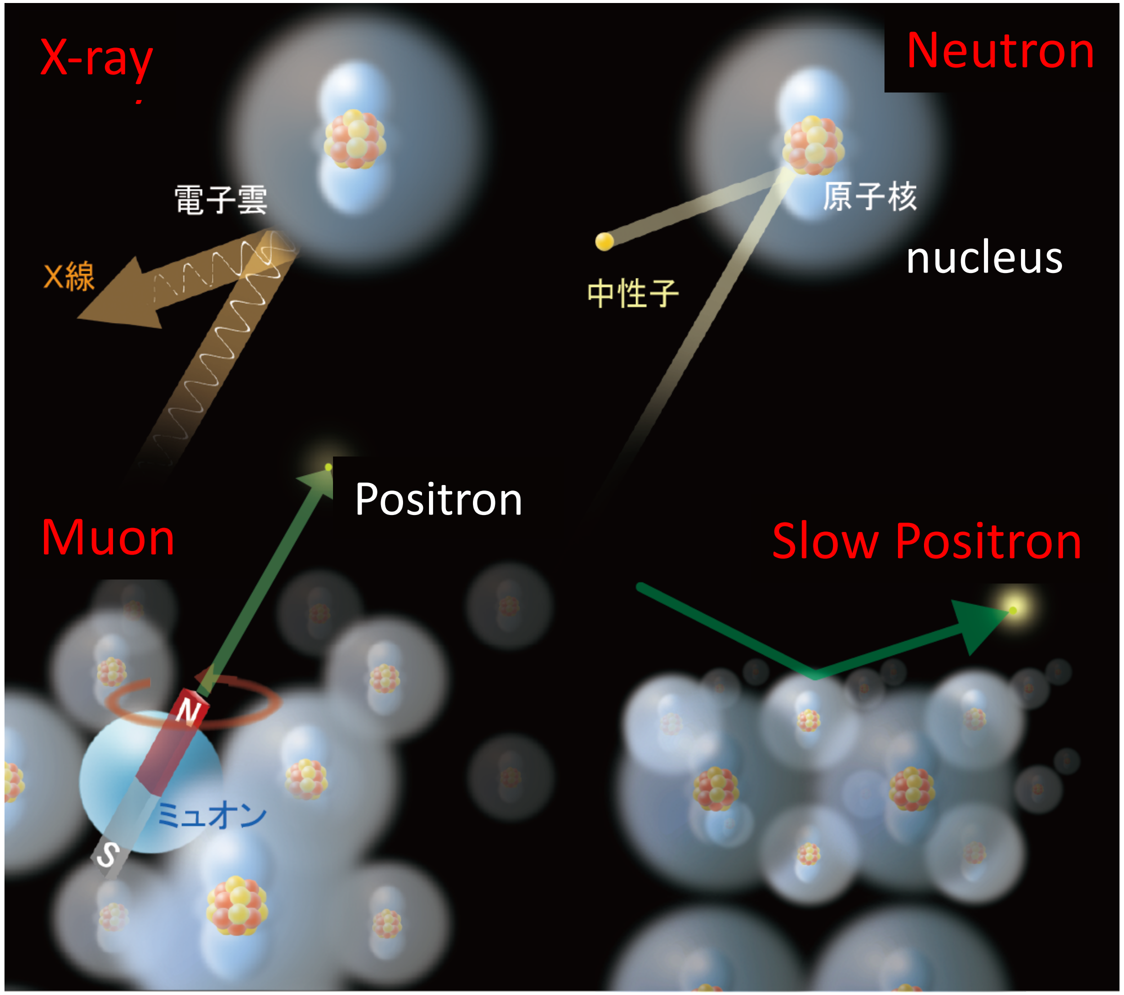
Materials science at KEK - Neutron Scattering-

Toshiya Otomo
Institute of Materials Structure Science, KEK

Institute of Materials Structure Science

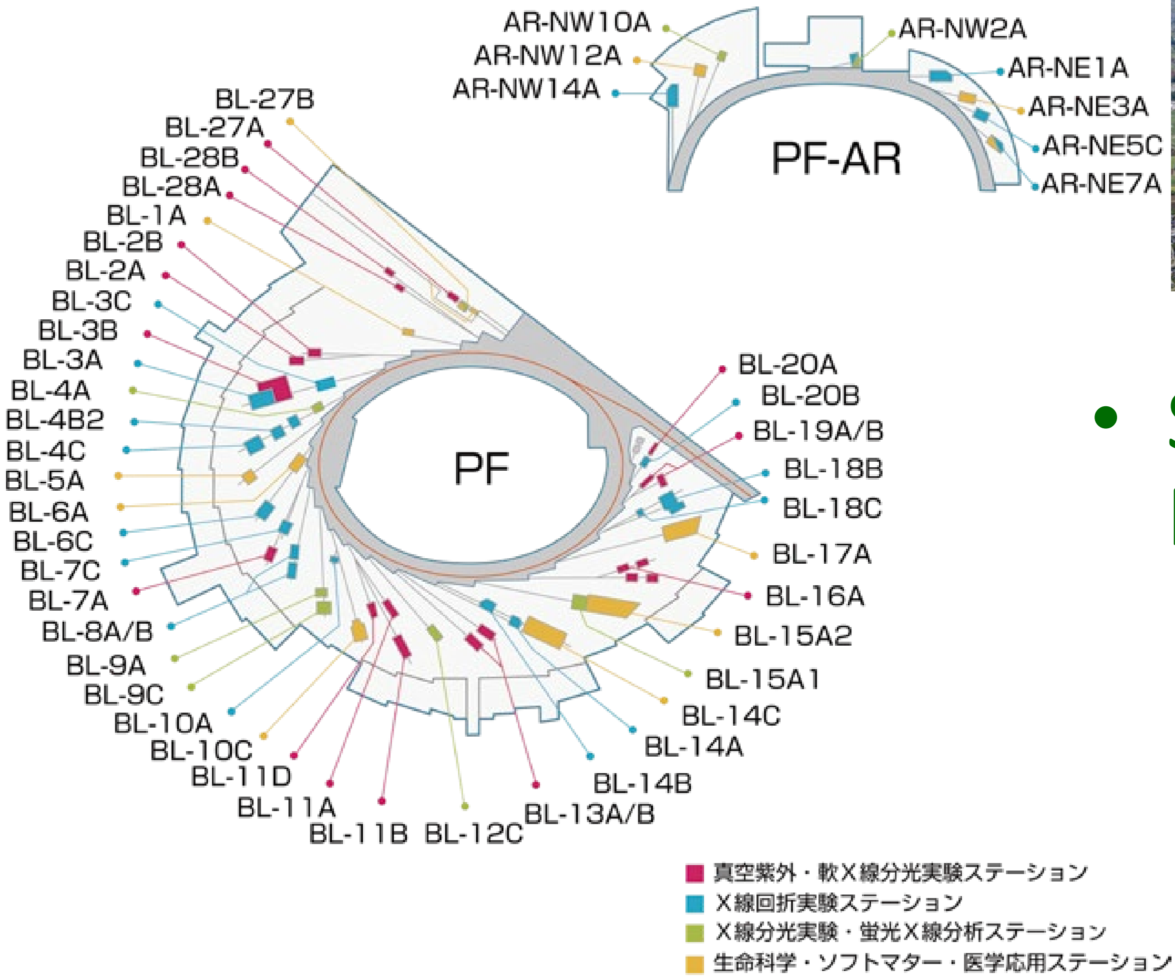
- Researches of the structure and functionality of materials using accelerators.
  - Synchrotron radiation, neutrons, muons and slow positrons
  - investigate a wide range of materials from those at the atomic level to polymers and biomolecules.

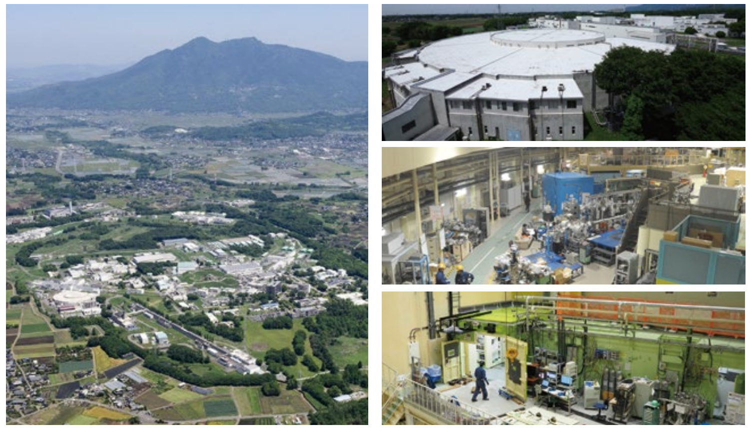
“Quantum Beams” of IMSS



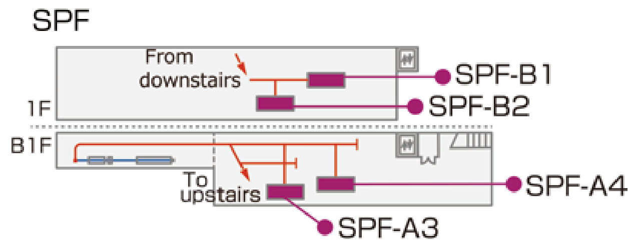
Institute of Materials Structure Science (Tsukuba)

- Synchrotron Radiation Facility (Photon Factory, PF)



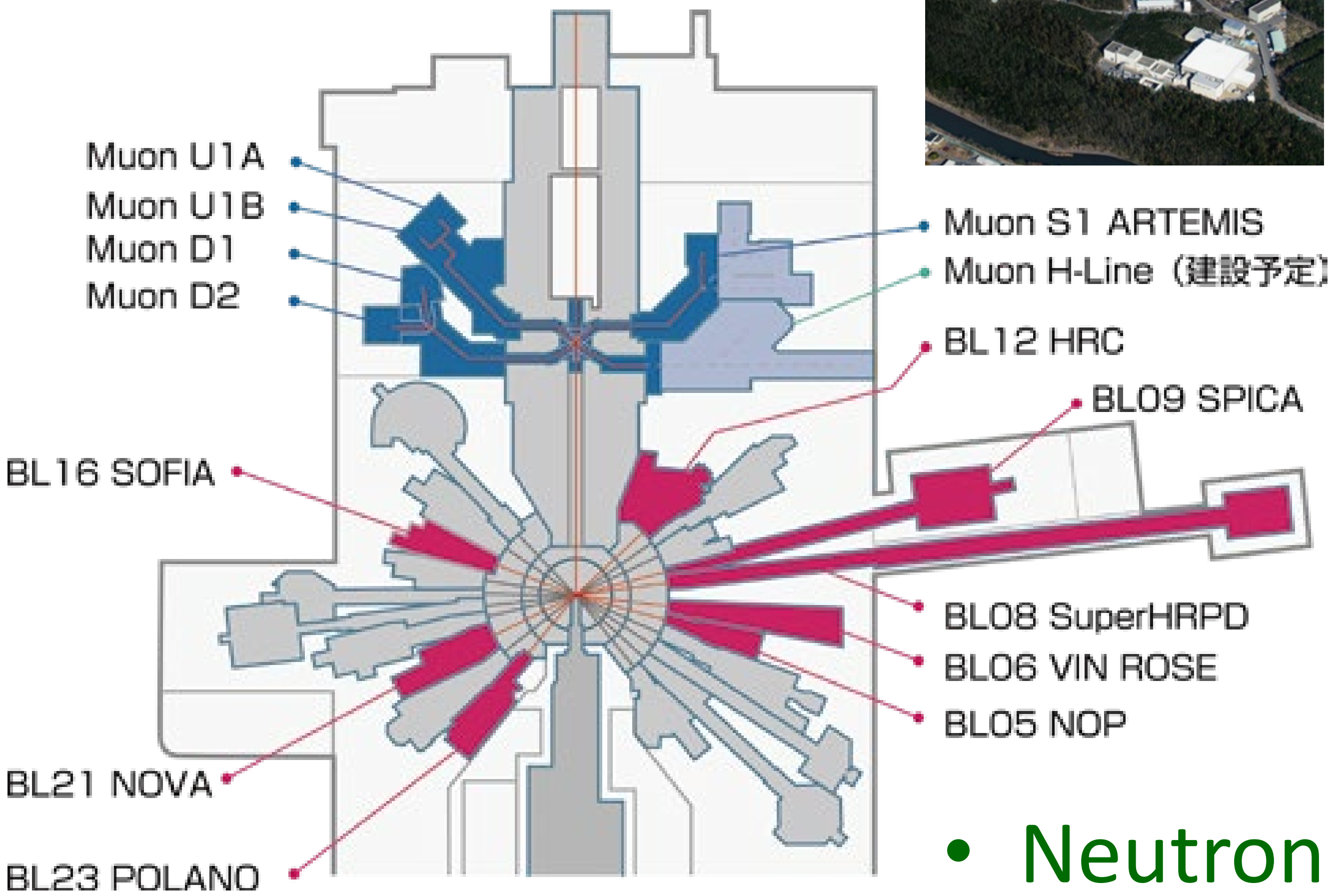


- Slow Positron Facility (SPF)



Institute of Materials Structure Science (Tokai)

- Muon facility

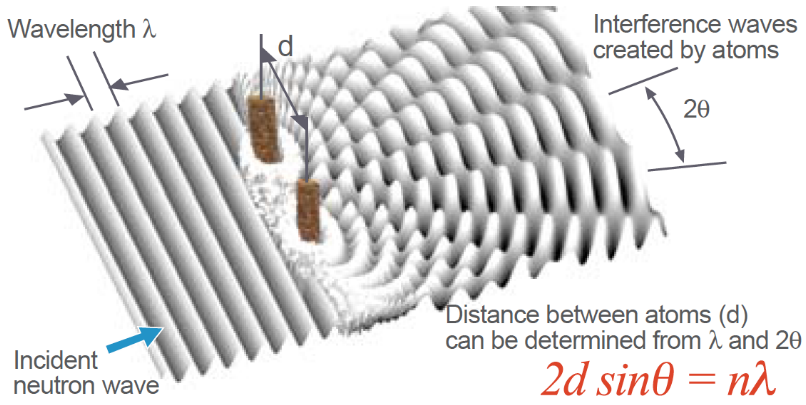


- Neutron facility

Neutron

mass : 1.67 x 10^{-24} g（≈ proton）
charge : zero
spin : 1/2

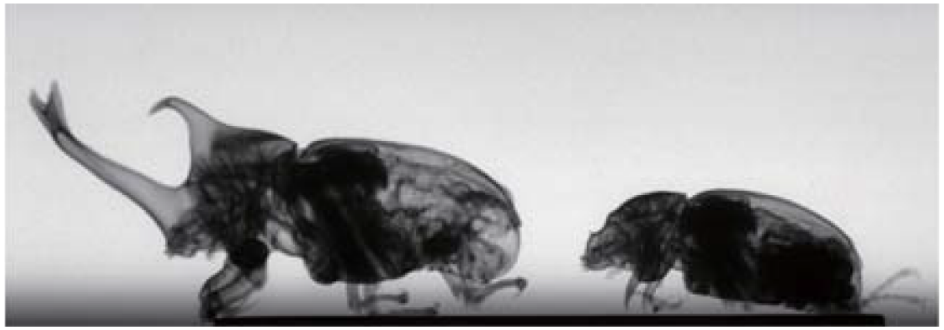
Observation of atomic structure

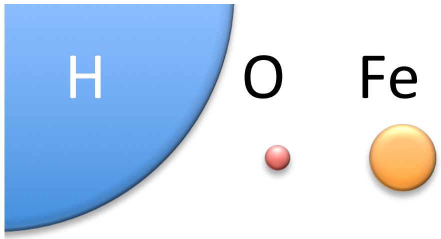


Observation of dynamics

Microscopic motions in a proton

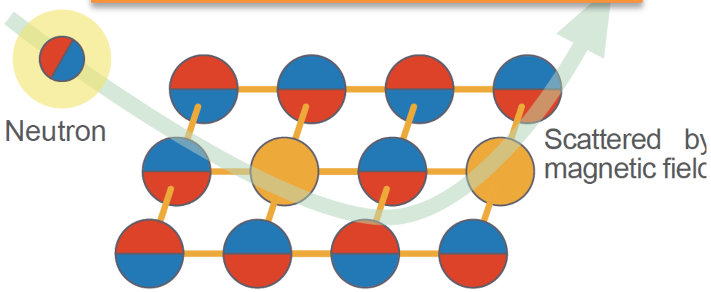
High transmittance





Sensitivities for light elements

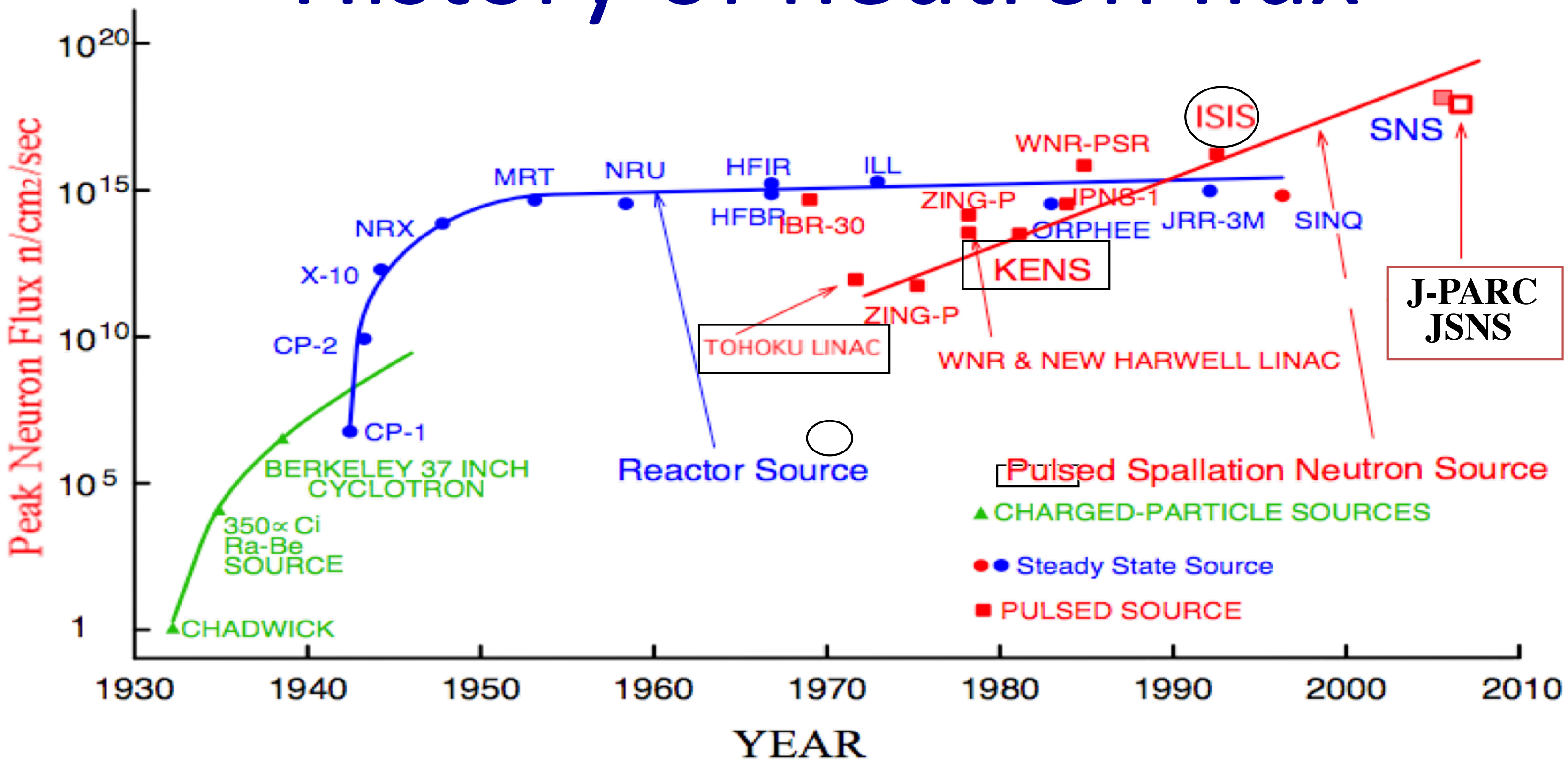
Observation of magnetic structure



J-PARC


LINAC
Rapid Cycle
Synchrotron
Neutrino
Expe.Facility
Materials & Life
Science Exp.
Facility (MLF)
Main Ring
Hadron exp.
Facility

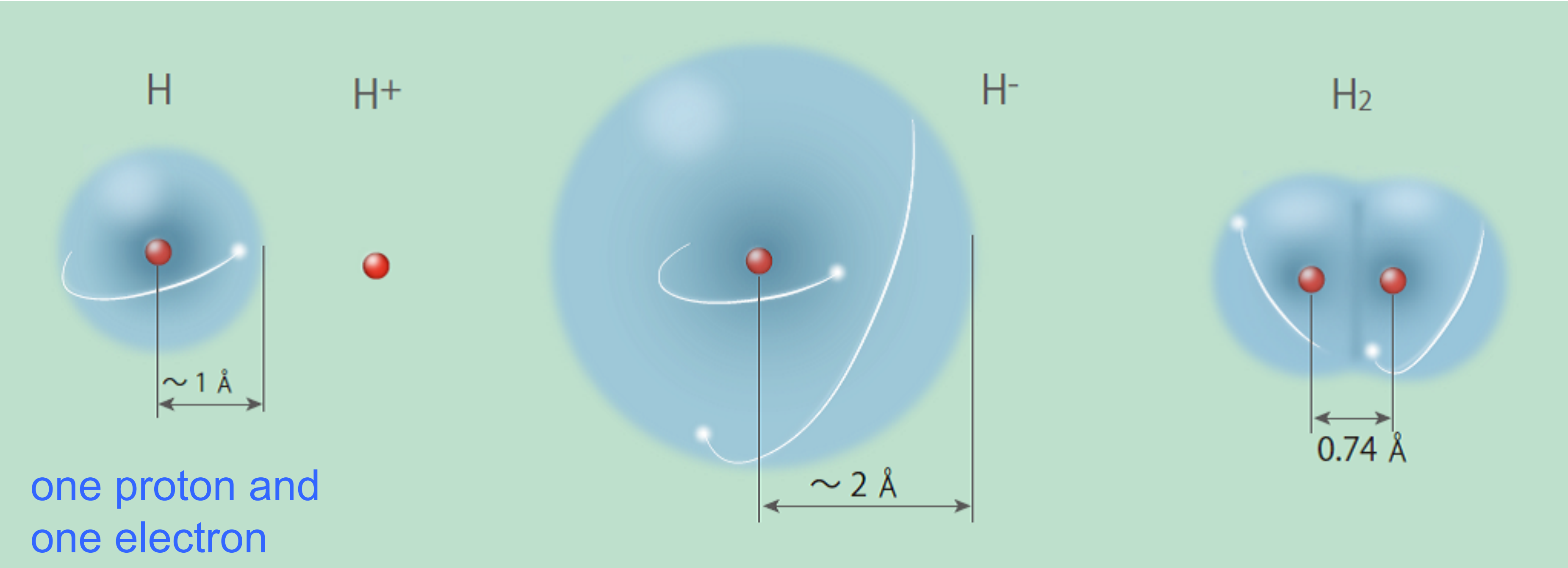
History of neutron flux



- China: CSNS project
- EU: ESS project
  - 5 MW

- Japan has long history of pulsed neutron source

Hydrogen can be cation and anion



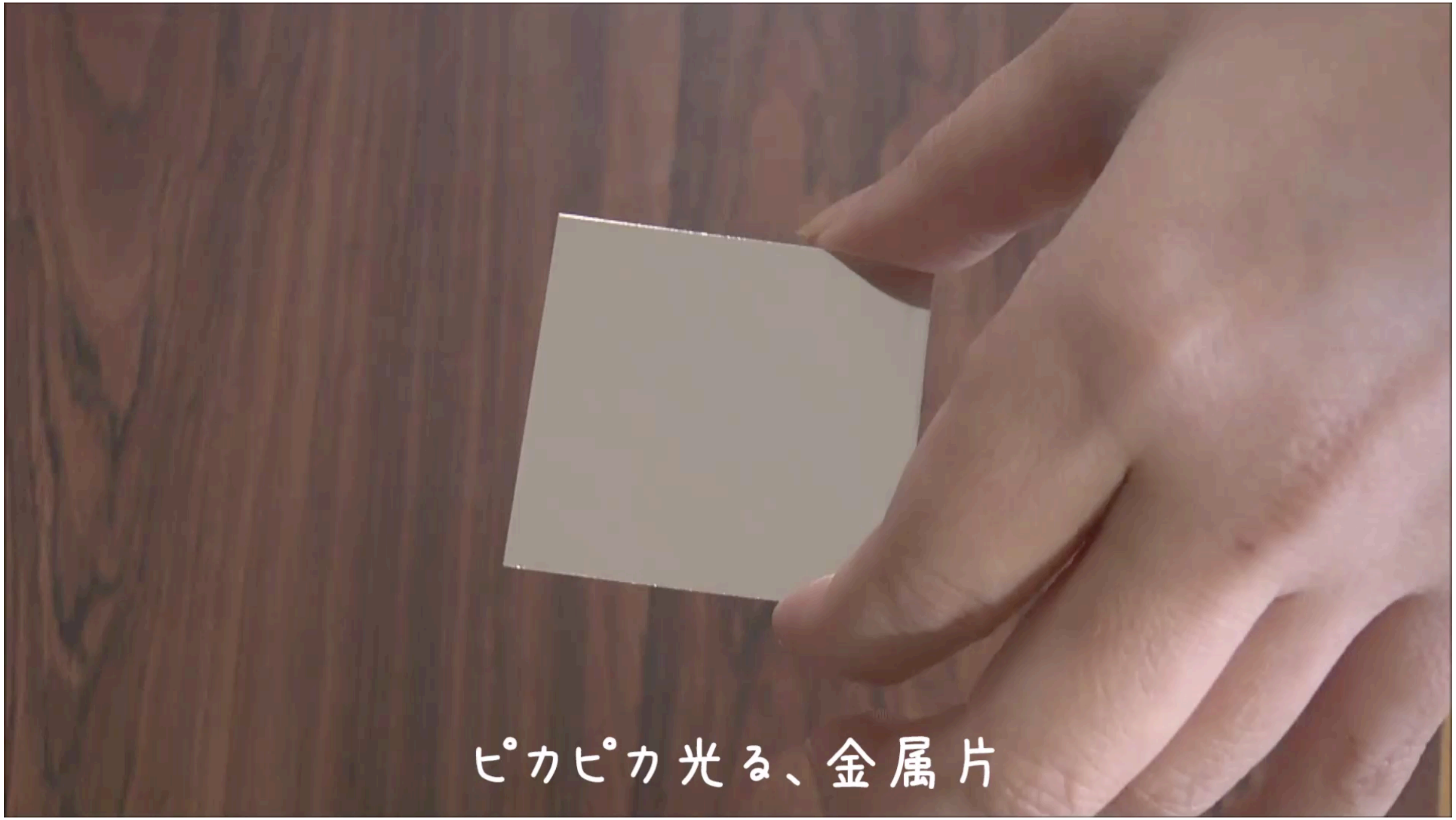
✓About 90% of
elements in universe

✓ About 63% of
elements in human body

Hydrogen

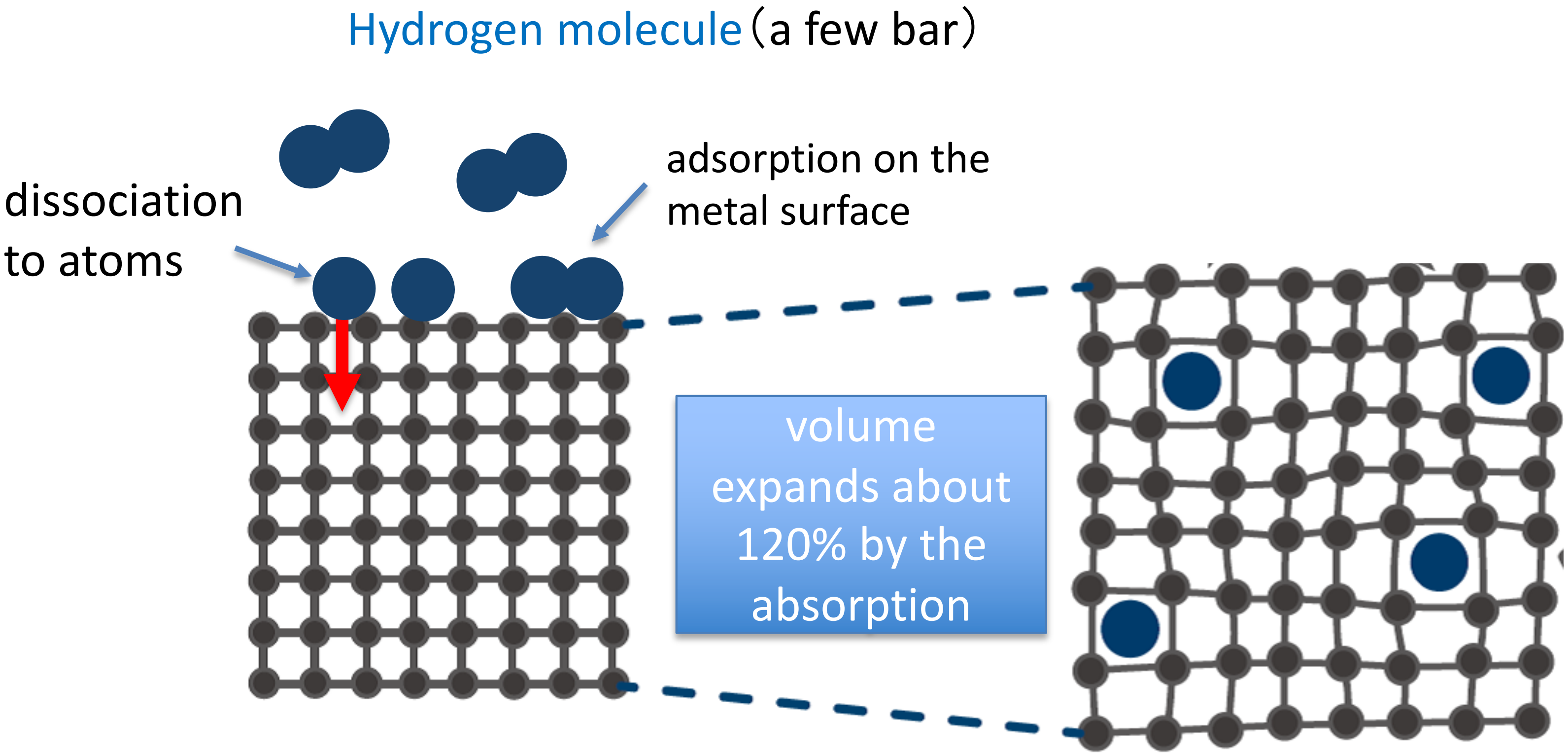
- Changes materials function
  - Hydrogen can be an electron donor and acceptor
- Key element of renewable energy
  - Hydrogen storage materials
  - High Tc superconducting materials
- Key element of our life
  - Hydrogen bond
  - H2O
- Most difficult element (atom) to observe
  - only one electron: difficult to detect with X-ray
  - neutron is the most powerful tool for the direct observation

What's happen if metals absorb hydrogen



sample provided by Atsumitec Co. Ltd.

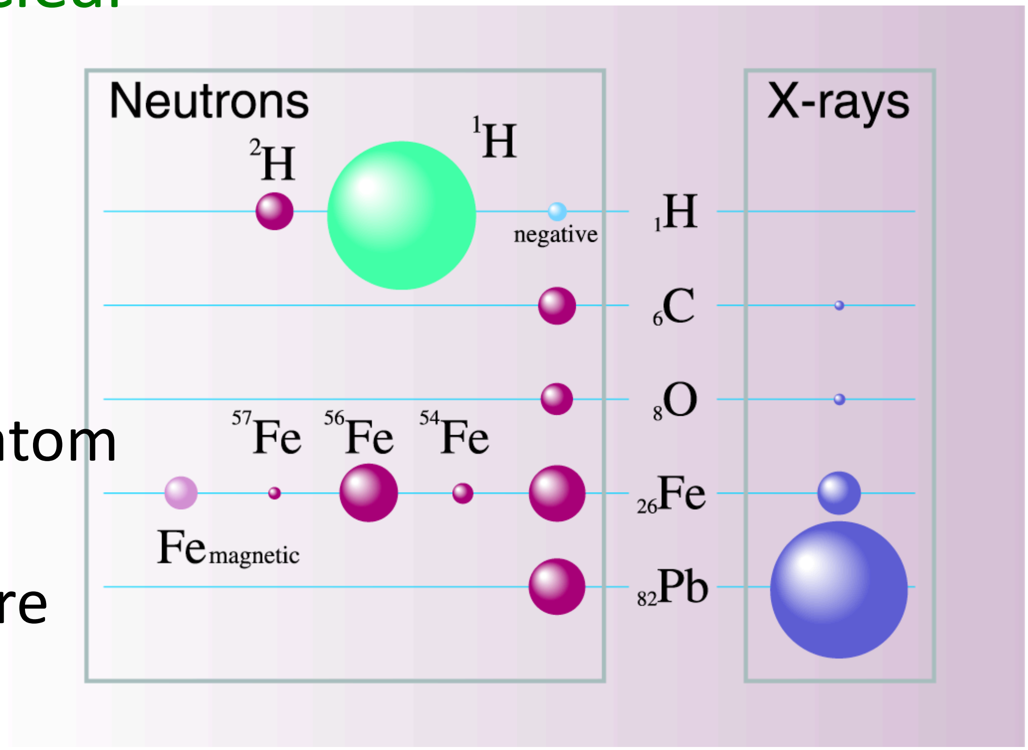
Hydrogen absorption



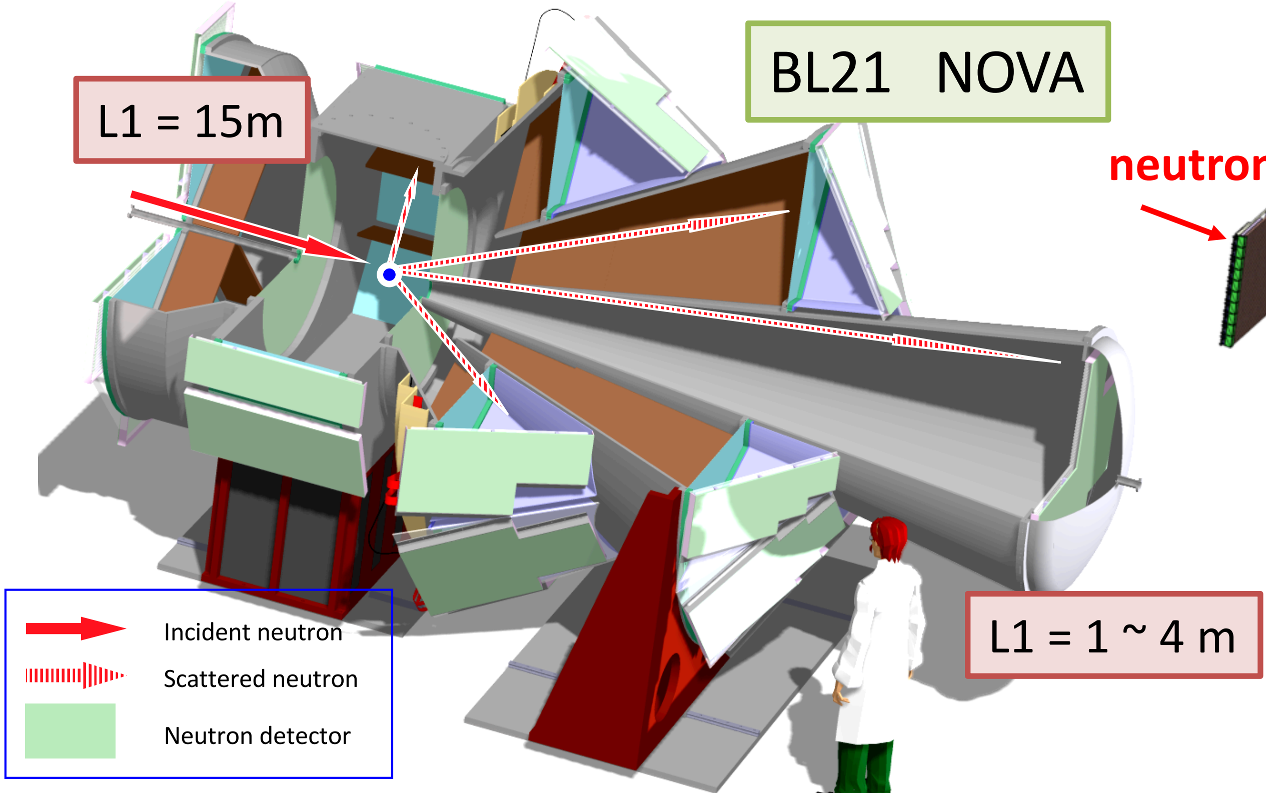
almost same density as liquid can be realized at room temp.

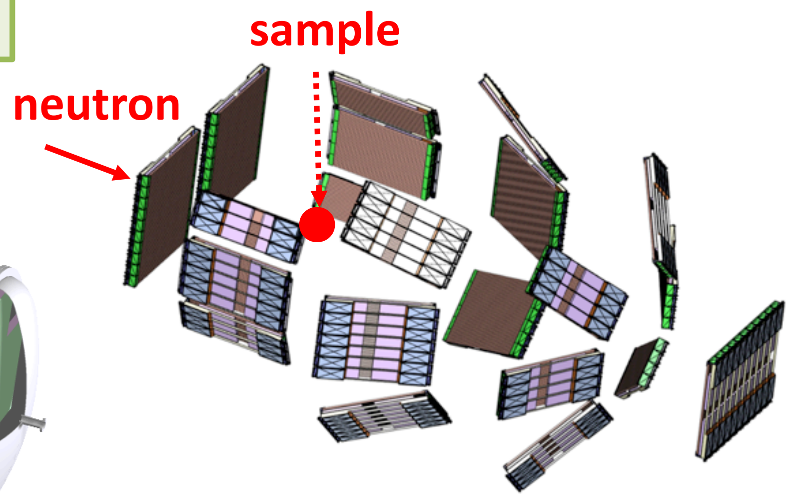
Neutron & Hydrogen

- Neutron scattered by nuclear
  - proton (H^+) is observable
- Large incoherent cross-section of ^1H
  - advantage to individual atom movement observation
  - disadv. to atomic structure observation -> use 2H
  - but often isotope effect between 1H and 2H is large



Neutron total scattering instrument





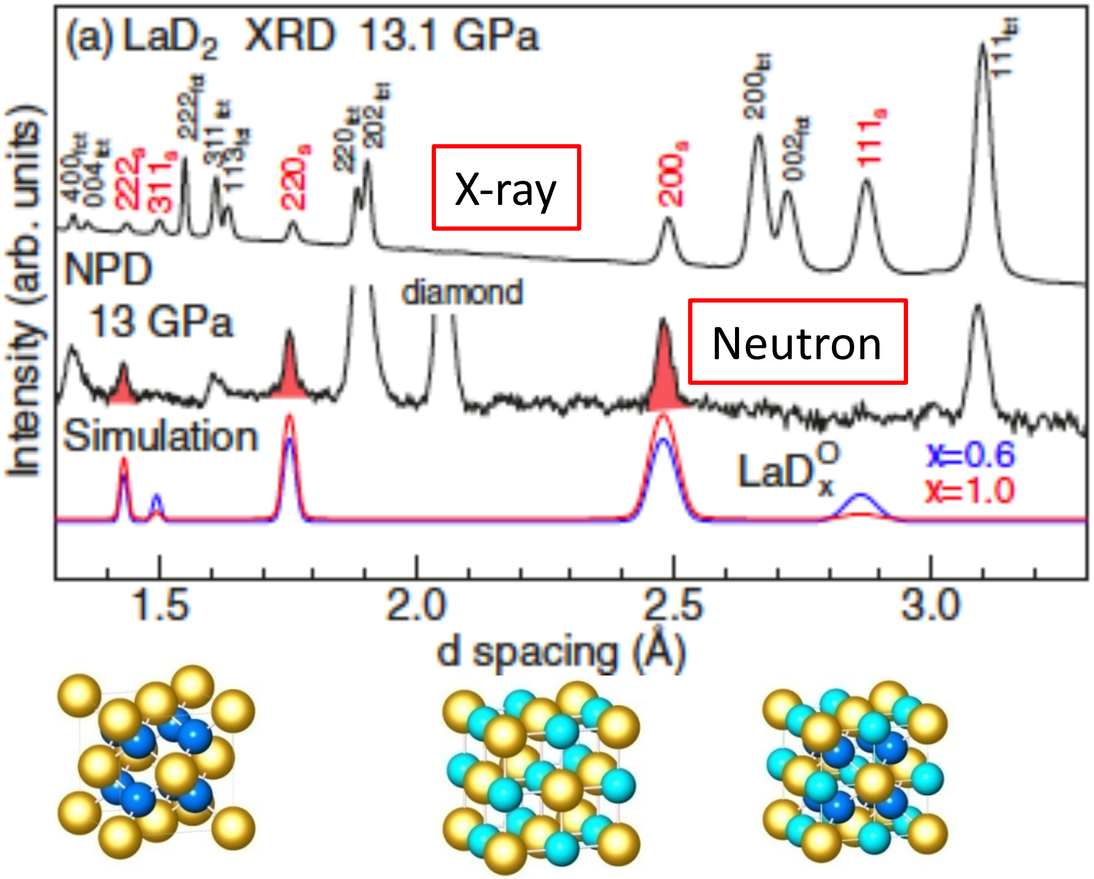
^{3}He PSD 20atm ½ inch 800mm

- Wide-Q measurement including small angle scattering : 0.03 Å^{-1} ~ 100 Å^{-1}
  - Accessible r_{max} of Pair Distribution Function : ~ 200 Å
- High Intensity Powder Diffractometer: $\Delta Q/Q$ ~ 0.35 %
  - Able to ordered and disordered phase
  - Minimum sample amount : ~ 1 mg
  - Magnetic structure measurement : ~ 0.3 μ_B (d ≤ 6.5 Å @ 0.6%)

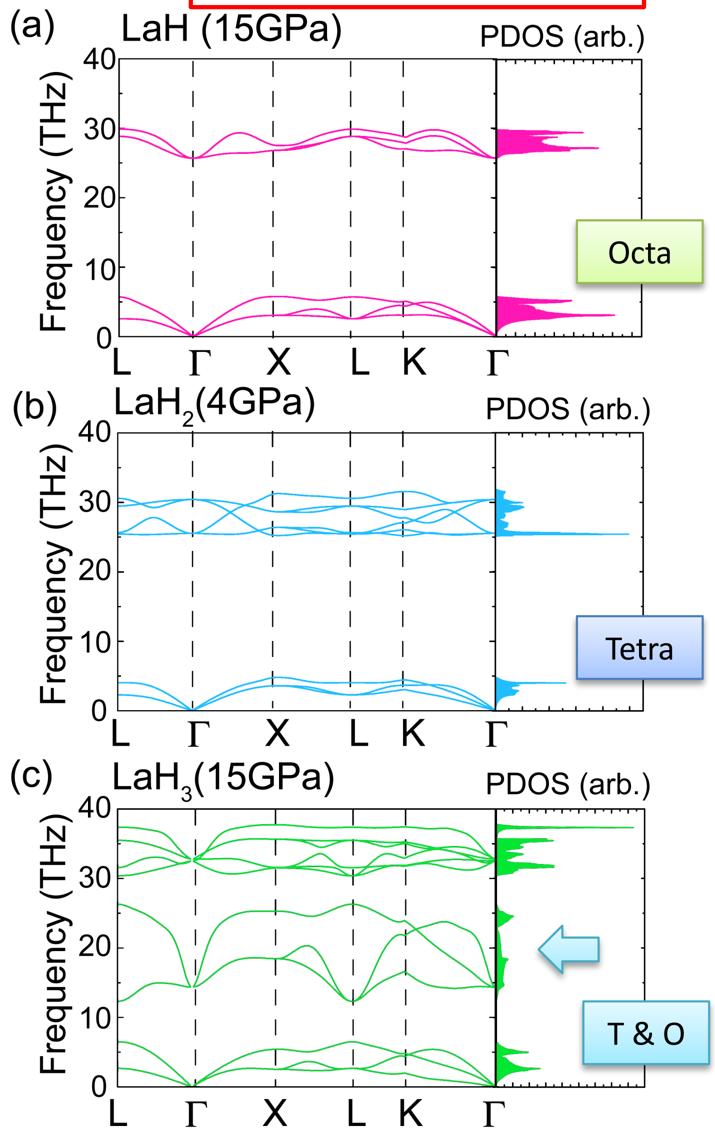
NEDO HydroStar

Analysis of LaD_2 above ten-thousands bar

It was found that LaD (mono-hydride) and LaD_3 exist above 11 GPa bar



LaD_2 (T-site) → LaD (NaCl-fcc, O-site)
+ LaD_3 (fcc, T- and O-sites)



Both neutron and X-ray were necessary to find LaD
Ab-initio calc. supported the existence of LaD and LaD_3

Machida, et al., Phys. Rev. Lett. **108**, 205501 (2012).

HYDRO STAR

Novel hydrogen storage materials developments

Ab-initio calc. predicted novel hydride with Cr and synthesized under high-pressure. X-ray was to used find the condition

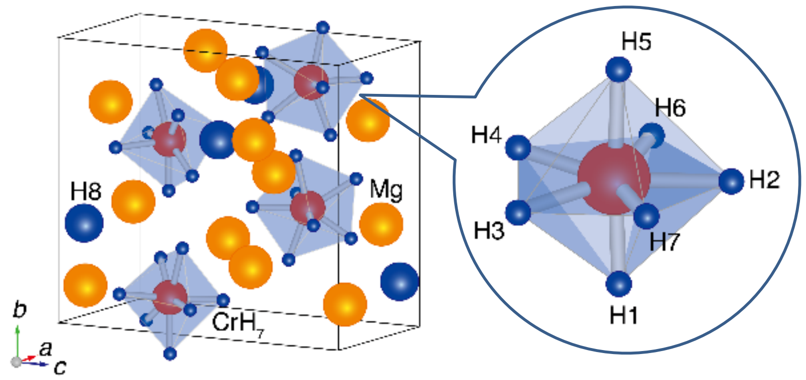
Tohoku Univ. : Takagi & Orimo

Neutron diffraction confirmed hydrogen position

MLF BL21（NOVA）

Establishment of synthetic method of high-density hydrogen storage

Open the way to synthesize very compact hydrogen storage material



Atomic structure of Mg_3CrH_8

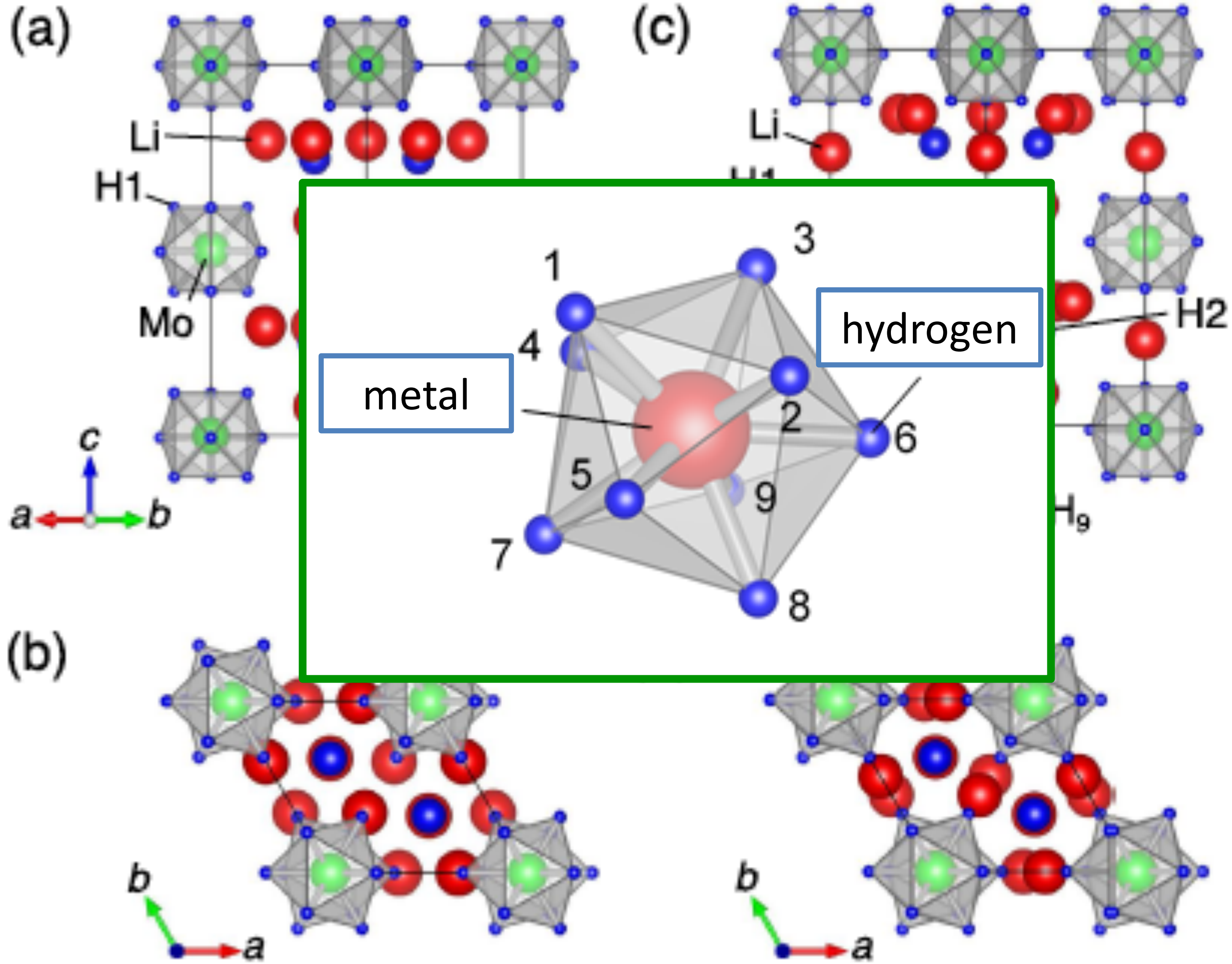
7 hydrogens connected to Cr and forms pentagonal bipyramid CrH_7 ion

（Courtesy：Tohoku Univ. Orimo lab）

Tohoku Univ. IMR/AIMR、JAEA、KEK、Toyota Central Lab.

Takagi, S., et al., Angewandte Chemie, 2015: on line

Nine hydrogen around Mo/W/Nb/Ta



(a) Li_5MoH_{11} Li_5WH_{11} (b) Li_6NbH_{11} Li_6TaH_{11}

The periodic table

www.webelements.com

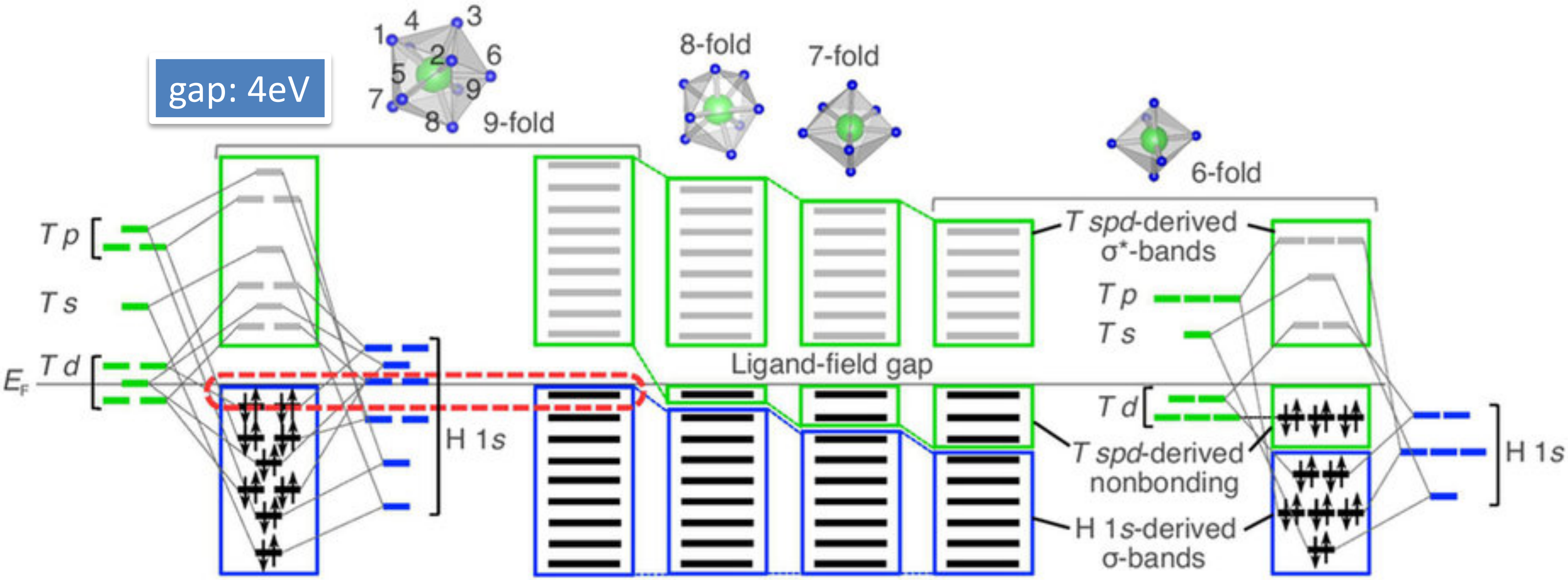
Key:
Element Name
Atomic number
Symbol
Atomic weight (mean relative mass)

1	2		3	4	5	6	7	8	9	10	11	12	13	14	15	16	17	18
Hydrogen 1 **H** 1.008																		Helium 2 **He** 4.0026
Lithium 3 **Li** 6.94	Beryllium 4 **Be** 9.0122												Boron 5 **B** 10.81	Carbon 6 **C** 12.011	Nitrogen 7 **N** 14.007	Oxygen 8 **O** 15.999	Fluorine 9 **F** 18.998	Neon 10 **Ne** 20.180
Sodium 11 **Na** 22.990	Magnesium 12 **Mg** 24.305												Aluminium 13 **Al** 26.982	Silicon 14 **Si** 28.085	Phosphorus 15 **P** 30.974	Sulfur 16 **S** 32.06	Chlorine 17 **Cl** 35.45	Argon 18 **Ar** 39.948
Potassium 19 **K** 39.098	Calcium 20 **Ca** 40.078(4)		Scandium 21 **Sc** 44.956	Titanium 22 **Ti** 47.867	Vanadium 23 **V** 50.942	Chromium 24 **Cr** 51.996	Manganese 25 **Mn** 54.938	Iron 26 **Fe** 55.845(2)	Cobalt 27 **Co** 58.933	Nickel 28 **Ni** 58.693	Copper 29 **Cu** 63.546(3)	Zinc 30 **Zn** 65.38(2)	Gallium 31 **Ga** 69.723	Germanium 32 **Ge** 72.63	Arsenic 33 **As** 74.922	Selenium 34 **Se** 78.96(3)	Bromine 35 **Br** 79.904	Krypton 36 **Kr** 83.798(2)
Rubidium 37 **Rb** 85.468	Strontium 38 **Sr** 87.62		Yttrium 39 **Y** 88.906	Zirconium 40 **Zr** 91.224(2)	Niobium 41 **Nb** 92.906(2)	Molybdenum 42 **Mo** 95.96(2)	Technetium 43 **Tc** [97.91]	Ruthenium 44 **Ru** 101.07(2)	Rhodium 45 **Rh** 102.91	Palladium 46 **Pd** 106.42	Silver 47 **Ag** 107.87	Cadmium 48 **Cd** 112.41	Indium 49 **In** 114.82	Tin 50 **Sn** 118.71	Antimony 51 **Sb** 121.76	Tellurium 52 **Te** 127.60(3)	Iodine 53 **I** 126.90	Xenon 54 **Xe** 131.29
Caesium 55 **Cs** 132.91	Barium 56 **Ba** 137.33	57-70 *	Lutetium 71 **Lu** 174.97	Hafnium 72 **Hf** 178.49(2)	Tantalum 73 **Ta** 180.95	Tungsten 74 **W** 183.84	Rhenium 75 **Re** 186.21	Osmium 76 **Os** 190.23(2)	Iridium 77 **Ir** 192.22	Platinum 78 **Pt** 195.08	Gold 79 **Au** 196.97	Mercury 80 **Hg** 200.59	Thallium 81 **Tl** 204.38	Lead 82 **Pb** 207.2	Bismuth 83 **Bi** 208.98	Polonium 84 **Po** [208.98]	Astatine 85 **At** [209.99]	Radon 86 **Rn** [222.02]
Francium 87 **Fr** [223.02]	Radium 88 **Ra** [226.03]	89-102 **	Lawrencium 103 **Lr** [262.11]	Rutherfordium 104 **Rf** [265.12]	Dubnium 105 **Db** [268.13]	Seaborgium 106 **Sg** [271.13]	Bohrium 107 **Bh** [270]	Hassium 108 **Hs** [277.15]	Meitnerium 109 **Mt** [276.15]	Darmstadtium 110 **Ds** [281.16]	Roentgenium 111 **Rg** [280.16]	Copernicium 112 **Cn** [285.17]	Ununtrium 113 **Uut** [284.18]	Flerovium 114 **Fl** [289.19]	Ununpentium 115 **Uup** [288.19]	Livermorium 116 **Lv** [293]	Ununseptium 117 **Uus** [294]	Ununoctium 118 **Uuo** [294]

*lanthanoids	Lanthanum 57 **La** 138.91	Cerium 58 **Ce** 140.12	Praseodymium 59 **Pr** 140.91	Neodymium 60 **Nd** 144.24	Promethium 61 **Pm** [144.91]	Samarium 62 **Sm** 150.36(2)	Europium 63 **Eu** 151.96	Gadolinium 64 **Gd** 157.25(3)	Terbium 65 **Tb** 158.93	Dysprosium 66 **Dy** 162.50	Holmium 67 **Ho** 164.93	Erbium 68 **Er** 167.26	Thulium 69 **Tm** 168.93	Ytterbium 70 **Yb** 173.05
**actinoids	Actinium 89 **Ac** [227.03]	Thorium 90 **Th** 232.04	Protactinium 91 **Pa** 231.04	Uranium 92 **U** 238.03	Neptunium 93 **Np** [237.05]	Plutonium 94 **Pu** [244.06]	Americium 95 **Am** [243.06]	Curium 96 **Cm** [247.07]	Berkelium 97 **Bk** [247.07]	Californium 98 **Cf** [251.08]	Einsteinium 99 **Es** [252.08]	Fermium 100 **Fm** [257.10]	Mendelevium 101 **Md** [258.10]	Nobelium 102 **No** [259.10]

Symbols and names: the symbols and names of the elements, and their spellings are those recommended by the International Union of Pure and Applied Chemistry (IUPAC - http://www.iupac.org/). Names have yet to be proposed for elements 113, 115, 117, and 118 and so those used here are IUPAC's temporary systematic names. In some countries, the spellings **aluminum**, **cesium**, and **sulphur** are usual.
Group labels: the numeric system (1–18) used here is the current IUPAC convention.
Atomic weights (mean relative masses): these are the IUPAC 2009 values and given to 5 significant figures. The last significant figure of each value is considered reliable to ±1 except where a larger uncertainty is given in parentheses. Representative values for those elements having an atomic weight interval are given (H, Li, B, C, N, O, Si, S, Cl, Tl). Elements for which the atomic weight is given within [brackets] have no stable nuclides and are represented by the element's longest lived isotope reported in the IUPAC 2009 values.

Level of H 1s (σ bond) electron at Fermi level



- At 94 GPa, the band gap will be disappear?
  ＝ High-Tc superconductivity realized with hydrogen？

Hydrogen & Superconductivity

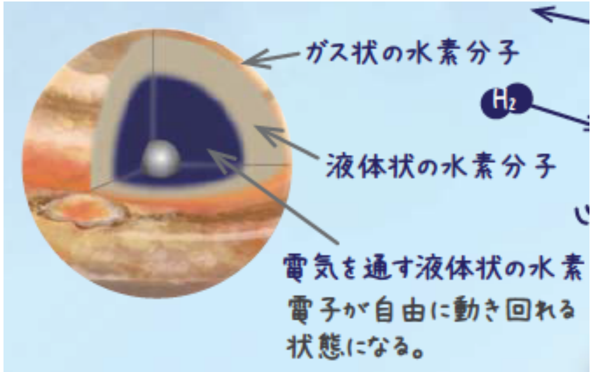
- BCS theory
  - Transition temperature, Tc $\propto {}^{1}/_{\sqrt{m}}$
  - If the hydrogen can commit the electron conductivity, Tc can be room temp?

- H_2S (2015)
  - 150 GPa, Tc = 200 K

- Pd-H/D (2016)
  - 10 MPa   52-61 K（not confirmed）

- La-H（2019)
  - 188 GPa, Tc = 260K

- H（2019)
  - 425 GPa, metallic phase observed（not confirmed）



Super conduction at the core of Jupiter?

Structure of hydrogen is essential to understand the pressure effect on the super conductivity

Summary

- Material science with 4 quantum beams at IMSS, KEK
  - Synchrotron radiation, neutrons, muons and slow positrons
  - Variety of experiments!

- Hydrogenous materials studies with neutron at J-PARC MLF were introduced.