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Analysis of Permeation Induced Transmutation from the Aspects of Deuterium Density and Electronic Structure in Pd Multilayer film (Pd/CaO/Pd)

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[1]Advanced Technology Research Center, Mitsubishi Heavy Industries, ltd., [2]Research Facility Center for Science and Technology, University of Tsukuba, [3]Institute of Industrial Science, Univ. of Tokyo,[4]Laboratory of Nuclear Science, Tohoku Univ., [5]Japan Synchrotron Radiation Research Institute (JASRI), [6]RIKEN SPring-8 Center, Hyogo Japan

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1.Introdcution

Features of the Present Method

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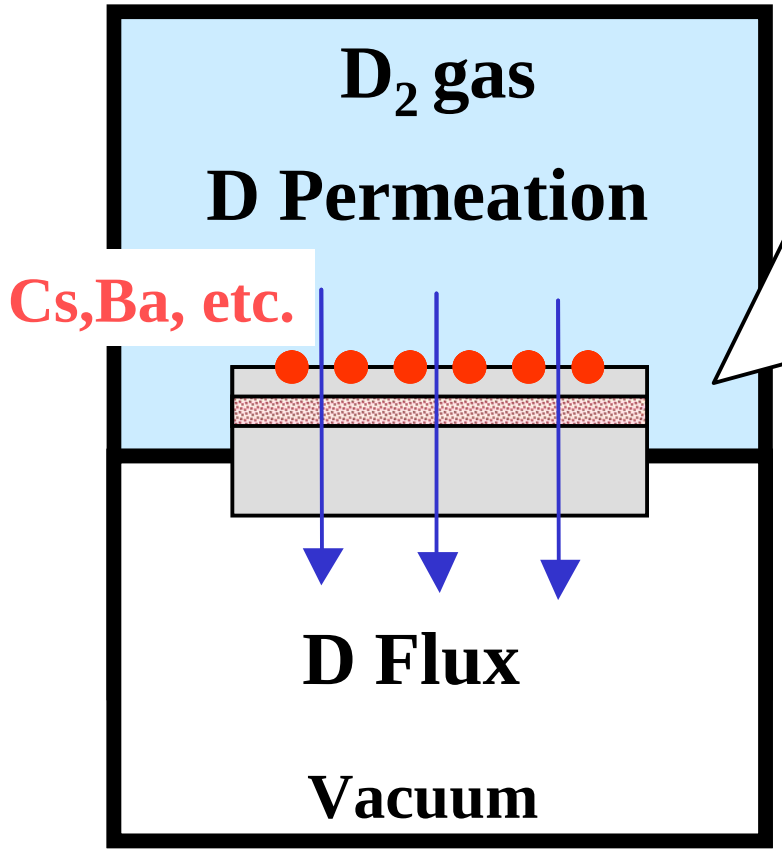
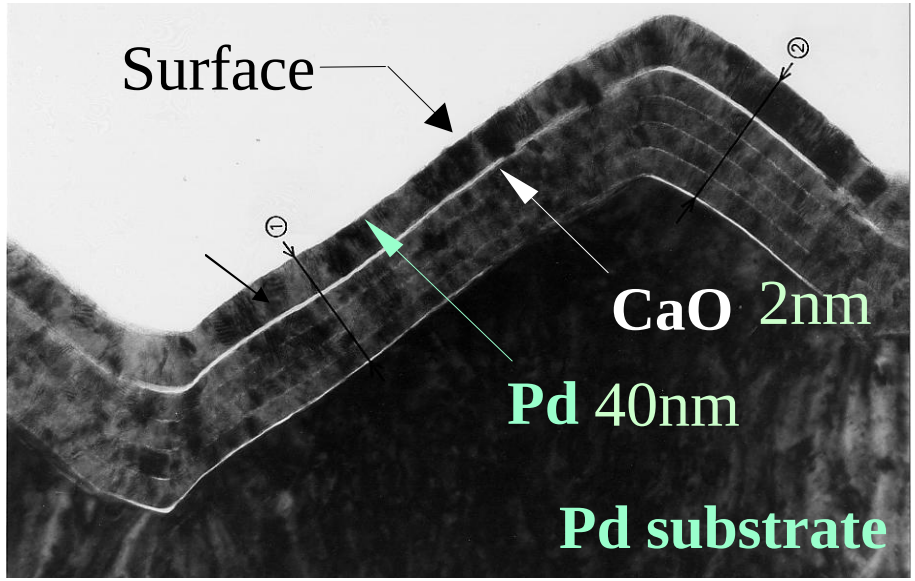
D_2 gas permeation through the Pd Complex



Pd nano-sized multilayer film



Reactions observed so far in MHI

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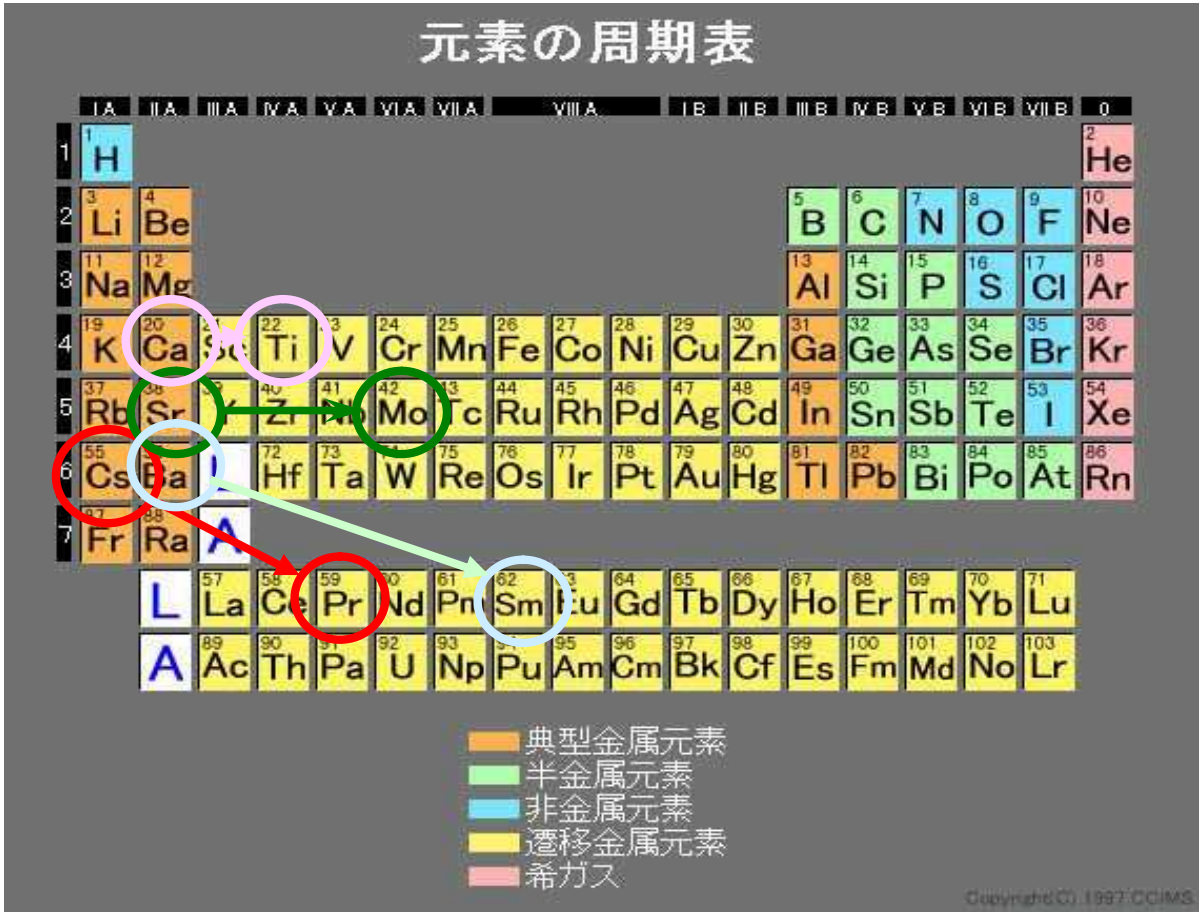

$$^{133}_{55}Cs \xrightarrow{4d(2\alpha)} {}^{141}_{59}\mathrm{Pr}$$

$$^{88}_{38}Sr \xrightarrow{4d(2\alpha)} {}^{96}_{42}Mo$$

$$^{138}_{56}Ba \xrightarrow{6d(3\alpha)} {}^{150}_{62}Sm$$

$$^{137}_{56}Ba \xrightarrow{6d(3\alpha)} {}^{149}_{62}Sm$$

$$^{44}_{20}Ca \xrightarrow{2d(\alpha)} {}^{48}_{22}Ti$$

Alkali and alkaline earth metals seem to be transmutable.

Similar Experiments at Iwate Univ.

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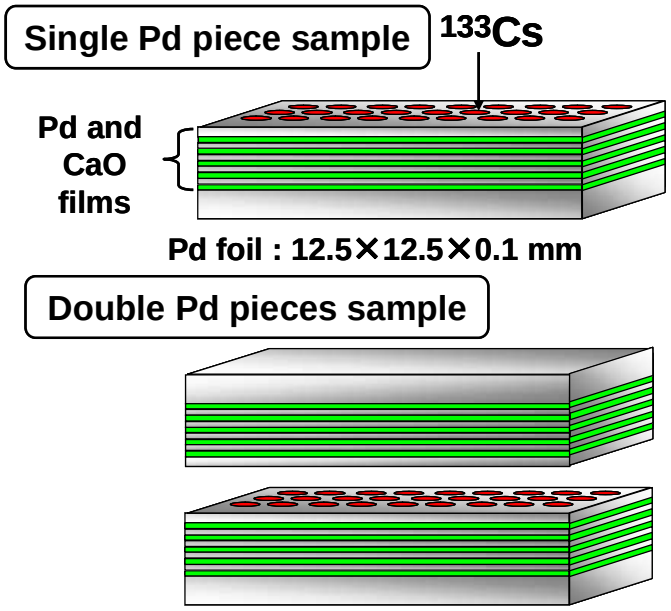

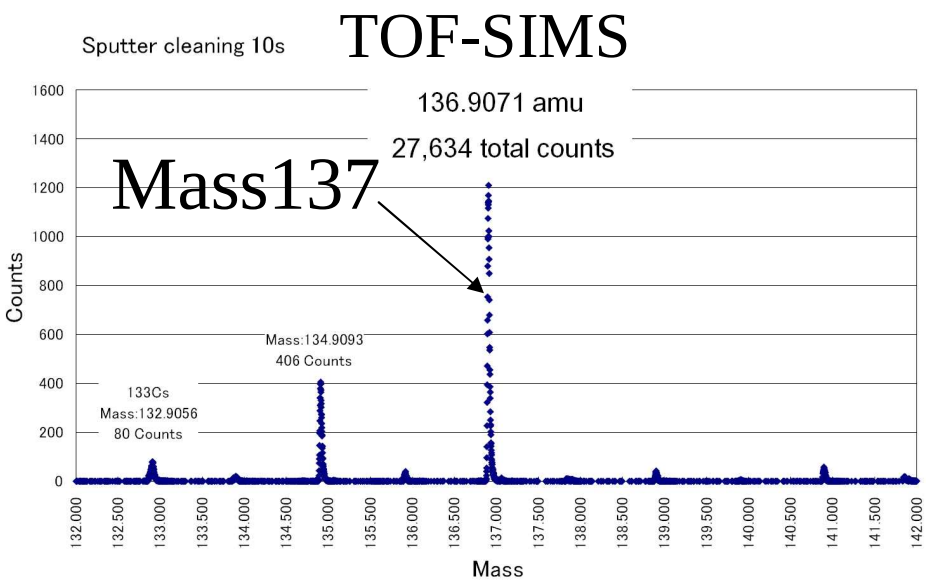

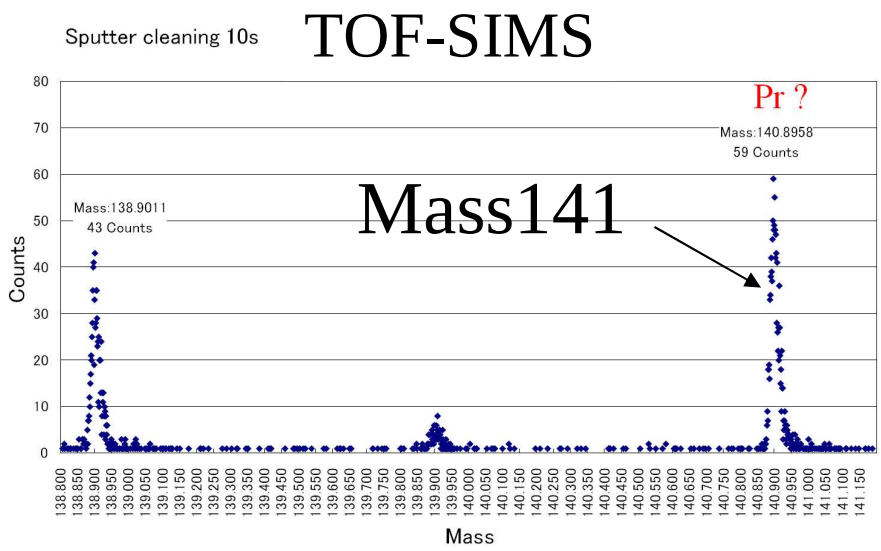

Increase of mass number 137 by deuterium permeation using both single and double Pd/CaO/Pd film.

Increase of mass number 141 only when ^{133}Cs was given.

Key factors in permeation experiments

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Key factors based on experimental results

Candidates for key factors

Evidence

D Flux
Temperature
Work function
Stress
Grain boundary
Nano size
D/Pd
Target element
Etc…
CaO Multilayer
Plasmon

More Essential Factors Necessary !

Theoretical Models

Hypothesis

Local Deuteron Density
Electronic Structure
Hopping Rate
……

Hydrogen density measurement using a resonant nuclear reaction

First Principle Cluster Calculation

ICCF15

2-1 Experimental Evidences related to the importance of *Local Deuteron Density*

Detection of Localized Pr

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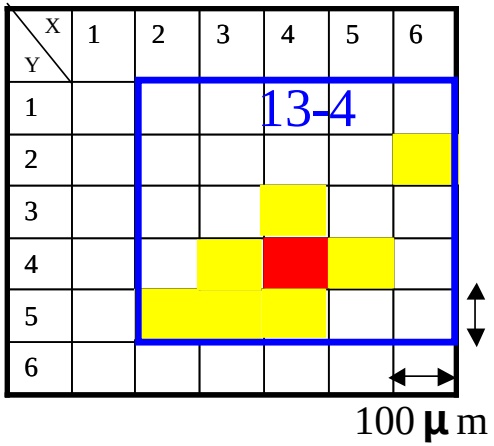
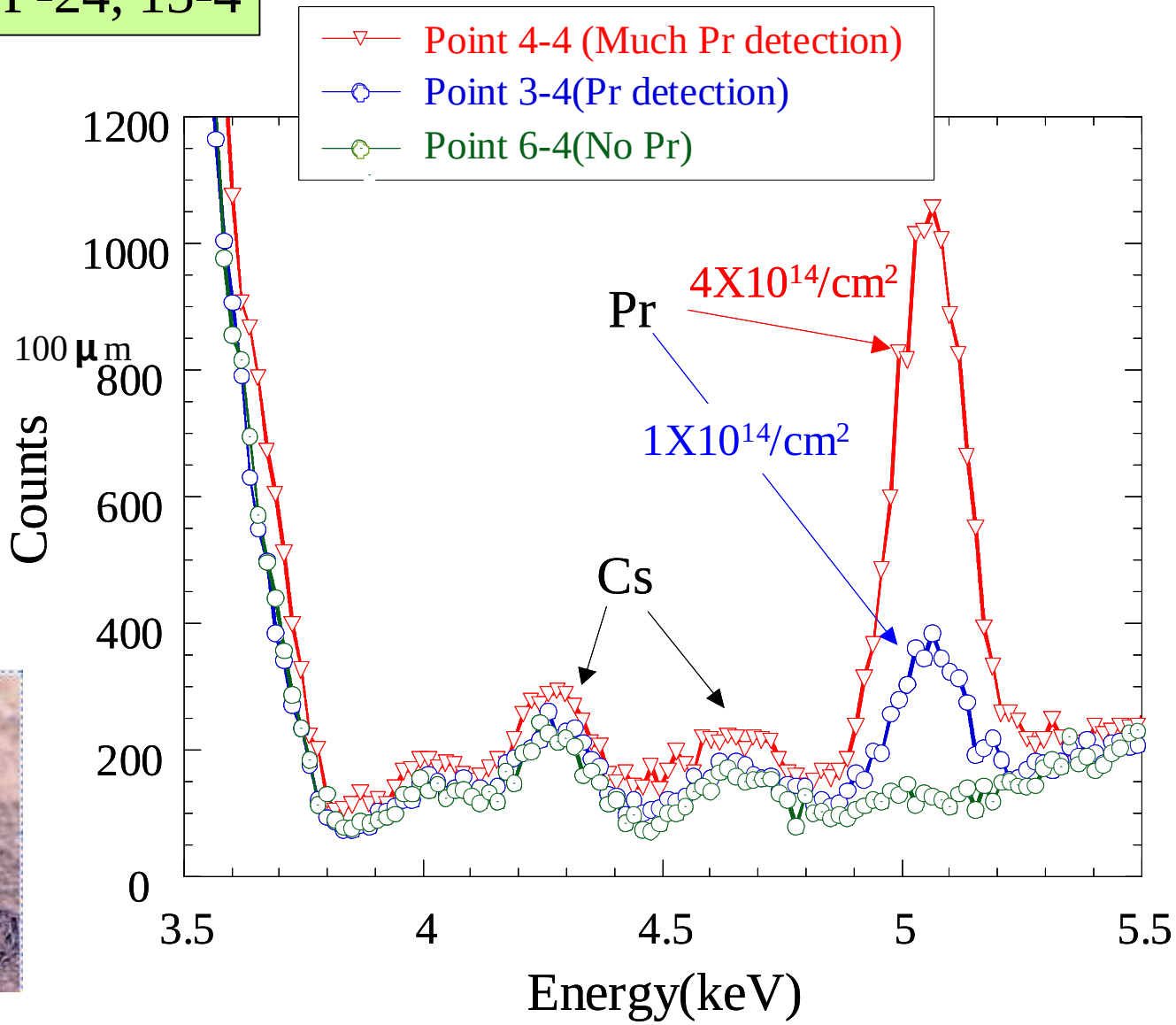
100 micron beam;SP-24, 13-4



Much Pr detection

Pr detection

No Pr



Further smaller beam analysis of Pr

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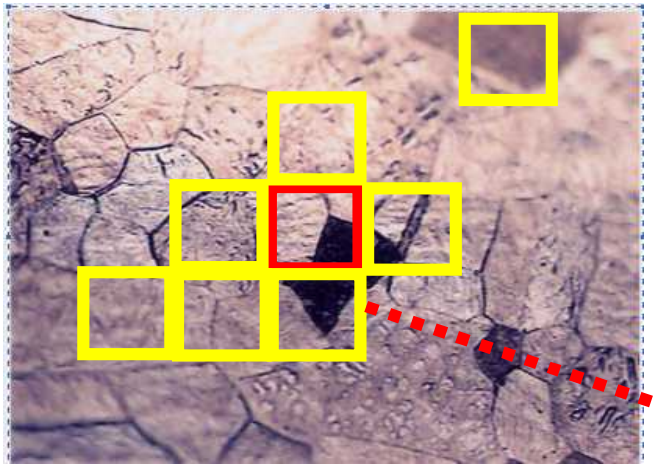
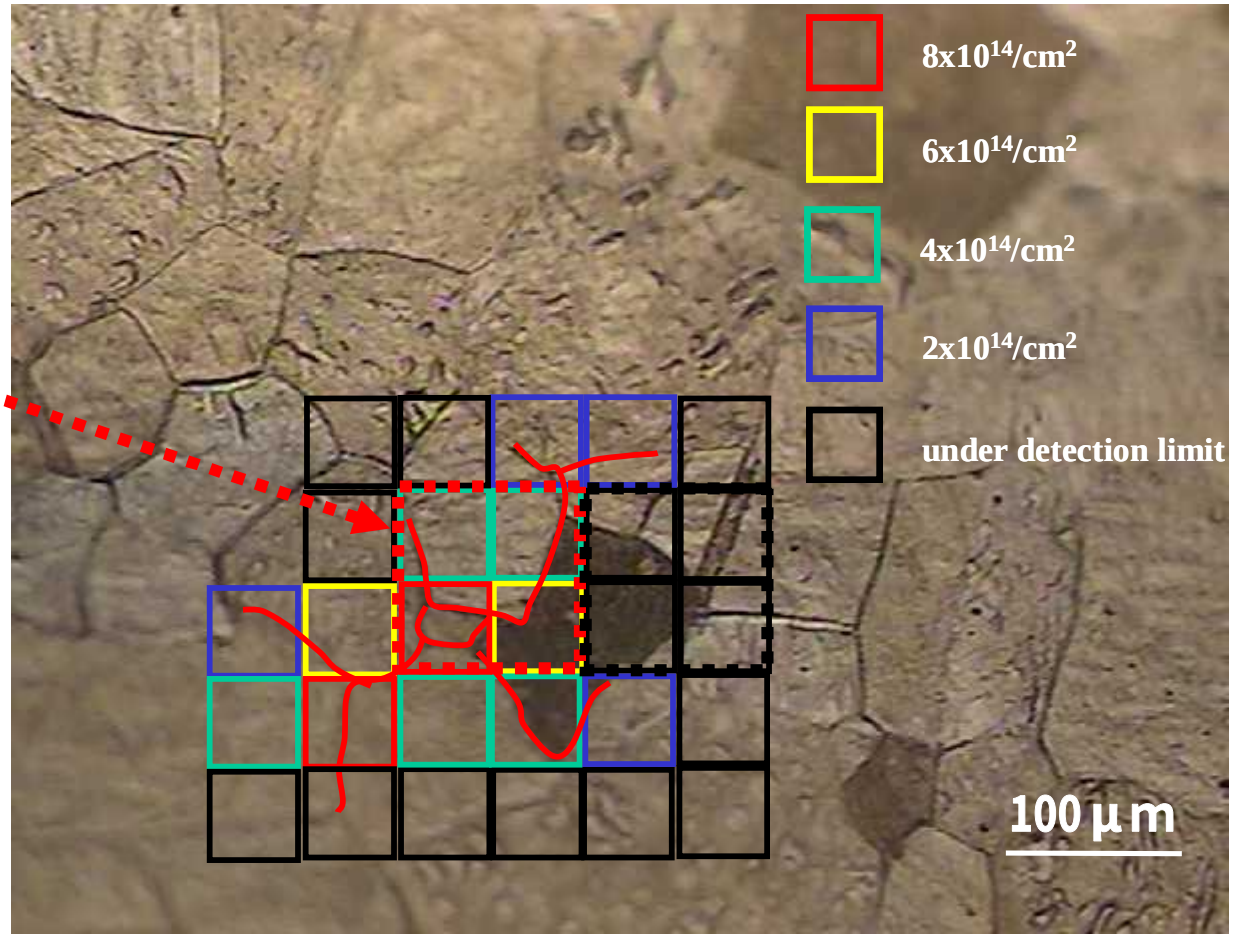
100 μm beam;SP-24



50 μm beam;SP-24



Smaller X-ray beam provides **more localized Pr distribution.**

Existence of hot spots? → **Transient High Deuterium Density?**

D^+ Ion Bombardment Experiment Performed at Tohoku Univ.

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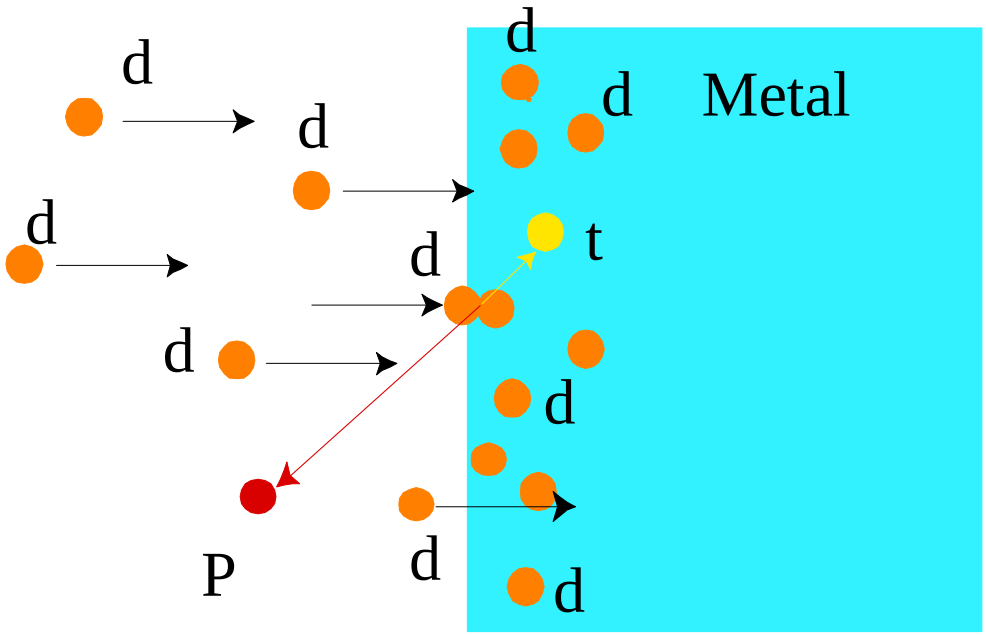
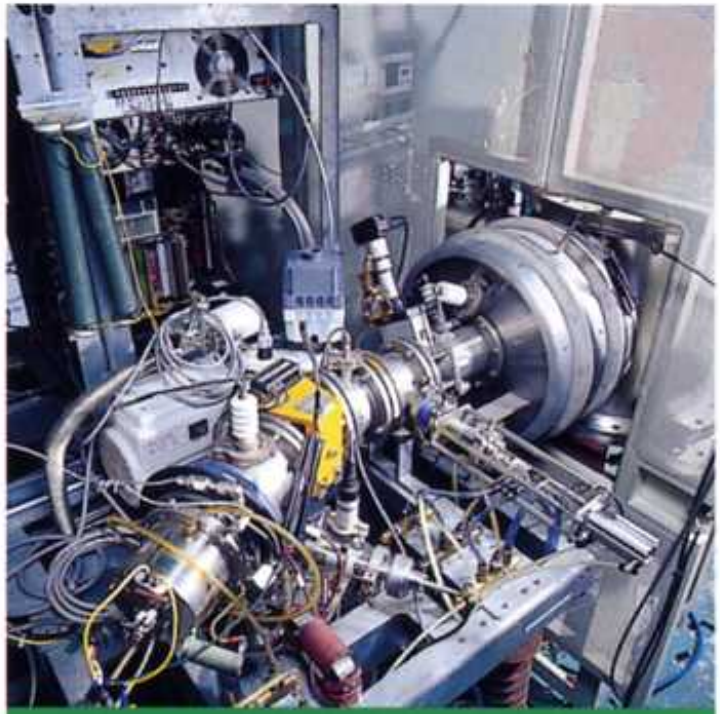
D^+ Ion beam bombardment on metal target



Experimental Apparatus



Deuterium Density measured by D^+ Ion Bombardment Experiment

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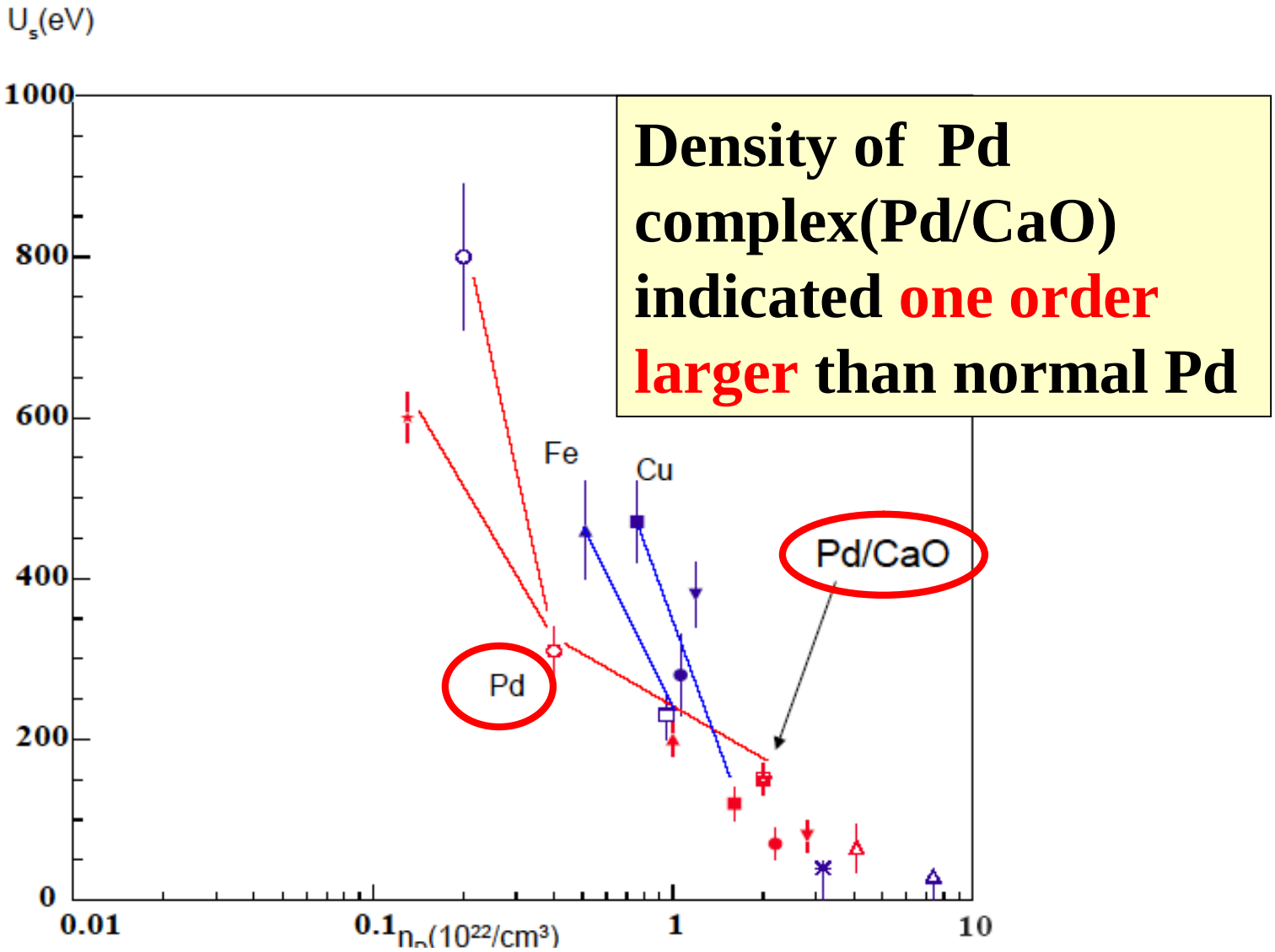

2-2 Experimental Evidences related to the importance of *Electronic Structure*

Dependence of intermediate layer

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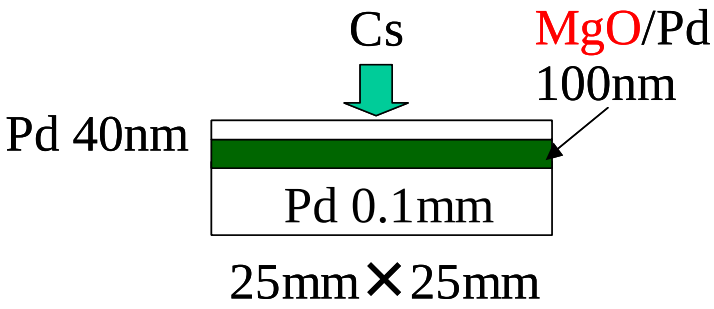
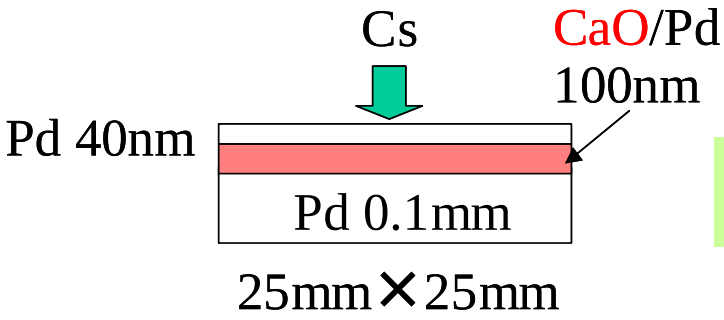


Work Function

MgO;3.3eV ⇒ NO Pr detected

3 cases



CaO;1.2eV ⇒ Pr detected !

many cases

Y_2O_3;2.2eV ⇒ Pr detected !

3 cases

Work function of the intermediate layer is important?

⇒ ***Toward the investigation of electronic structure***

Reactions observed so far in MHI

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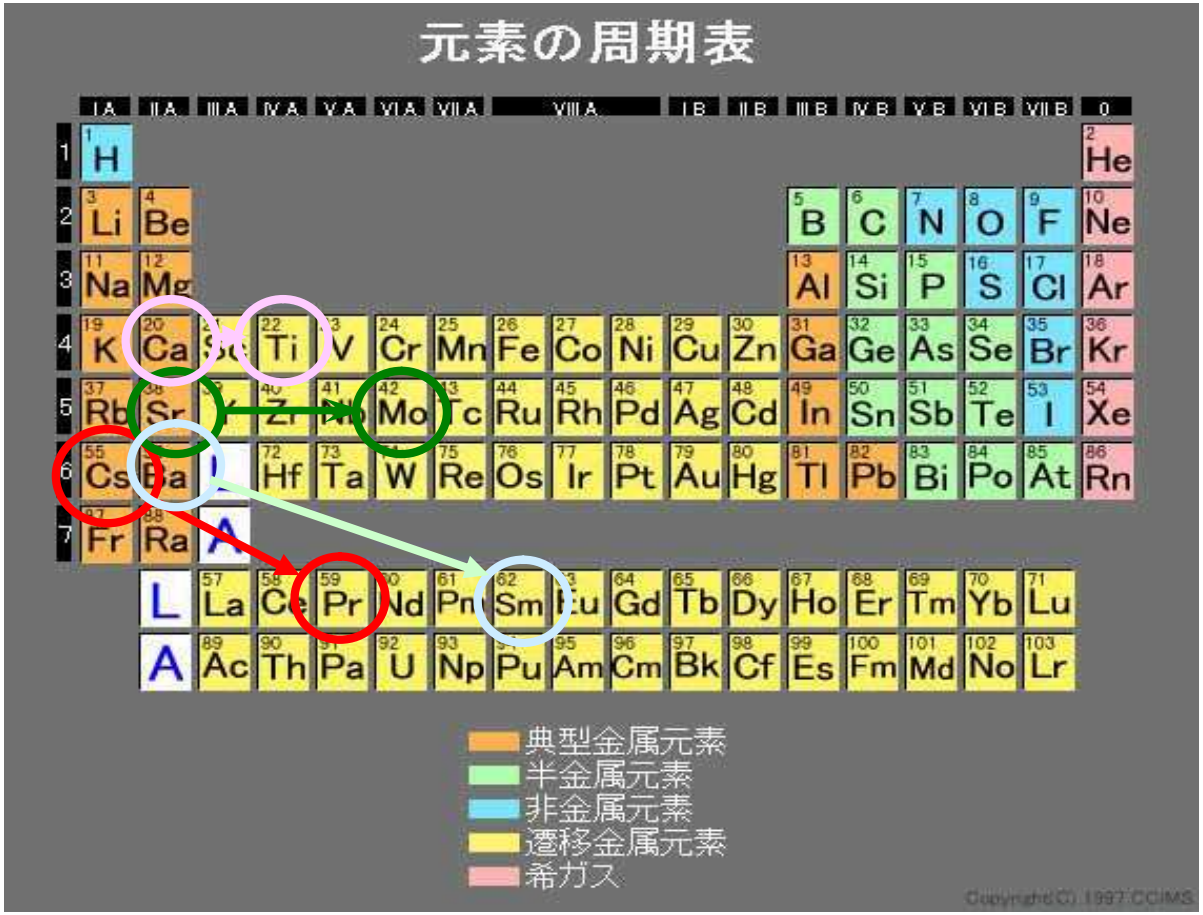

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$$^{137}_{56}Ba \xrightarrow{6d(3\alpha)} {}^{149}_{62}Sm$$

$$^{44}_{20}Ca \xrightarrow{2d(\alpha)} {}^{48}_{22}Ti$$

Electronic structure must be one of key factors.

3-1 Hydrogen density measurement using a resonant nuclear reaction

Resonant Nuclear Reaction

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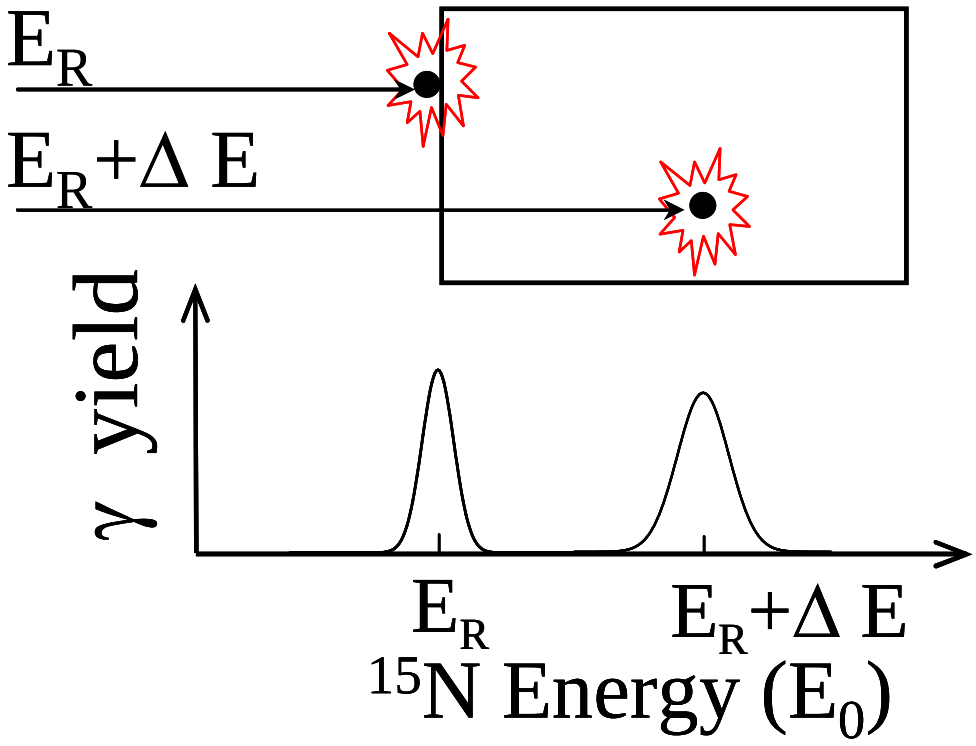
$^{15}N+^{1}H \rightarrow ^{12}C+\alpha+\gamma \quad (4.43MeV)$

$E_R=6.385MeV$



Hydrogen density distribution can be measured by 15N Ion beam

Schematic of the developed system

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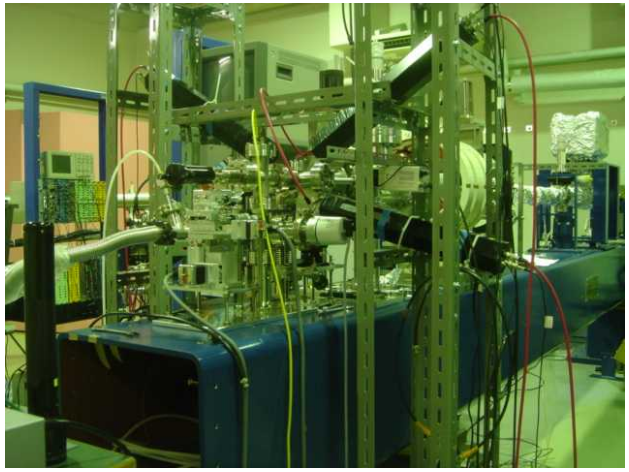
Resolution:
X-Y: 1μm
Z(depth):1nm~10nm



3-2 Hydrogen behavior during permeation

α-β phase tradition in Pd

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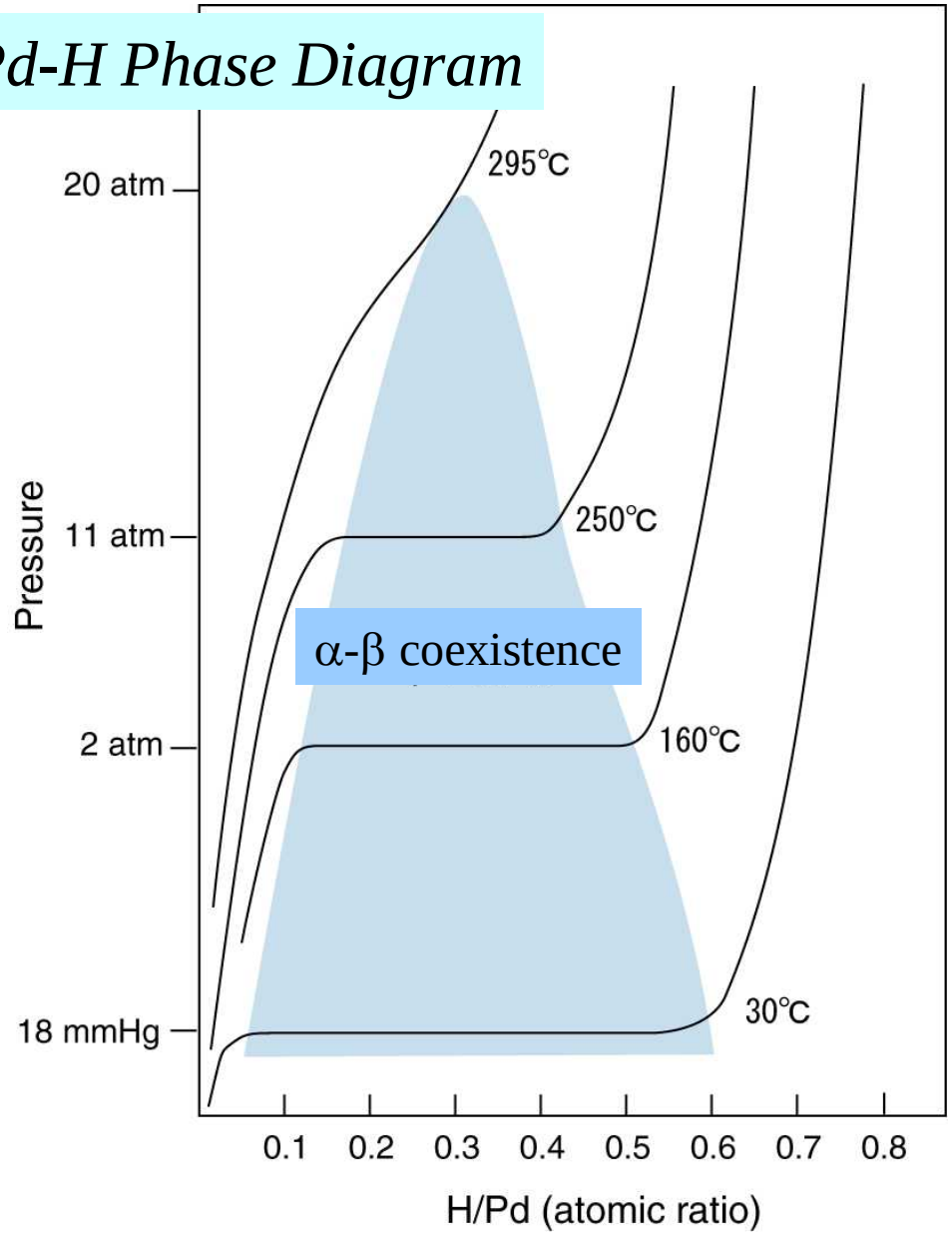
Pd-H Phase Diagram



PdH_x at RT

α-phase: x～0.06

β-phase: x～0.6

α-phase → β-phase

How is 3D distribution when hydrogen is permeating through Pd?

Experimental Set-up

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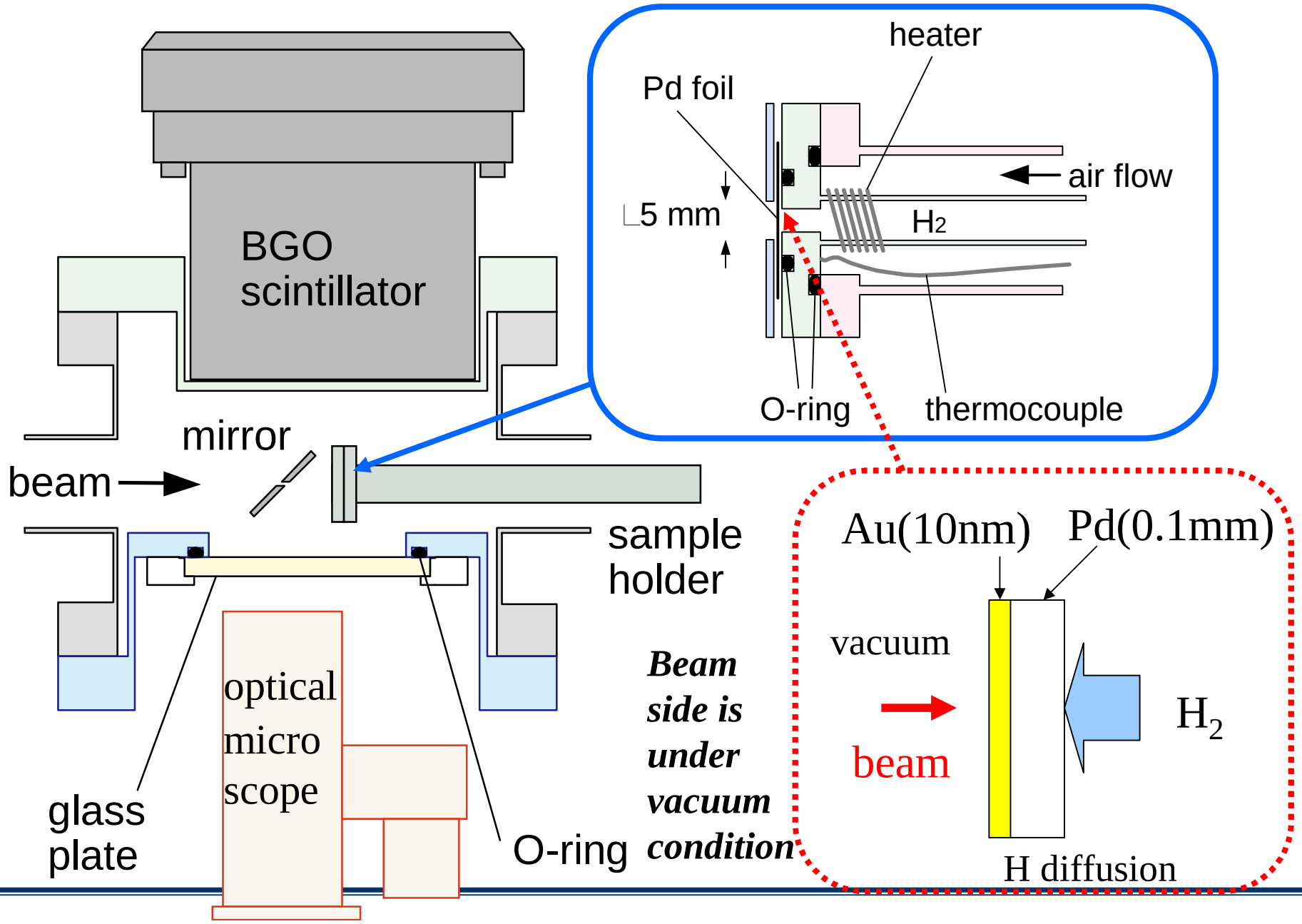

Time Dependent Site-specific NRA

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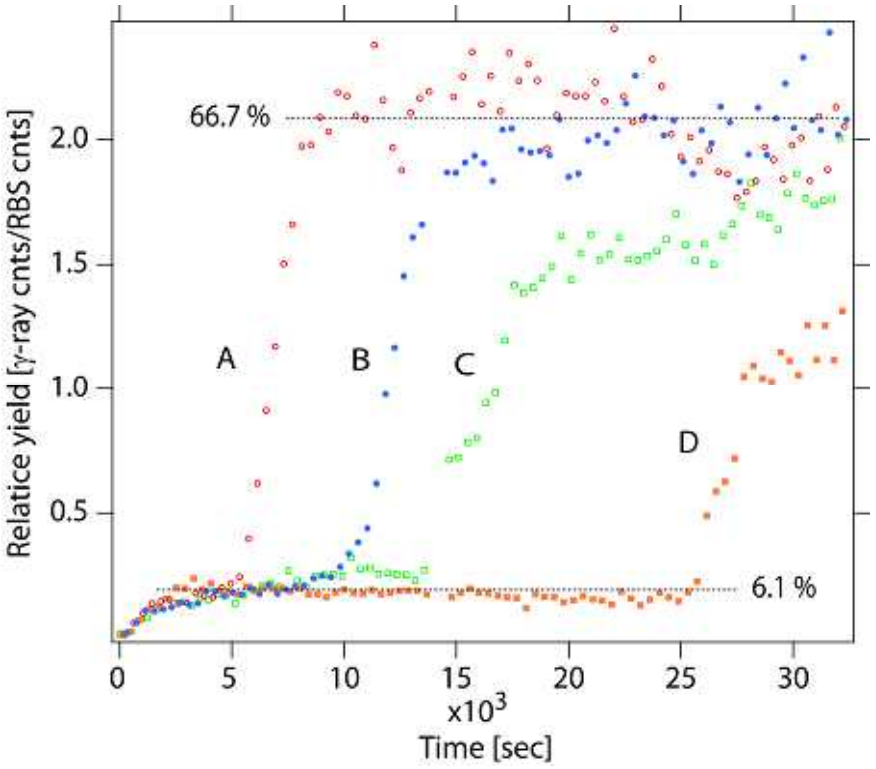
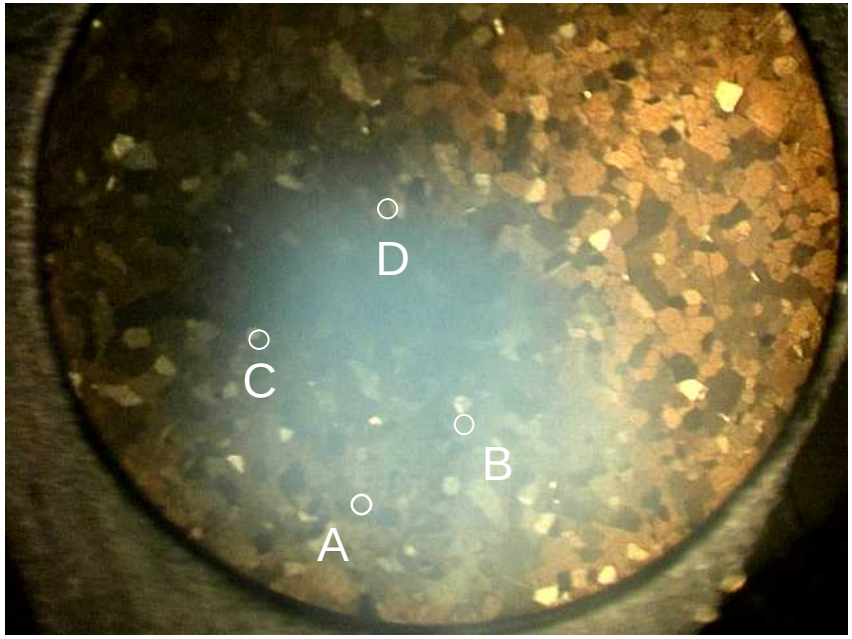
Depth ~200nm, Beam dia. <150μm, Time dependence for A~D





D. Sekiba, K. Fukutani , Y. Iwamura et al. unpublished.

To α-phase: almost simultaneously ~3,000sec
To β-phase: Seems to be **dependent on the distance from the center**

⇨ **Effect by stress or other factors?**

NRA under Normal Pressure

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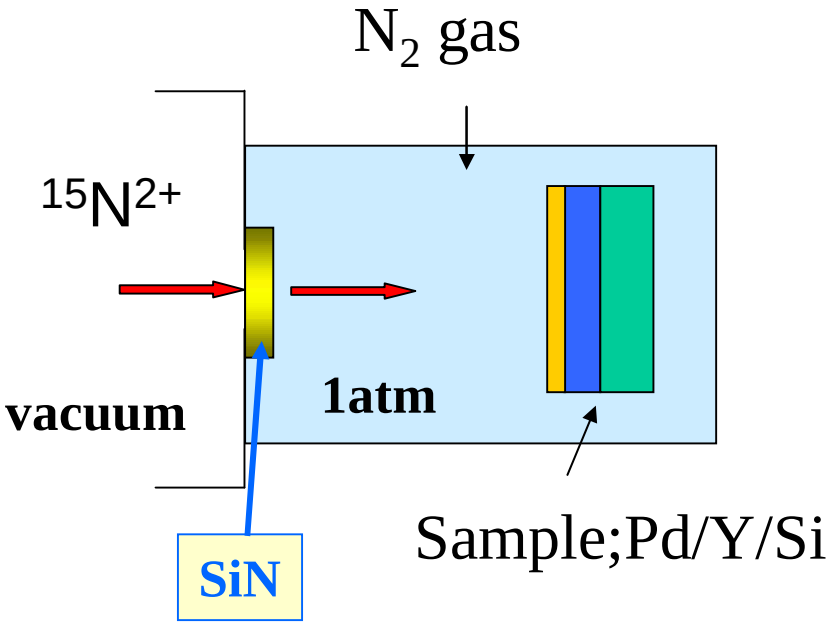
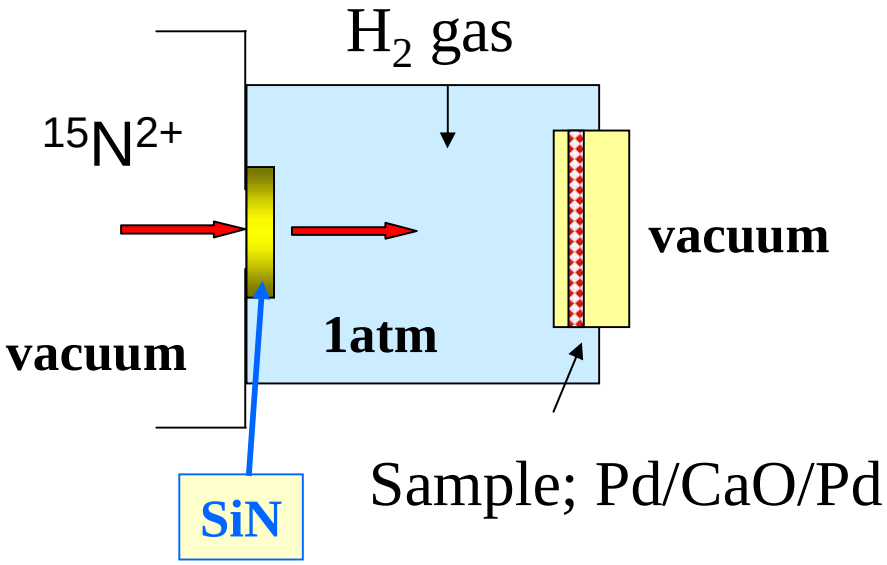


Recently, we made a success to measure hydrogen distribution under normal pressure(~1am).



Next, we will be able to measure hydrogen distribution in Pd/CaO/Pd multilayer.

3-3 Theoretical Approach to electronic structures for both targeted and transmuted elements

Assumption

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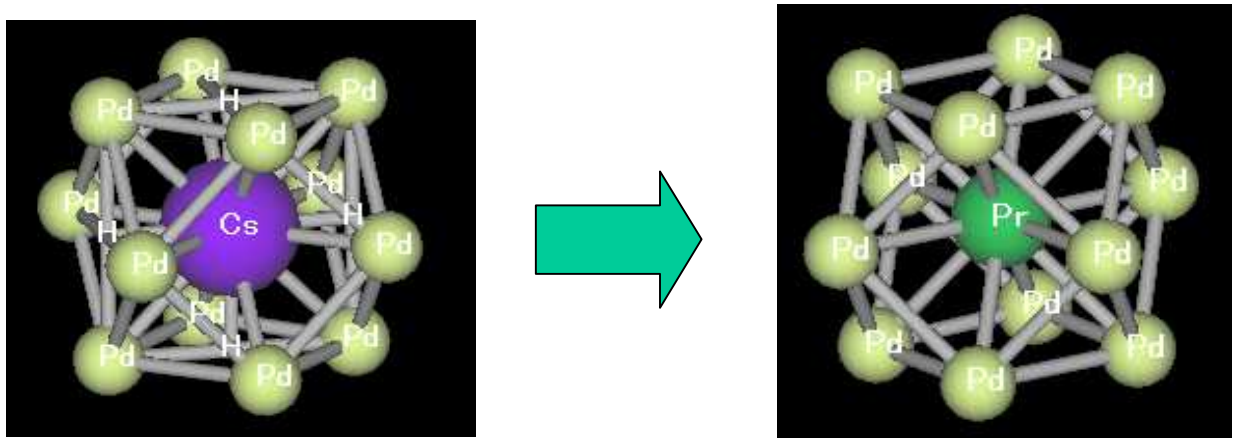
A Characteristic Electronic Structure might be found for both targeted and transmuted elements.

For example,

Electronic Structure



Cs:4H(close to Cs) in Pd	≈	Pr in Pd
Initial state	≈	Final state

Method of Calculation

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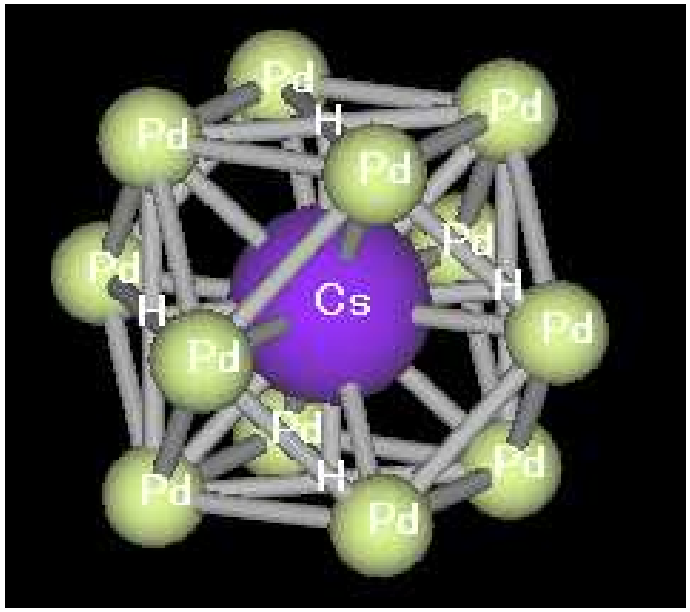
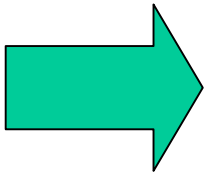
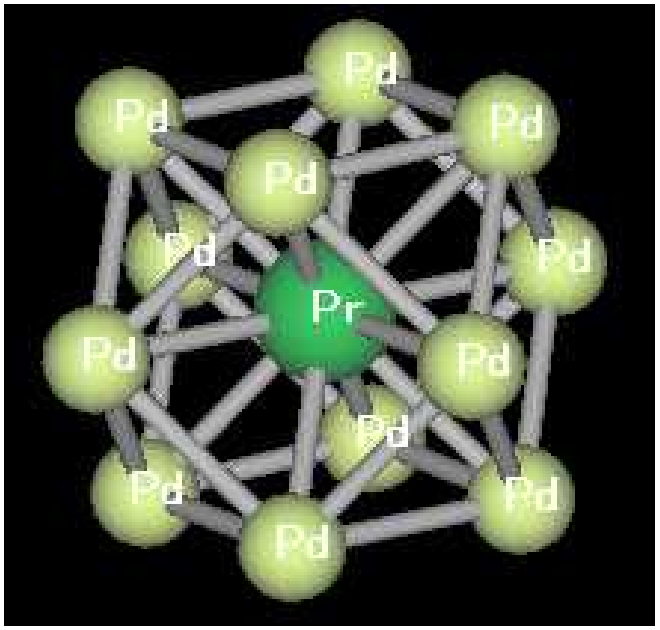
Software **ADF**(Amsterdam Density Functional software)
Density-functional scheme







(Cs+4H):12Pd Cluster

Pr:12Pd Cluster

Energy Levels for Cs, Pd and Pr

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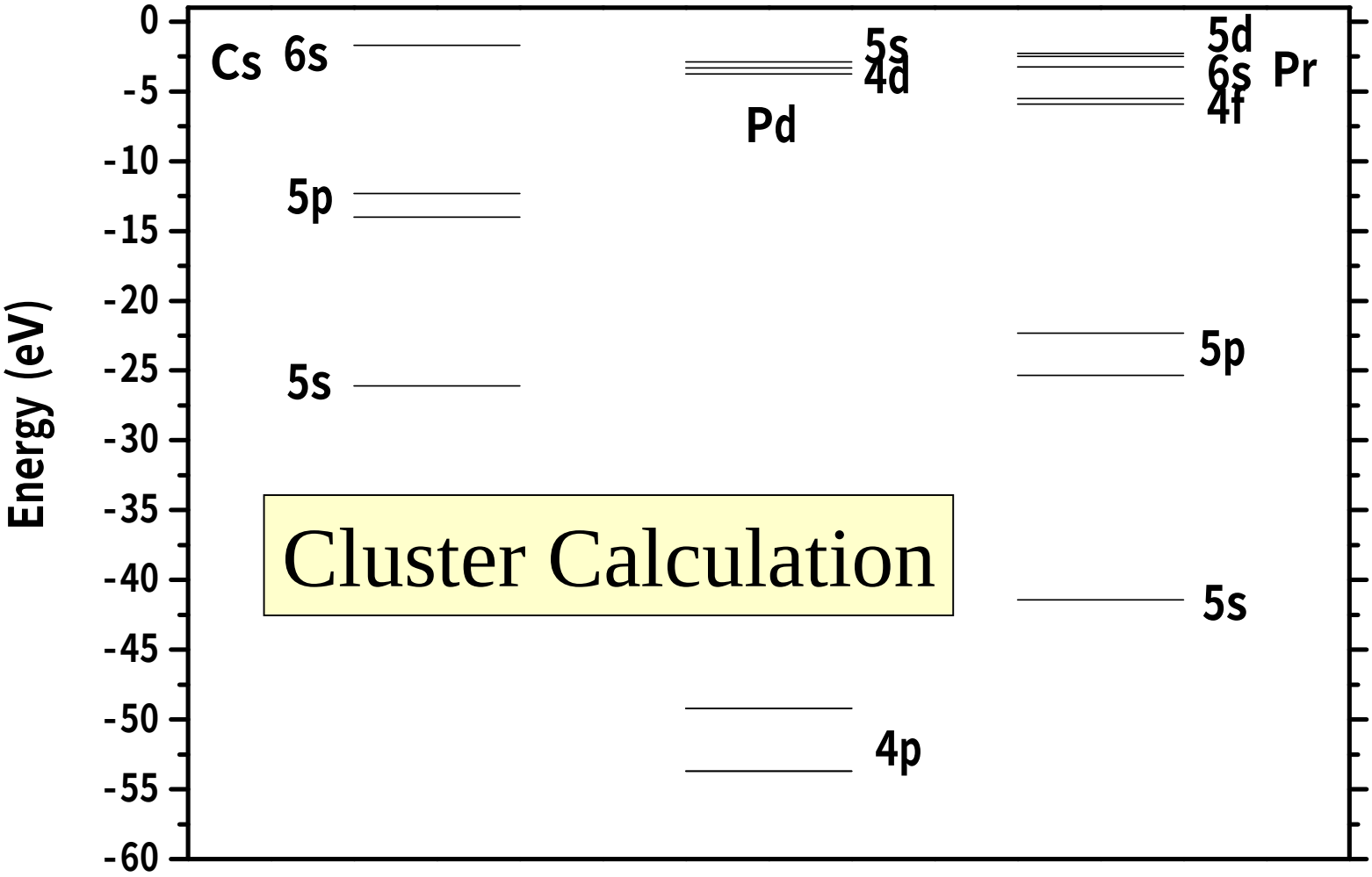

Agree with experimental values

Change of Total Electronic Energy

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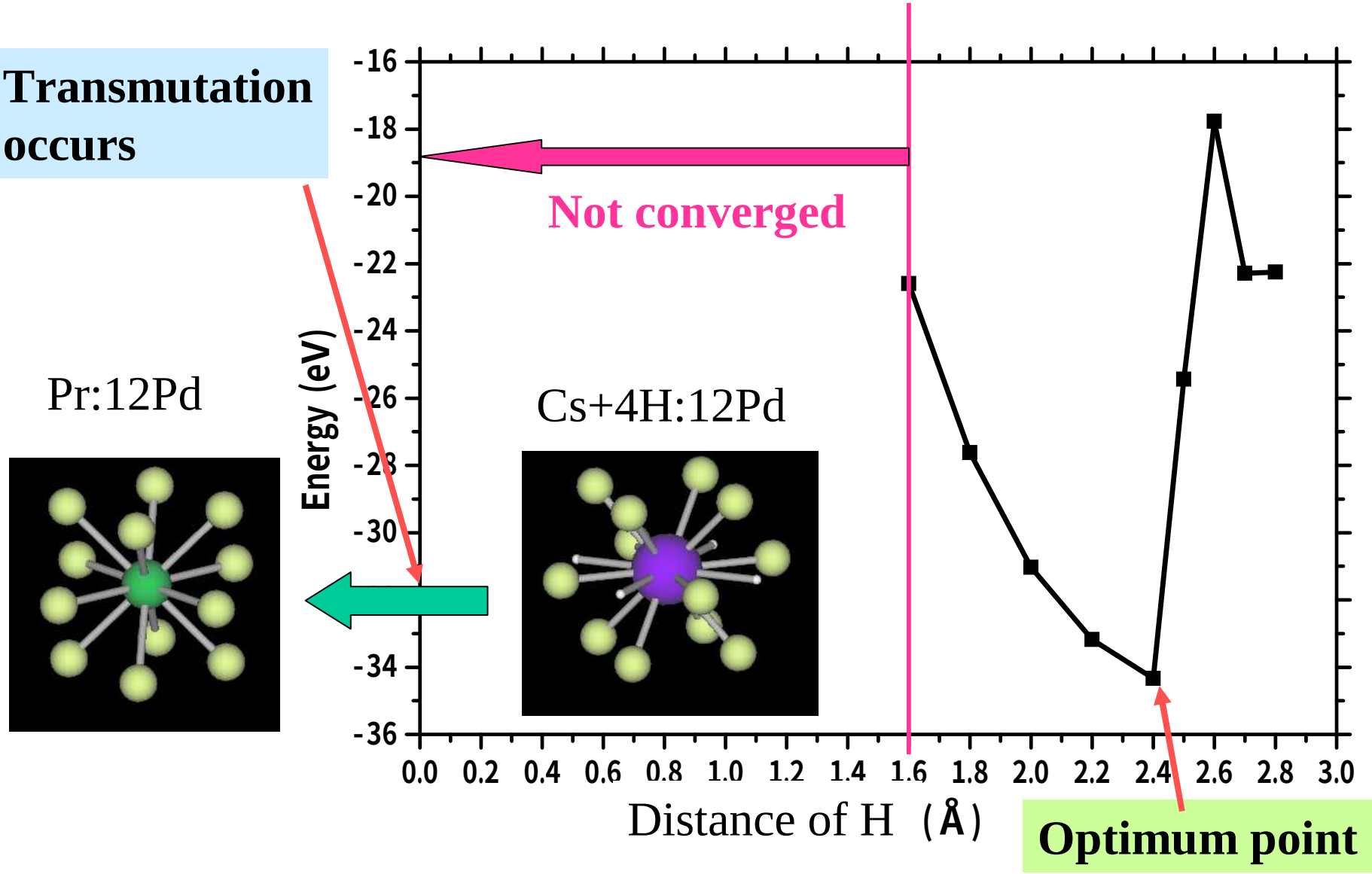
Transmutation occurs



Change of Electronic Structure

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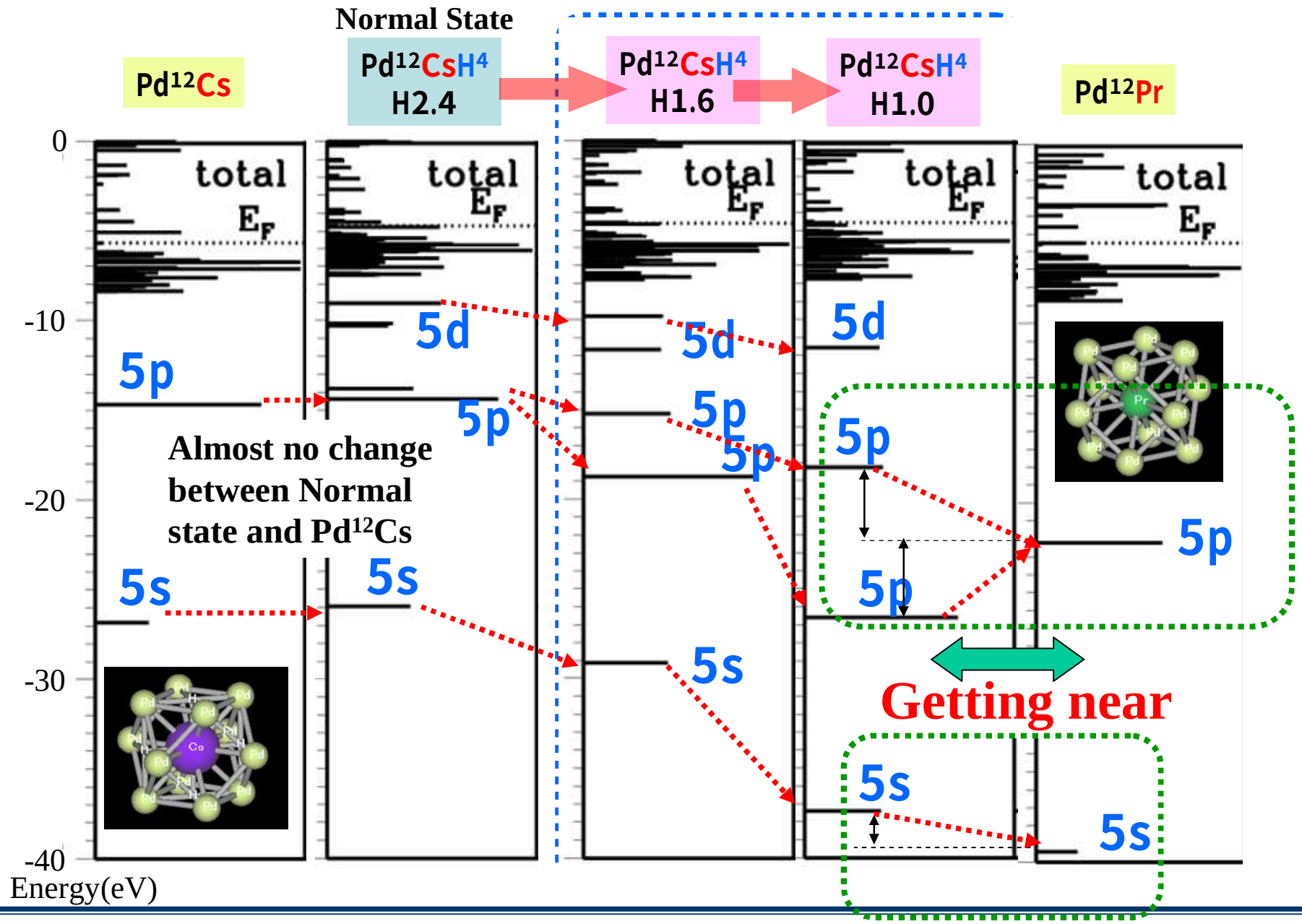

Concluding Remarks

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1. **Low energy nuclear transmutations from Cs into Pr, Sr into Mo, Ba into Sm and Ca into Ti have been observed in the Pd complexes, which are composed of Pd and CaO thin film and Pd substrate, induced by D2 gas permeation.**
2. **Local Deuteron Density and Electronic Structure in the Pd multilayer seems to be one of the essential factors that govern this phenomenon.**
3. **Using A micro-beam NRA system, position dependence for phase transition in Pd sample was observed. Phase transition from α to β seemed to be dependent on the distance from the center; it might be correlated with inner stress in the Pd. *Local deuteron density measurement***
4. **We have started first principal calculation for this phenomenon based on the assumption that a characteristic electronic structure might be found for transmuted elements. *Electronic Structure Calculation***

Acknowledgements

I would like acknowledge
Prof. A.Takahashi,
Prof. H. Yamada,
Prof. S. Narita
for their supports and valuable discussions.

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