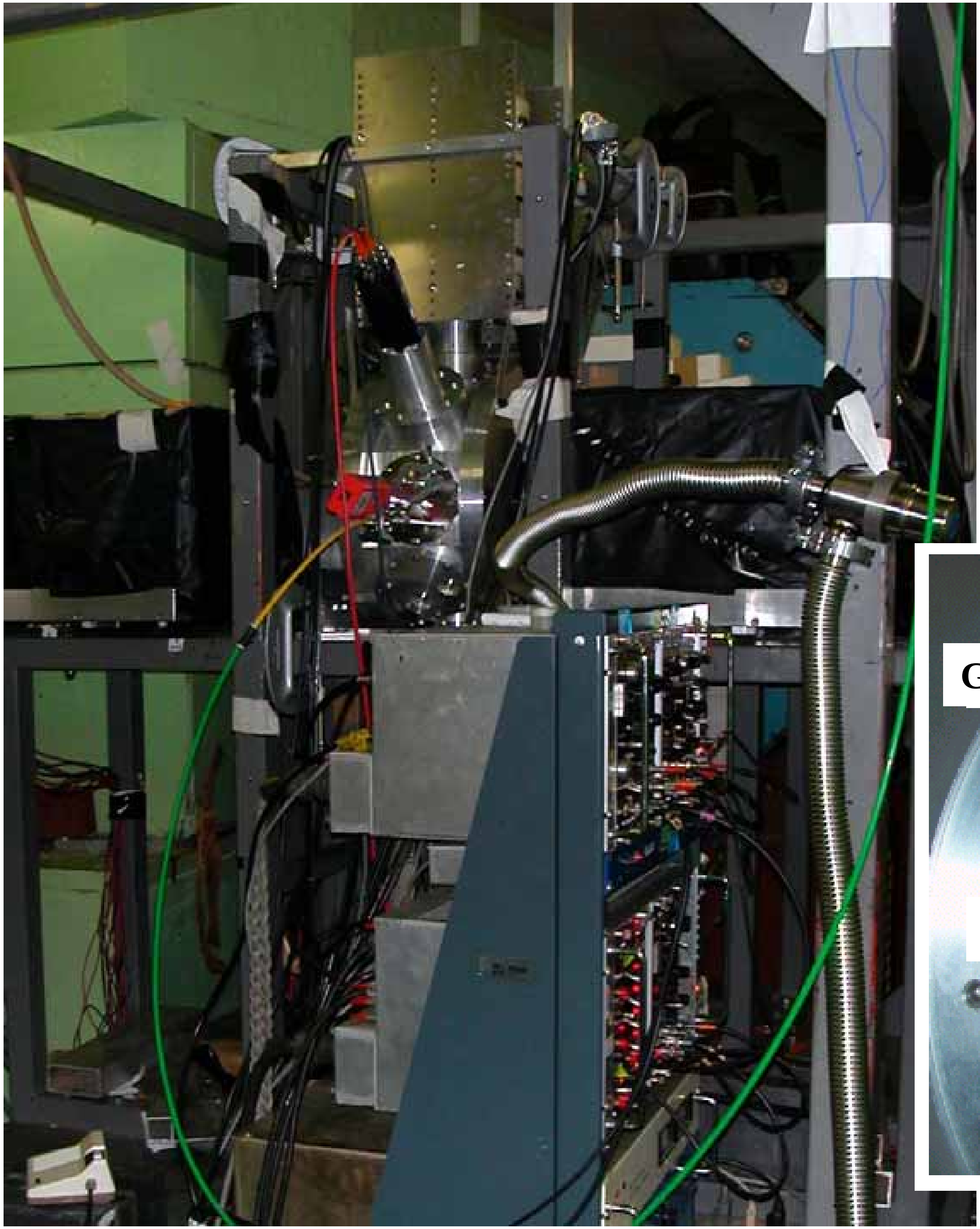
Outline of a measuring system





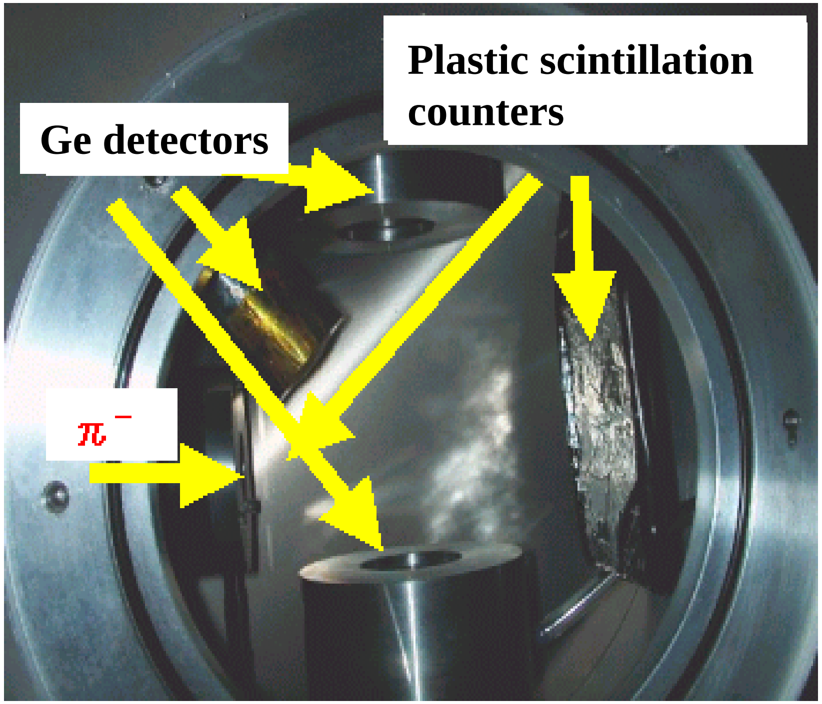
at π μ channel of KEK-PS

- **Data taking: List mode recording of energy and timing signals of each detector**
- **He-flow chamber installed plastic scintillation counters (PS-3 & PS-4)**
- **Energy range: 3 keV 〜 800 keV**



■ Overview of the experimental setup at πμ-chennel of EP-2

■ Interior of the measuring chember



Executed Beam Time

E546

Beam line: π μ (K2-parasite)
Approved beam time: 35 shifts
Executed beam time: 30 shifts
Executed cycle: 03[6-1]

Experimental

- Construction of improved setup
- Data taking： 13 samples (Zn, Mo-MoO3, Sn-SnO2, Gd2O3, Ho-Ho2O3, Yb2O3, Lu2O3, Ta, HgO, Pb)

E567(+extension)

Beam line: π μ (K2-Parasite)
Approved beam time: 30＋15 shifts
Executed beam time: 55 shifts
Executed cycles: 05[1、4-2]

Experimental

- Data taking： 18 samples (Cu, Ag, NaI, Xe, CsF, BaO, Eu2O3, Tb2O3, Dy2O3, Er2O3, Tm2O3, Lu2O3, Hf, W, Ir, Au, Tl2O3, UO2(ac)2

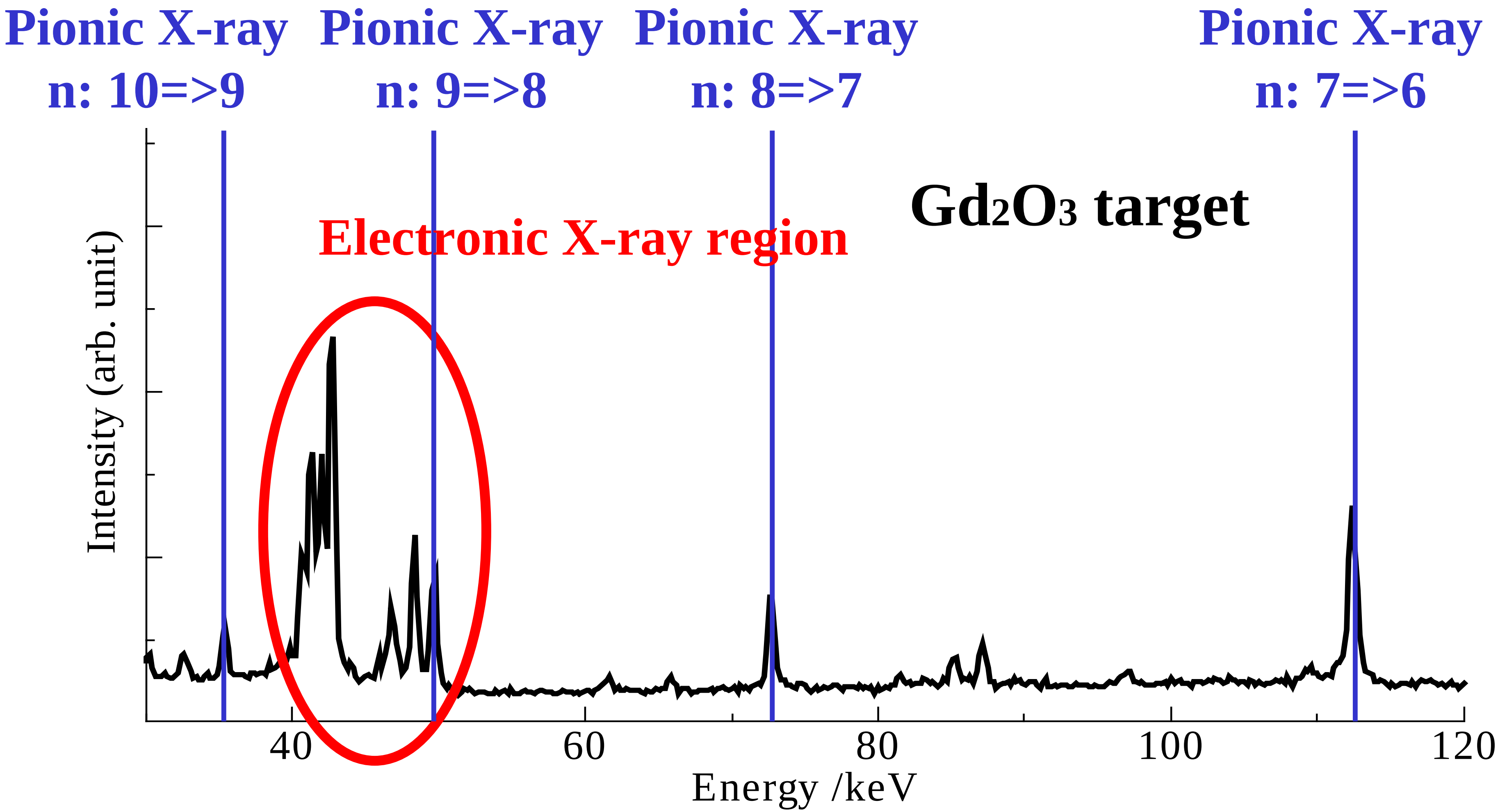
→ Our proposals were accepted as the K2 or EP2 parasite experiments.
Some test experiments were also performed to develop the measuring system.

Measured sample list

Target	atomic number	Thickness (g/cm^2)	total STOP event /10^6	Target	atomic number	Thickness (g/cm^2)	total STOP event/10^6
Cu	29	0.179	126	Ho_2O_3	67	0.530	64
Zn	30	0.143	112	Er_2O_3	68	0.577	161
Mo	42	0.051	120	Tm_2O_3	69	0.587	166
Ag	47	0.210	148	Yb_2O_3	70	0.488	93
Sn	50	0.073	170	Lu_2O_3	71	0.621	127
NaI	53	0.806	235	Hf	72	0.665	233
Xe	54	1.09	113	Ta	73	0.333	40
CsF	55	0.834	244	W	74	0.386	48
BaO	56	0.622	208	Ir	77	1.13	101
Eu_2O_3	63	0.294	109	Au	79	0.578	68
Tb_4O_7	65	0.491	93	HgO	80	0.688	130
Dy_2O_3	66	0.516	116	Tl_2O_3	81	0.356	151
Gd_2O_3	64	0.449	82	Pb	82	0.340	55
Ho	67	0.220	23	$UO_2(ac)_2$	92	0.574	356

5. Results and Analysis

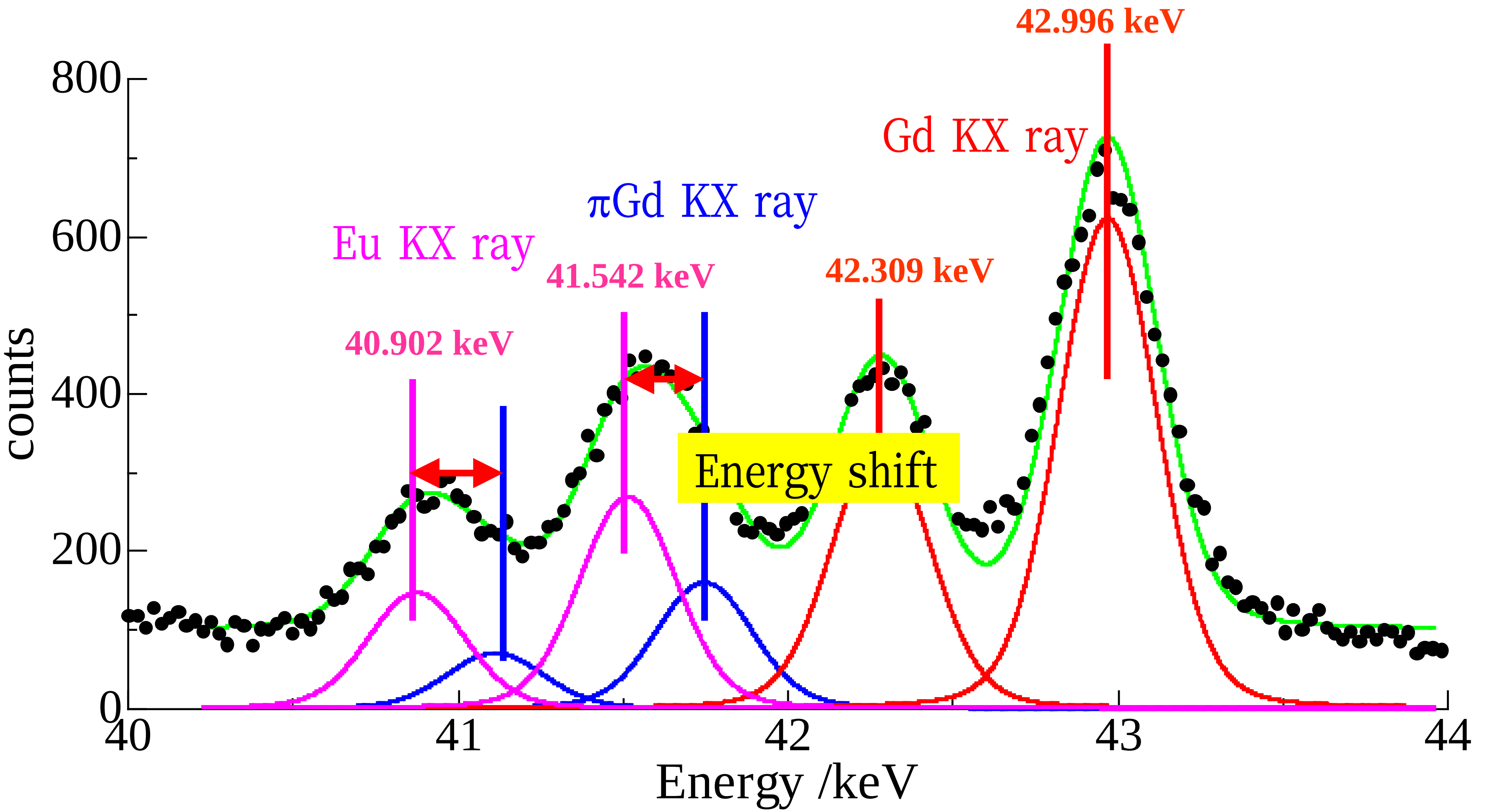
■ Measured photon spectrum



"n" means principle quantum number

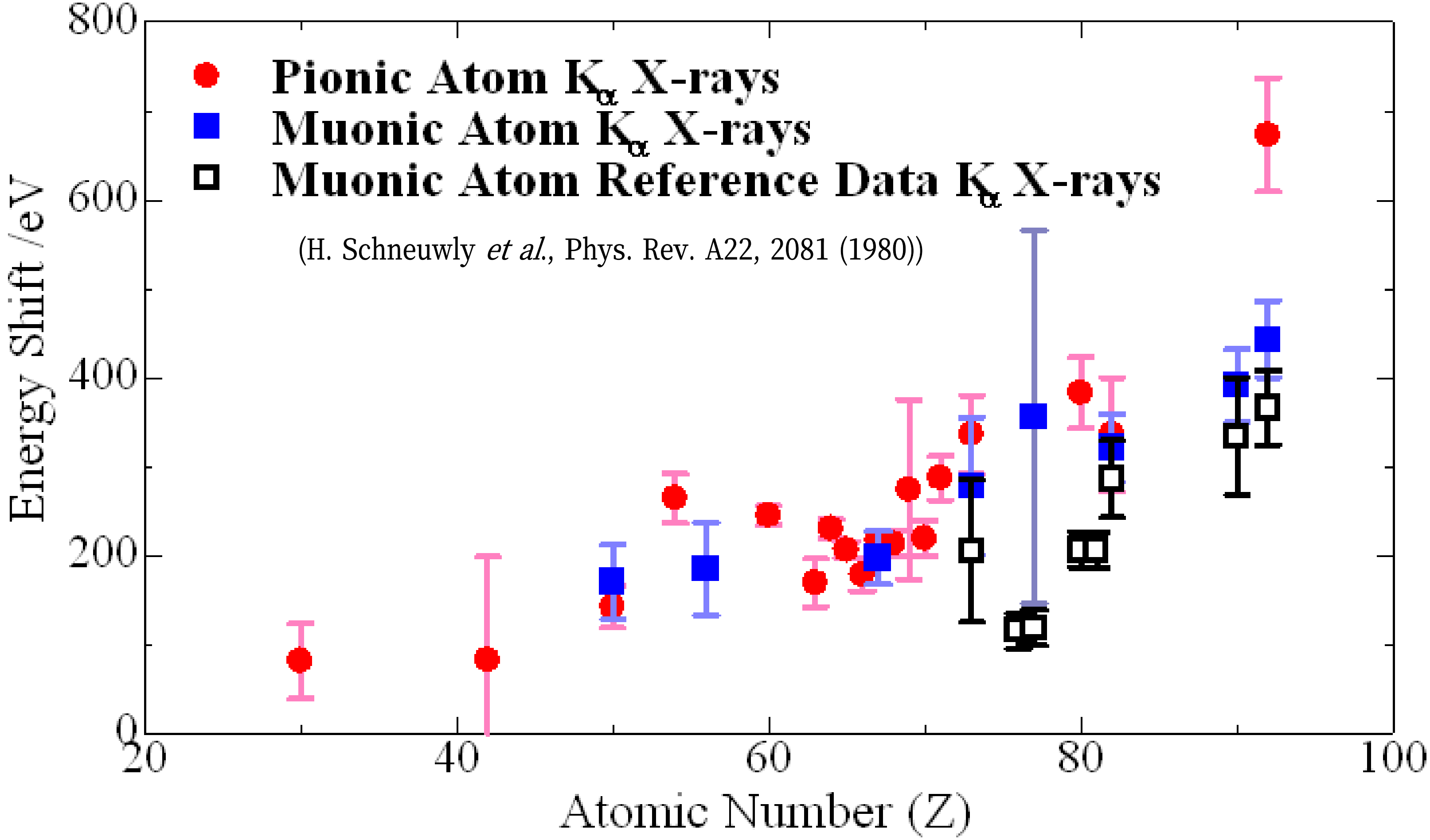
Peak Analysis

Example-1︰ X-ray spectrum for a Gd_2O_3 target with its fitting lines. The peak centers of the electronic X-ray and the intensity ratio of $K\alpha_1$ / $K\alpha_2$ for Z and Z-1 atoms were fixed in the fitting analysis.



Results of Exp-1 → Discussion

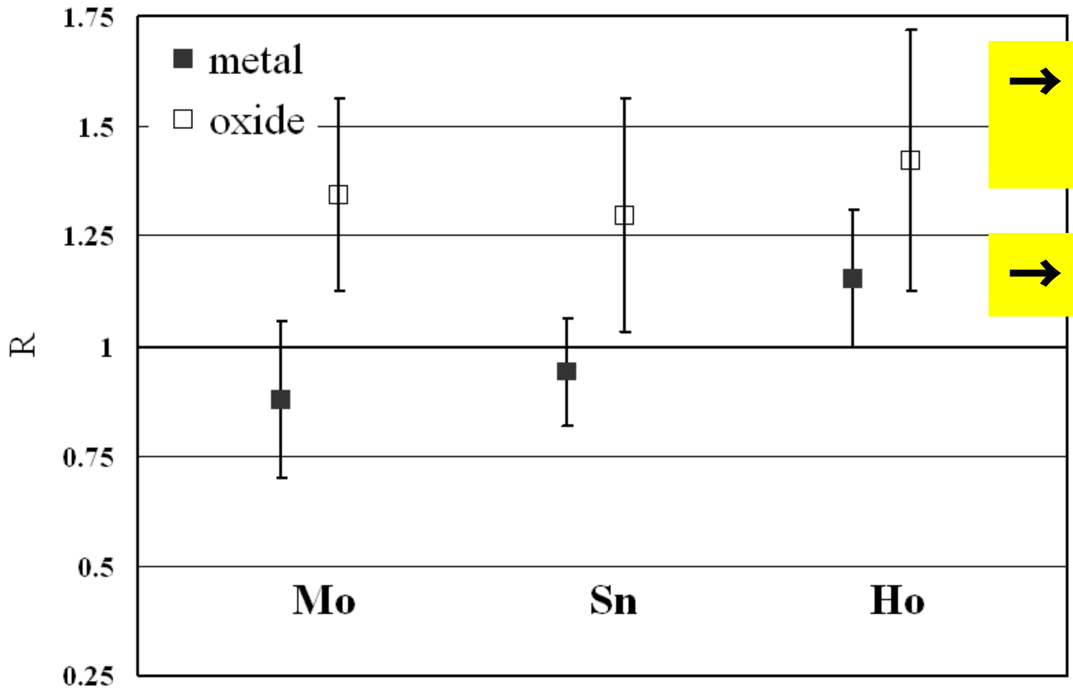
Atomic number dependence of the energy shift of the electronic Kα X rays



Ref.: K. Ninomiya, et al., Radiochim. Acta 93, 515-518 (2005);
J. Radioanal. Nucl. Chem., 272(3), 661-664 (2007).

Results of Exp-2

Electronic KX-ray structures (K_β/K_α) correlated with several pionic X-ray emissions



→ Obvious difference between metal and the oxides

→ Highly ionized atomic state

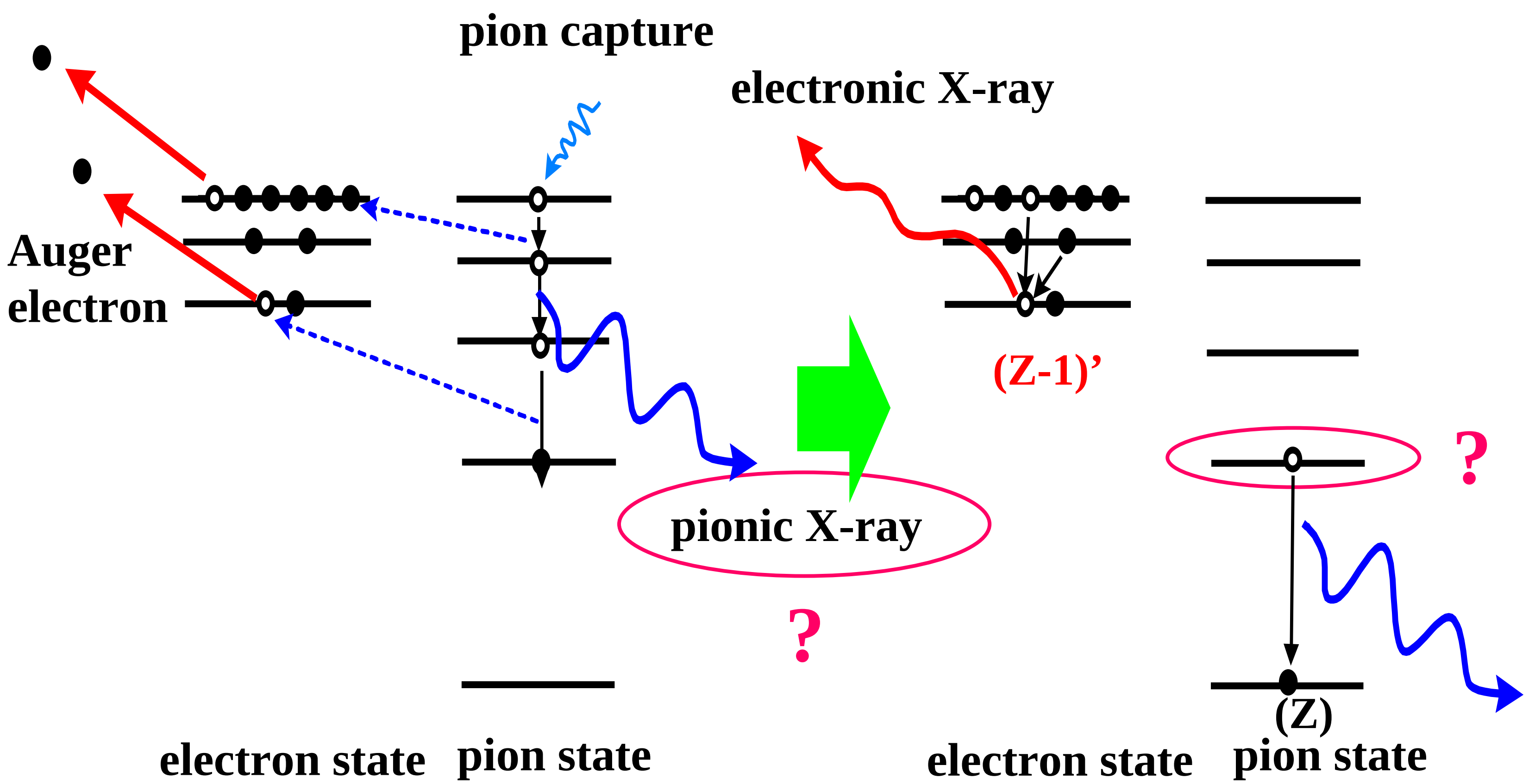
→ No information on the molecular state

R = (K_β/K_α ratio of in-beam experiment) / (K_β/K_α ratio of photoionization experiment)
The intensity ratios are taken as averaged values of those correlated with several pionic X rays.

6. Discussion

6.1. Qualitative discussion

■ K-hole creation and the pion orbital



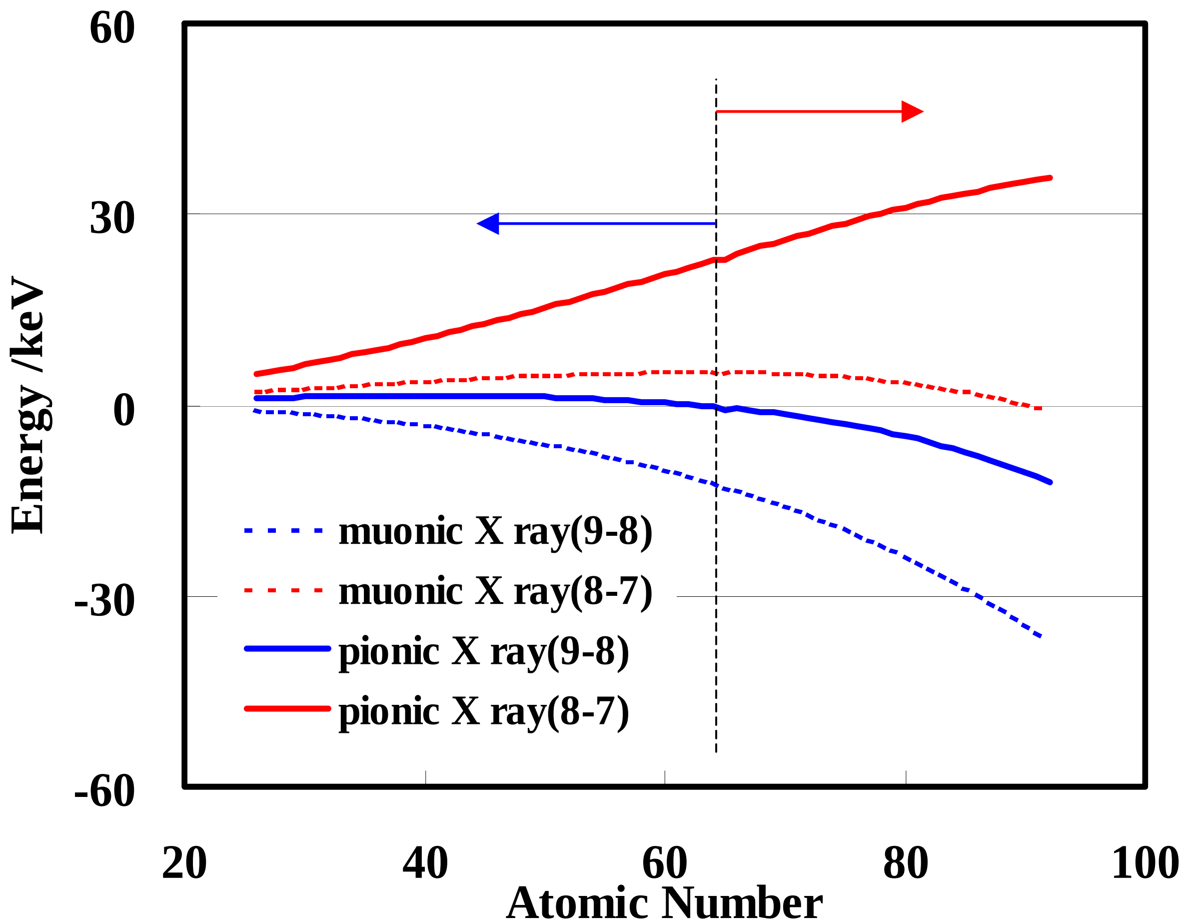
■ Qualitative discussion:

π/μ atomic transition causing K-hole creation

(Comparison between Mesonic transition energy and K-shell binding energy)

(K-shell binding energy) - (mesic X-ray energy) > 0 : Auger Process

→ eKX Ray Emission



K shell refilling rate > π μ transition rate

The KX ray is emitted, when the pion or muon is in the final state of the relevant transition.

Muonic atoms: n=7

Pionic atoms: n=8 for Z<65

n=7 for Z>65

Z-dependence:

Energy shift (n=8) > (n=7)

Energy shift (high-Z) > (low-Z)

6.2. Quantitative discussion

■ Cascade Calculation

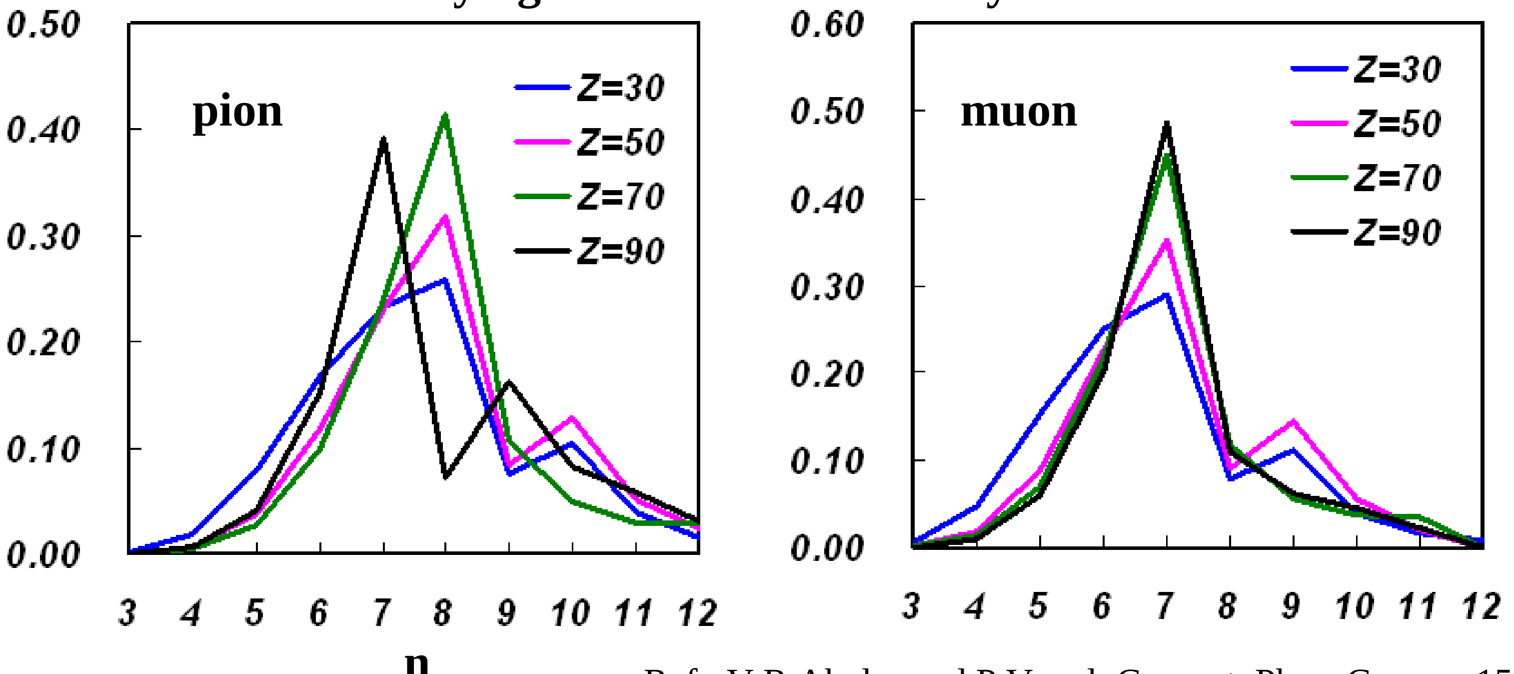
Using Cascade Calculation for Muonic Atom (by Akylas-Vogel code [ref.]),
Determination of the Parameters ← Reproducibility of muonic or pionic X ray intensity patterns

↓

(Calculation under the optimum conditions)

↓

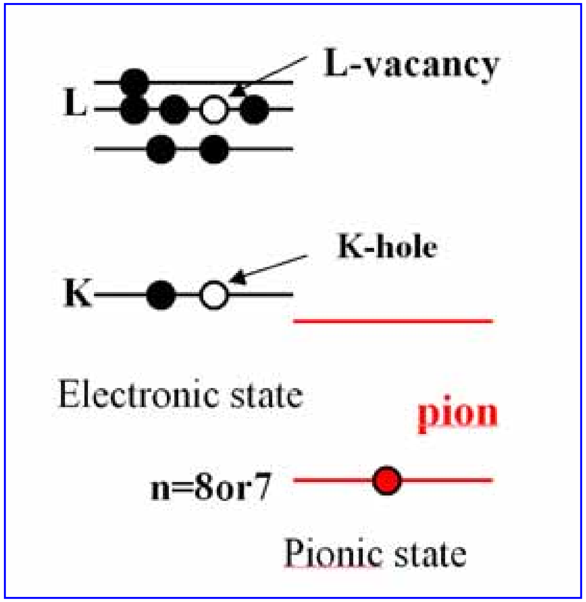
K-hole creation prob. → Pionic or muonic orbital (n) on which the pion or muon is lying at emission of K X ray



Ref.: V.R.Akylas and P.Vogel, Comput. Phys. Comm., 15, 291(1978).

Calculation of energy level of atomic system containing a pion or muon

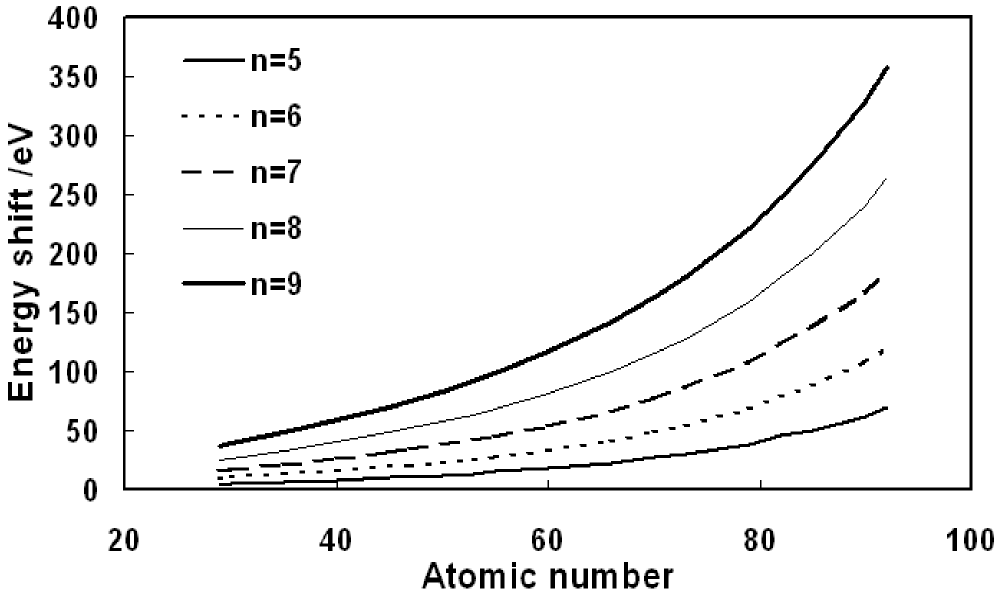
Calculation method: MCDFGME code [ref.]
This code can calculate the total energy of atomic system containing one exotic particle on a particular orbit.

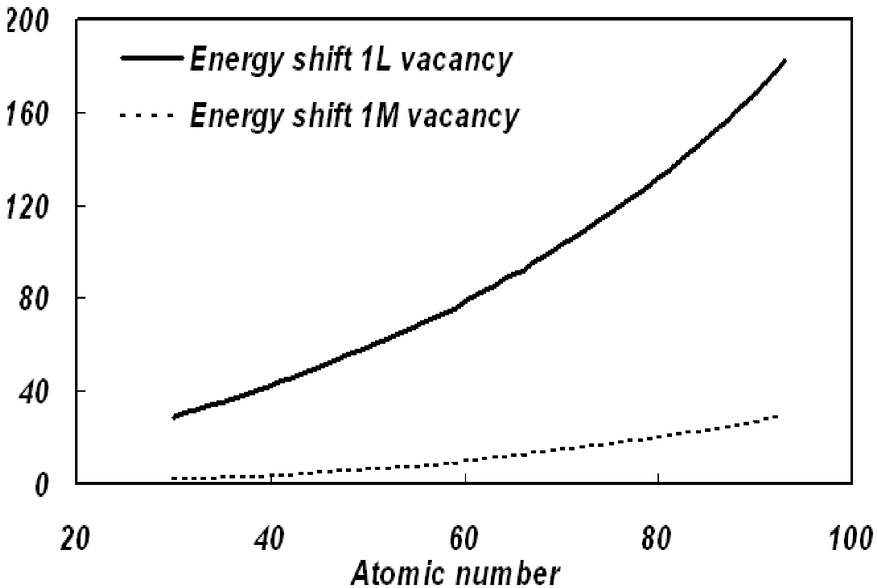


Calculation for Pionic Atom (Z) ⇔ for Ordinary Atom (Z-1)

↓

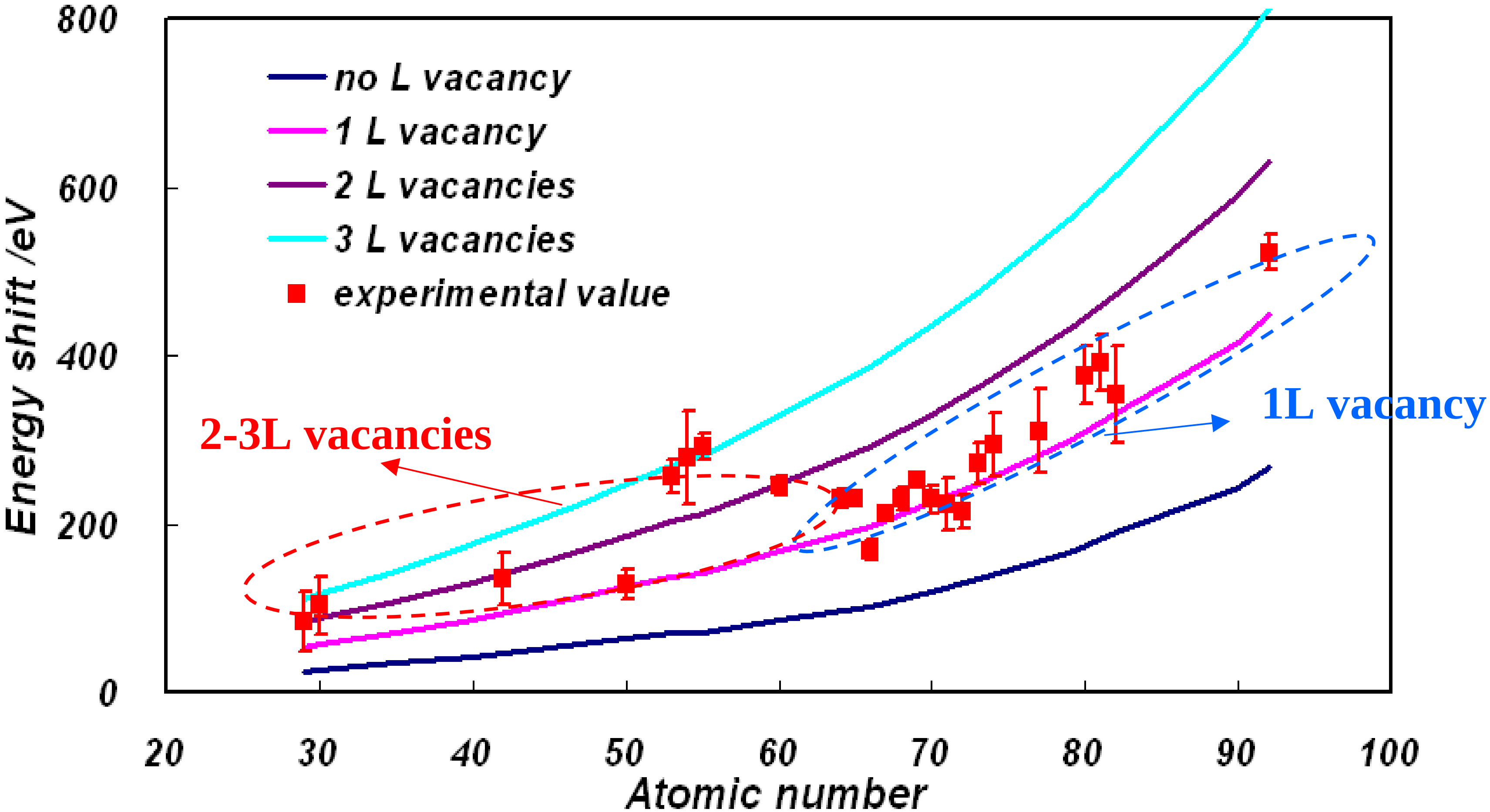
Energy shift : for various pionic state (n) and L vacancies





Ref.: J.P.Desclaus and P.Indelicato, MCDFGME code, http://dirac.spectro.jussieu.fr/.

Comparison between the theoretical and experimental energy shifts under the condition of various L vacancies



7. Summary and Perspectives

1) Energy shifts in electronic KX rays of pionic atoms were measured for various metal and compounds ranging Z=29 to 92.
2) Atomic process after formation of pionic atoms was discussed from a comparison between the energy shifts obtained and theoretical calculations based on a static model.
3) It was suggested that the energy shift is caused by the incomplete screening effect of orbital pion as well as L electron vacancies created by the pionic cascade.

Atomic state after pion/muon capture is not perturbed than one expected.
→ Possibility of chemical reaction of muonic atoms ?
→ Another formation method of pionic/muonic atoms ?

New proposal (→ RIKEN-RAL, *J-PARC*)

- **Formation process of muonic atoms and molecules through the muon transfer from muonic hydrogen**
- **Chemical reaction of muonic atoms**

Publication List

Original Papers

[1] K. Ninomiya, et al., "Energy Shift of Electronic X Rays Emitted from Pionic Atoms", Radiochimca Acta. **93**, 515-518 (2005).

[2] K. Ninomiya, et al., "Study of Electronic X rays Emitted from Pionic and Muonic Atoms " J. Radioanal. Nucl. Chem., **272**(3), 661-664 (2007).

Papers presented at international conferences

[1] K. Goto, et al., "Development of the Measuring System for Electronic X Rays Following Atomic Capture of Negative Pions", Asia-Pacific Symposium on Radiochemistry 2001, Fukuoka, Oct. 30 – Nov. 1, 2001.

[2] K. Ninomiya, et al., "Energy Shift of Electronic X rays Emitted from Pionic Atoms", 6th International Conference on Nuclear and Radiochemistry (NRC6), Aachen, Germany, 8/29-9/3 2004.

[3] K. Ninomiya, et al., "Electronic X-ray Energies Emitted from Pionic and Muonic Atoms", The 6th International 21 Century COE Symposium on Integrated EcoChemistry(COEIEC 6), Osaka, Japan, 2005.10.11-13.

[4] K. Ninomiya, et al., "Study of electronic X rays emitted from pionic and muonic atoms", Asia-Pacific Symposium on Radiochemistry (APSORC-05), Beijing, China, 2005.10.17-21.

[5] K. Ninomiya, et al., "Electronic KX-ray Energy Shift of Pionic and Muonic Atoms", The 8th International 21st Century COE Symposium on Integrated EcoChemistry, Kyoto, 2006.8.28-2.

[6] K. Ninomiya, et al., "Atomic Structure of Pionic and Muonic Atoms during Atomic Cascade ", The 9th International 21st Century COE Symposium on Integrated EcoChemistry, Kyoto, 2007.1.16-18.