北海道大学
HOKKAIDO UNIVERSITY

28th Aug. - 1st Sep. 2023

Ministry of Education, Culture, Sports, Science and Technology (MEXT) International Nuclear Human Resource Development Initiative Project (Subsidy for Promotion of Nuclear Human Resource Development) "Establishment of New Nuclear Education Center for Future Society by Strengthening Institutional Collaboration".

Neutron Activation Analysis and Elemental Analysis Experiments

Hirotaka Sato

Graduate student recruitment in progress!

Hokkaido University Graduate School of Engineering Division of Applied Quantum Science Laboratory of Neutron Beam Science and Engineering

Website: https://www.eng.hokudai.ac.jp/labo/QBMA/

Contents ①

- **Quantum Beam Science and Neutron Beam Science**
- **Particle accelerators (using the electron linear accelerator at Hokkaido University as an example)**
- **Neutron sources using particle accelerators (focusing on pulsed neutron sources)**
- **Neutron energy spectrum**
- **Neutron transport and detection**
  - Energy decomposition of white neutrons by TOF method
  - Neutron imaging using two-dimensional position sensitive detectors
- **Reaction cross sections between neutrons and nuclei**
- **Radiation Safety of Neutron Beam Facilities**

Contents ②

- **Principles of Elemental Analysis by Neutron Activation Analysis**
- **Applications of Neutron Activation Analysis**
  - Investigation of the Cause of Death of Napoleon
  - The Hayabusa Project
- **Pulse Height Spectra of NaI Scintillation Detectors**
- **Energy Calibration and Energy Resolution of NaI Scintillation Detectors**
- **Backgrounds and their countermeasures**
- **Identification and Qualitative Analysis: Elemental (Nuclide) Analysis**
- **Quantitative analyses: neutron flux estimation, elemental quantification**

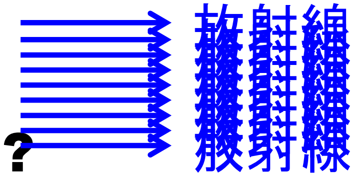
Time schedule planning (loosely interacting and having fun!)

	Mon.	Tue. TA 3 people?	Wed. TA 3 people?	Thu. TA 3 people（morning）	Fri. TA 3 people?
8:45-10:15	Education and training for radiation facilities	Neutron activation analysis experiment (10 pps, 15 min)	Spectral measurements of activated samples	additional experiment（Time, distance, shielding, and so on?）	presentation
10:30-12:00	Explanation of facilities, tour of facilities	NaI scintillation gamma-ray spectrometer test	Element (nuclide) identification	Data analysis of additional experiments	presentation
13:00-14:30	Introduction to quantum beam/Neutron science	Energy calibration experiment	Neutron flux estimation (element determination)	Research and presentation preparation	presentation, summary, and dismissal
14:45-16:15	Introduction to Accelerators, Neutron sources, and Neutron engineering	Energy resolution evaluation, background	Setting the theme of the presentation	Research and presentation preparation	Black: Lectures, etc. Blue: Practice Red: Practical training in a controlled area
16:30-18:00	Introduction to Neutron activation analysis	Discussion (buffer)	Discussion (buffer)	Research and presentation preparation	

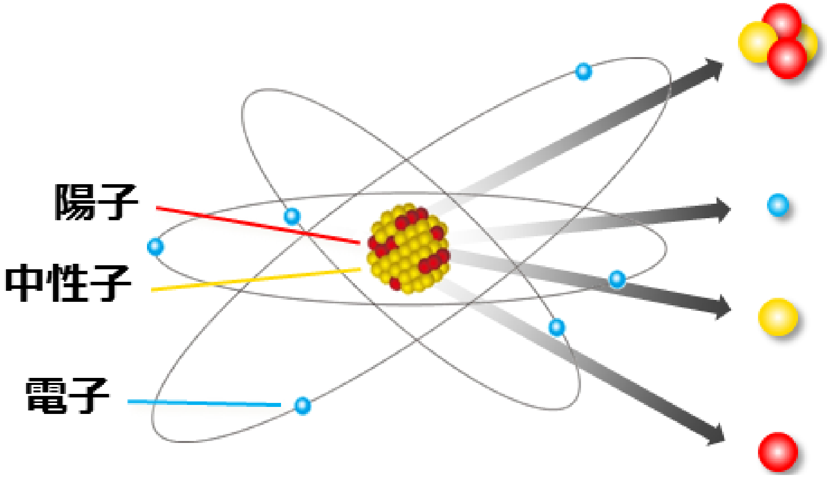
Quantum beam science and Neutron beam science

What is a quantum beam?

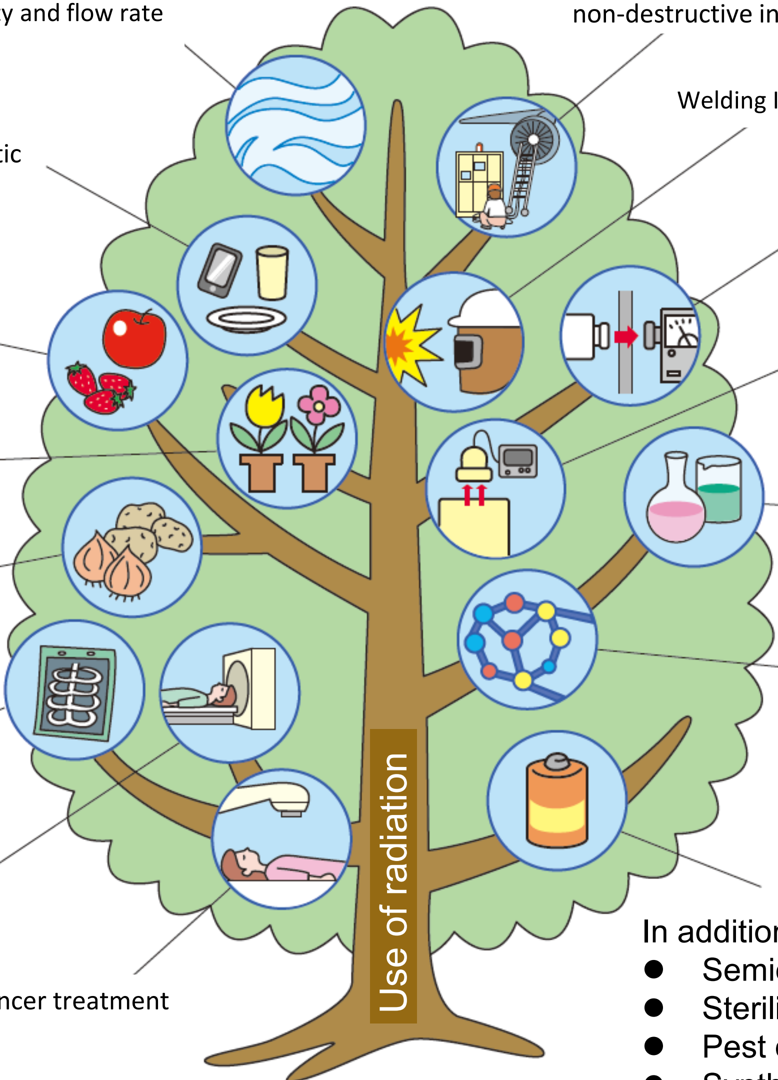
- **Beams of radiation**
- Beam：High intensity（large number）and highly directional（same direction）flux



- **So, what is radiation?**
- Particle beam flowing with high kinetic energy
  - High energy electromagnetic waves（light and radio-waves = photons）



Examples of the use of radiation are manifold.



Use of radiation

Investigation of flow velocity and flow rate

Change (absorption)

reinforced plastic

Investigation of ripening, etc.

Change (absorption)

Selective breeding

Germination Prevention

X-ray examination

View (transparent)

X-ray CT

Cure (absorption)

Cancer treatment

non-destructive inspection

View (transparent)

Welding Inspection

Thickness measurement

From the inside of your room to the edge of the universe

New drug development

Chemical structure research

See (scatter)

isotope battery

Make (absorption)

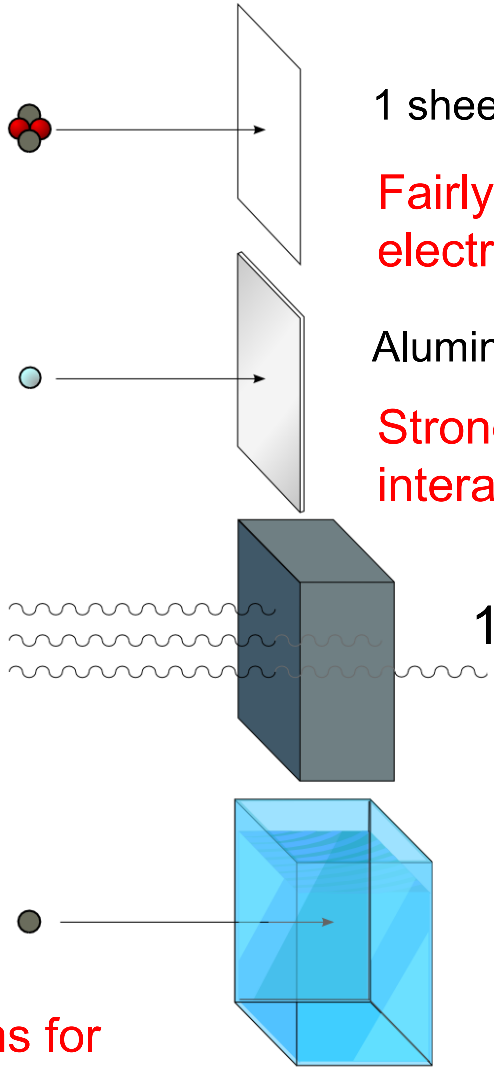
In addition to this

- Semiconductor processing
- Sterilization of medical instruments
- Pest detoxification
- Synthesis of new elements (Nihonium), etc.

Image source: Federation of Electric Power Companies of Japan, Nuclear and Energy Drawings 2015 6-2-5

Interaction of different types of radiation with different materials

Ionizes and excites matter.
Scattered and absorbed by the substance.



Helium-4 nuclei(heavy charged particle) α

1 sheet of paper

Fairly strong electromagnetic interaction

Electrons(charged particle) β

Aluminum several mm thick

Strong electromagnetic interaction

Ectromagnetic waves(x-rays and gamma rays) γ

10 cm thick lead

Neutrons n

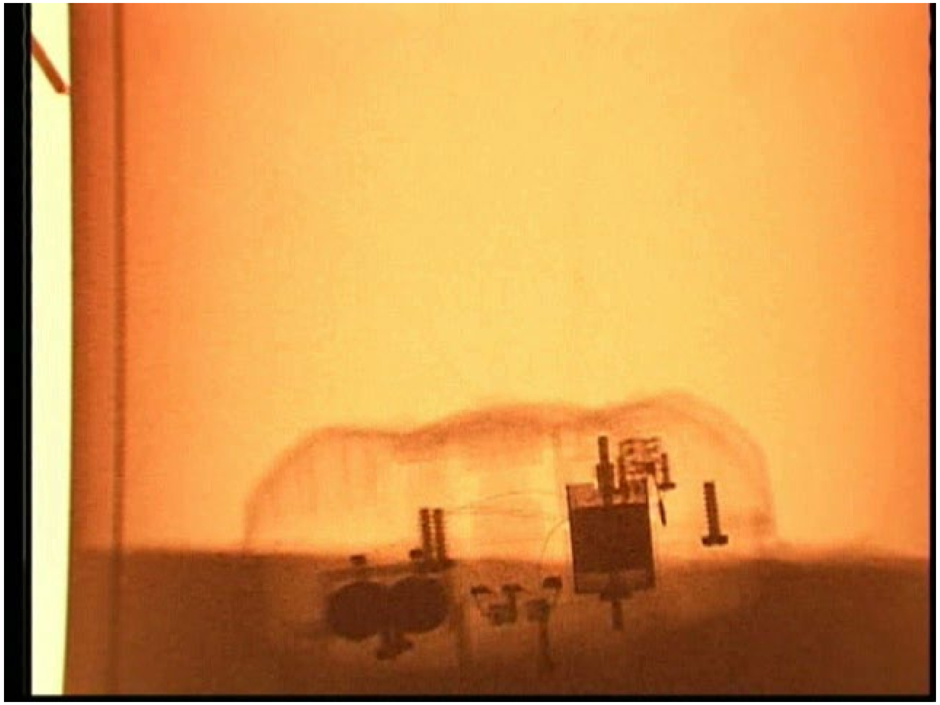
10 cm thick water
1 cm thick boron carbide
1 mm thick cadmium

Frequently used as quantum beams for materials and life sciences.

Picture: http://ja.wikipedia.org/wiki/%E6%94%BE%E5%B0%84%E7%B7%9A

Time-resolved imaging: Dynamic radiography/tomography

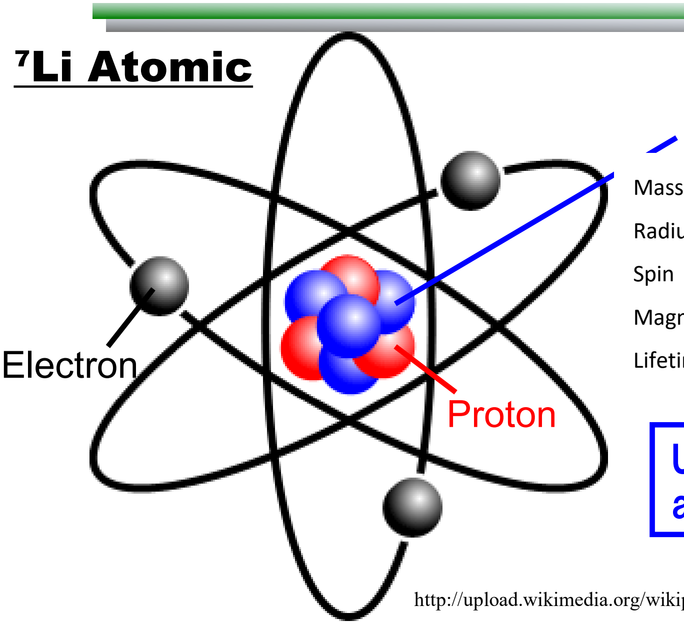
70 keV X-ray



25 meV neutron

Neutron properties and applications

^{7}Li Atomic



Neutron

Mass	$1.674928(1) \times 10^{-27}$ kg
Radius	0.7 fm
Spin	$-\hbar/2$
Magnetic moment	$-9.6491783(18) \times 10^{-27}$ JT^{-1}
Lifetime	885.9 ± 0.9 sec

Unique characteristics are utilized.

http://upload.wikimedia.org/wikipedia/commons/e/e2/Stylised_Lithium_Atom.png

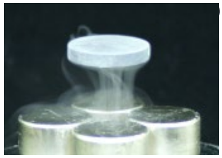
✓ **Electrically neutral**
- High material permeability
- Ability to identify light elements and isotopes

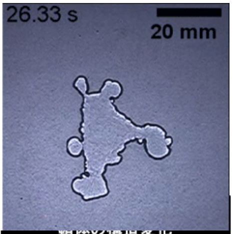
✓ **Mass close to the nucleus of an atom**
- Capable of nondestructively analyzing microscopic spatio-temporal information of materials.

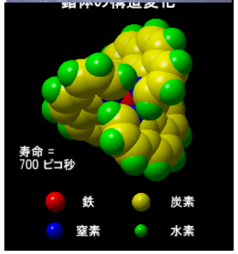
✓ **With spin and magnetic moment**
- Exploration of magnetic information

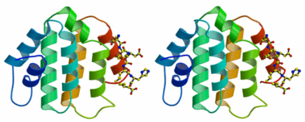
Applications in the fields of materials, life, earth, planet, and physics

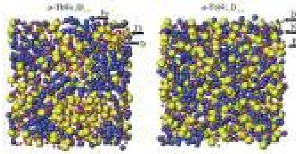
- Atomic and molecular structure
- analysis
- Dynamics analysis of atoms and
- molecules
- Fluoroscopy（Imaging）
- Elemental and isotopic analysis
- Particle physics and nuclear physics











※画像は全てKEKホームページよりお借りしました。

Applications in the energy sector

Nuclear power generation
- Transmutation of long-lived radioactive waste

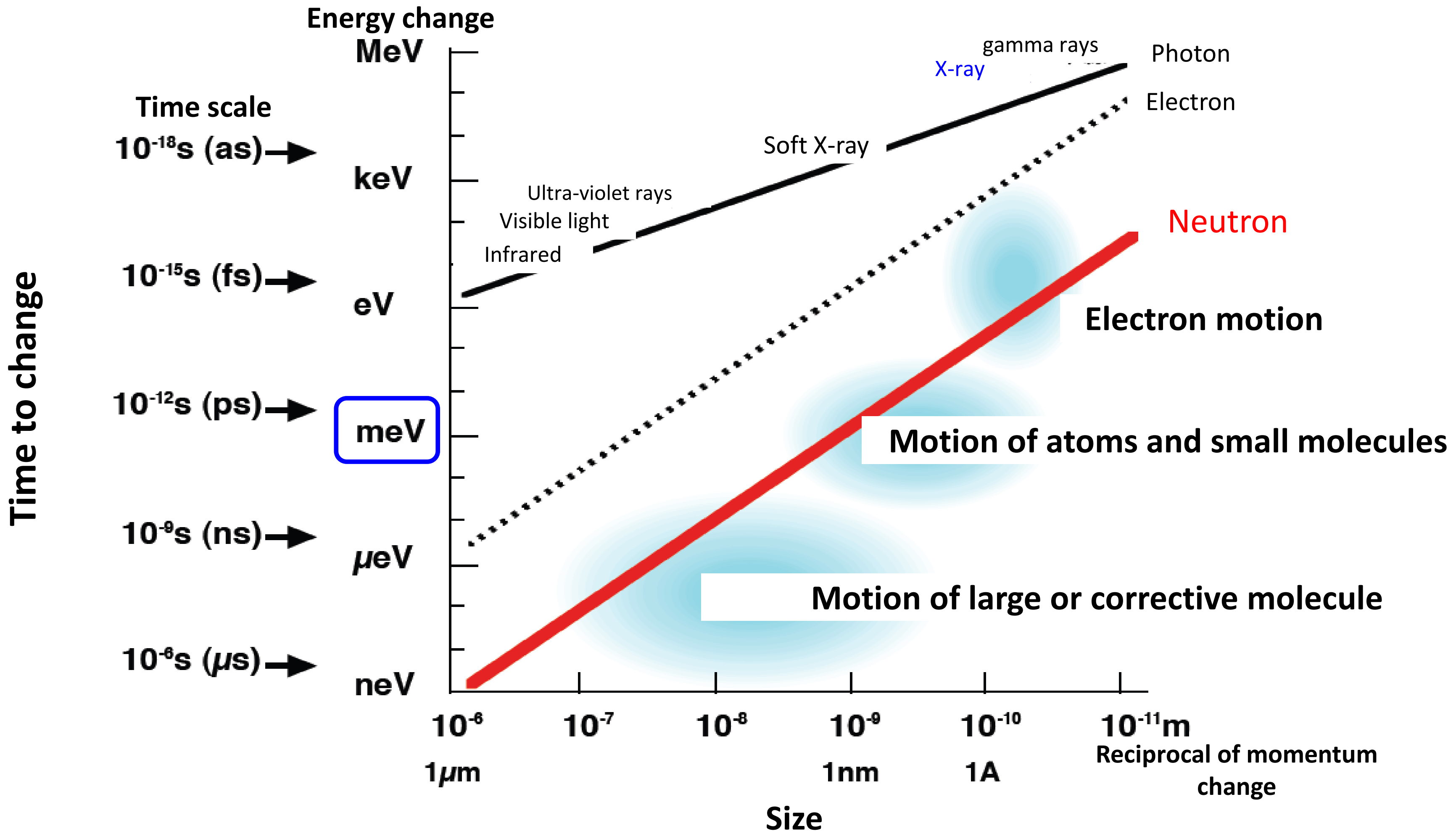
Medical applications

- (Boron) neutron capture therapy
- RI drug production

Other applications

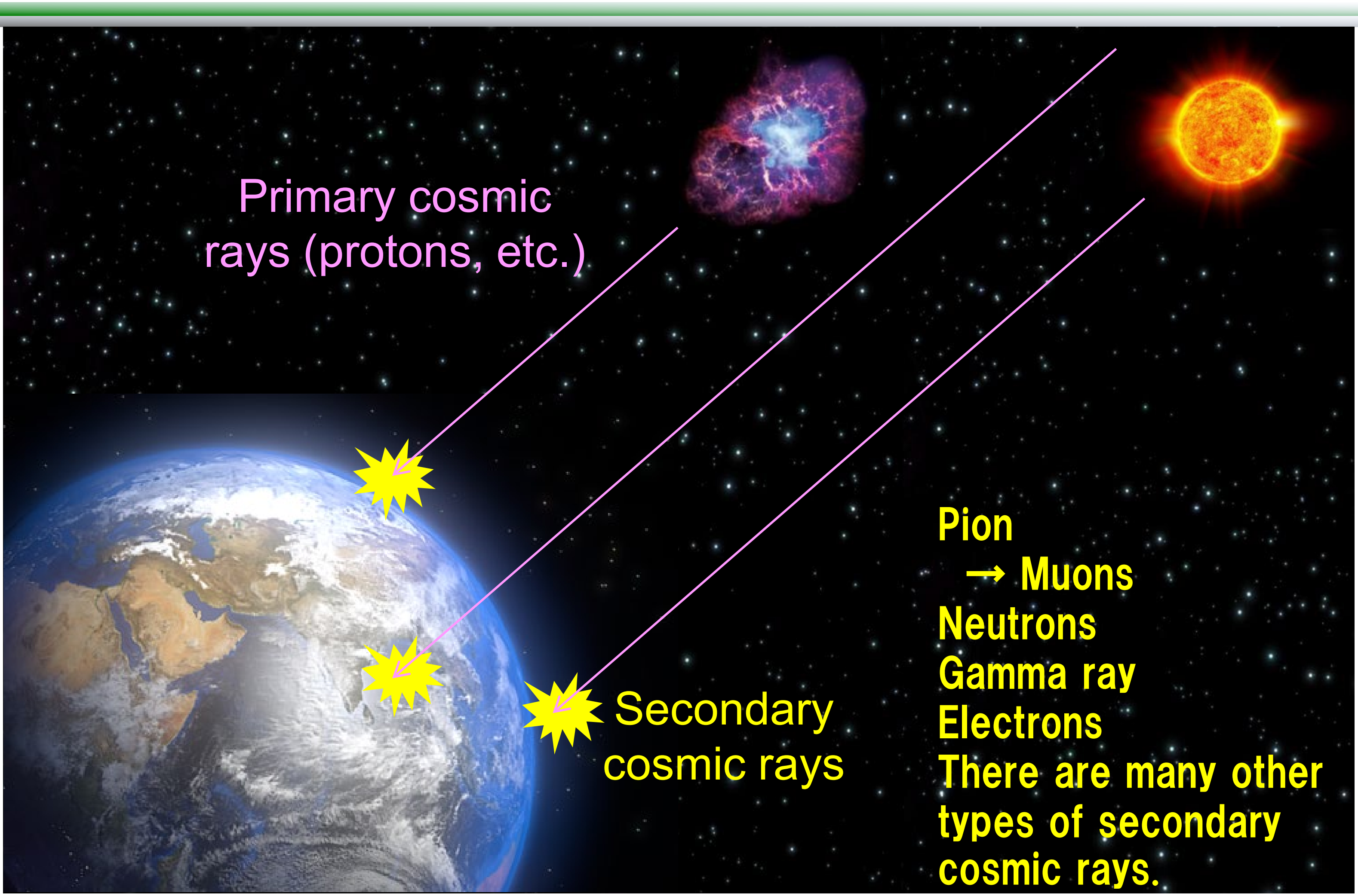
- Neutron Irradiation Testing (Materials & Electronics)
- Moisture meters, ...

Mass near nuclear → Matching material analysis



Compared to other radiation, it is necessary to lower the energy to meV.

Radiation source ①:
Cosmic rays（Ultra-high energy）



Primary cosmic rays: Radiation that comes from space. Protons are the main type of cosmic rays. Secondary cosmic rays: Particles formed when primary cosmic rays react with the atmosphere. Muons, etc.

Radiation source ②: Radioactive isotope (RI)

image source：http://blog-imgs-38.fc2.com/w/i/n/wine4wine/R0010667.jpg

- **Alpha (alpha) radiation sources**

  Am (Americium)-241 (about 5.5 MeV),

  Po (Polonium)-210 (about 5.3 MeV), etc.

- **Beta (beta) sources**

  Sr-90 (Max. approx. 2.28 MeV),

  Na-22 (Beta+ (Positron) source), etc.

- **Gamma (gamma) radiation source**

  Co-60 (Approx. 1.17 MeV & 1.33 MeV),

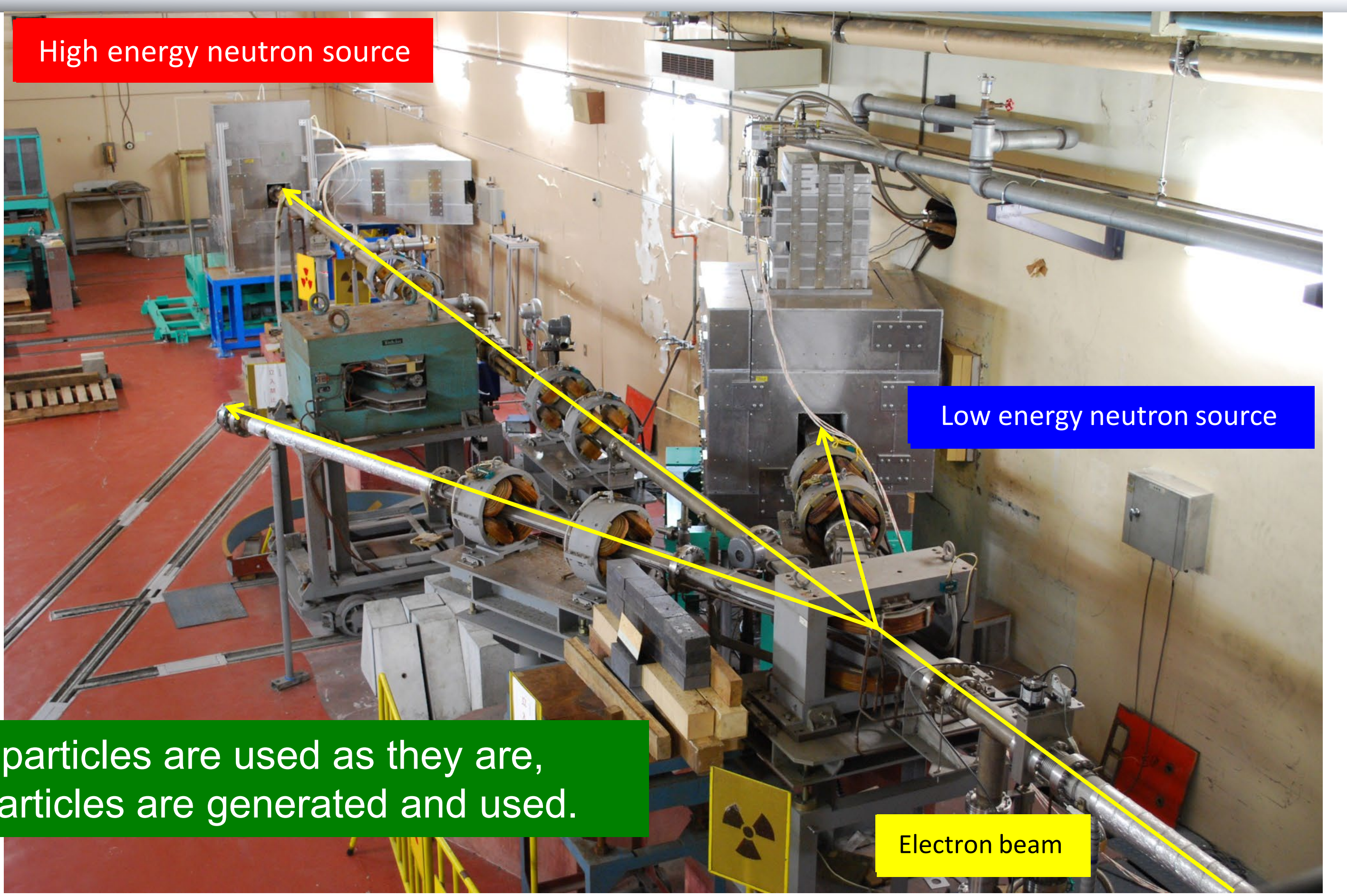
  Cs-137 (Approx. 0.66 MeV), etc.

- **Neutron sources: ^{252}Cf, ^{241}Am-Be, etc.**

The emission is low and radiation is emitted in all directions. but easy to use.

Radiation source ③:
Accelerator（high intensity･high energy）

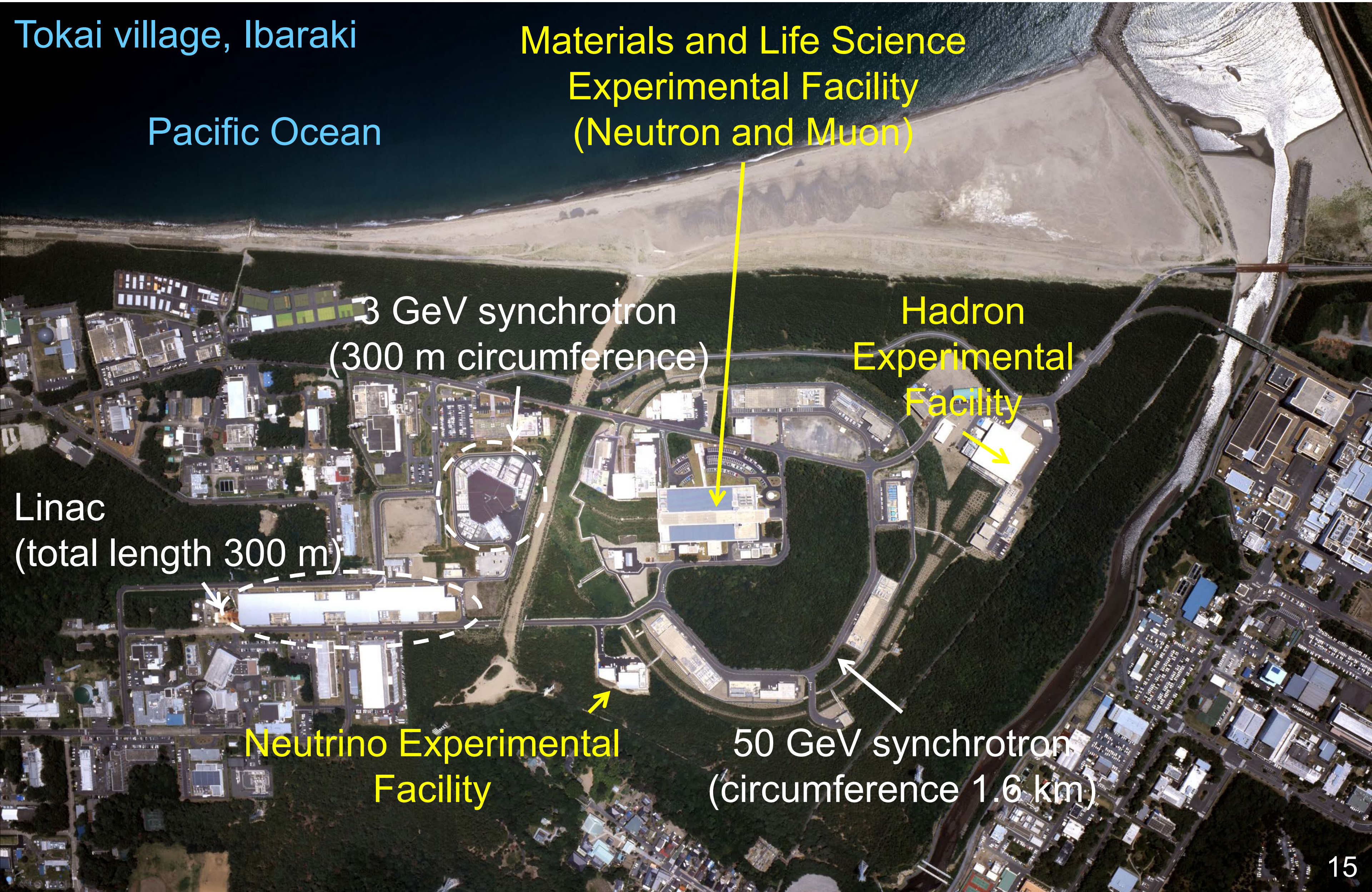
Photo: "Electron Accelerator Driven Neutron Experimental Facility" at faculty of engineering, Hokkaido University



Accelerated particles are used as they are, secondary particles are generated and used.

- Provides high-intensity, highly directional radiation fluxes（Quantum beams）.
- A wide variety of radiation that can be generated（from elementary particles to uranium nuclei）.
- Small accelerators can be found throughout the city（e.g., radiation therapy units in hospitals）.

Japan Proton Accelerator Research Complex (J-PARC)



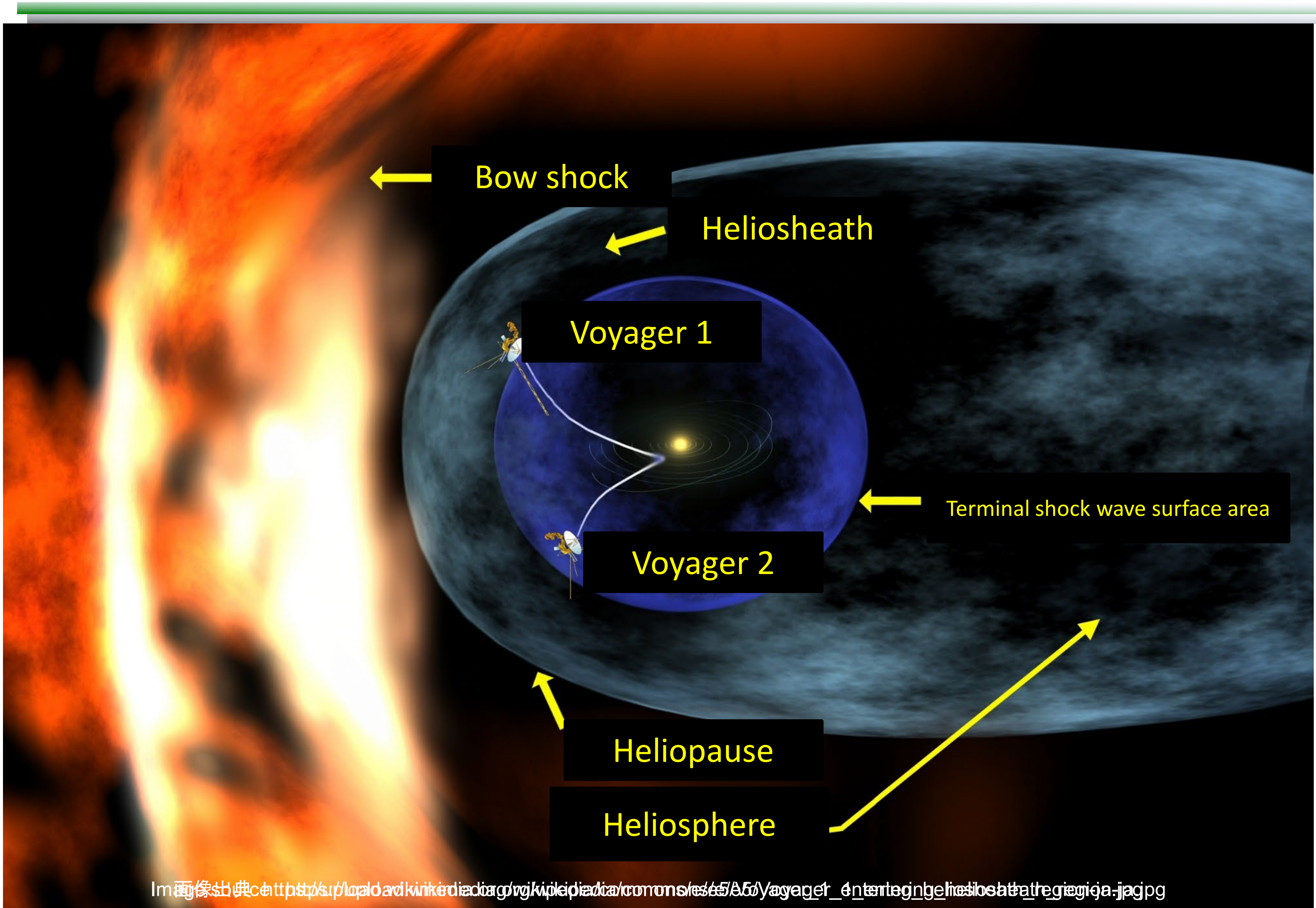
Fire alarm (Smoke detection)

Not widely used in Japanese households...

- **Am-241 (Alpha radiation source) is used to ionize smoke.**
- **Smoke is detected by detecting changes in ionizing current.**
- Capable of detecting smoke with high sensitivity.
- The equipment falls under the category of “Apparatuses loaded with Radioactive Materials," and is therefore subject to substantial restrictions.

Voyager space probe (Launched in 1977) in 2005

Currently, Voyager 1 is out of the solar sphere.



Bow shock

Heliosheath

Voyager 1

Terminal shock wave surface area

Voyager 2

Heliopause

Heliosphere

Image source: https://upload.wikimedia.org/wikipedia/commons/e/e5/Voyager_1_entering_heliosheath_region.jpg

画像出典：https://upload.wikimedia.org/wikipedia/commons/e/e5/Voyager_1_entering_heliosheath_region.jpg

Radioisotope (RI) batteries

Stable power can be supplied over a long period in places where sunlight is not sufficient to reach.

- An α-ray source (Pu-238, Po-210, etc.) is used.
- The heat generated when alpha rays are absorbed by a material is converted into electric power using a thermoelectric conversion element.

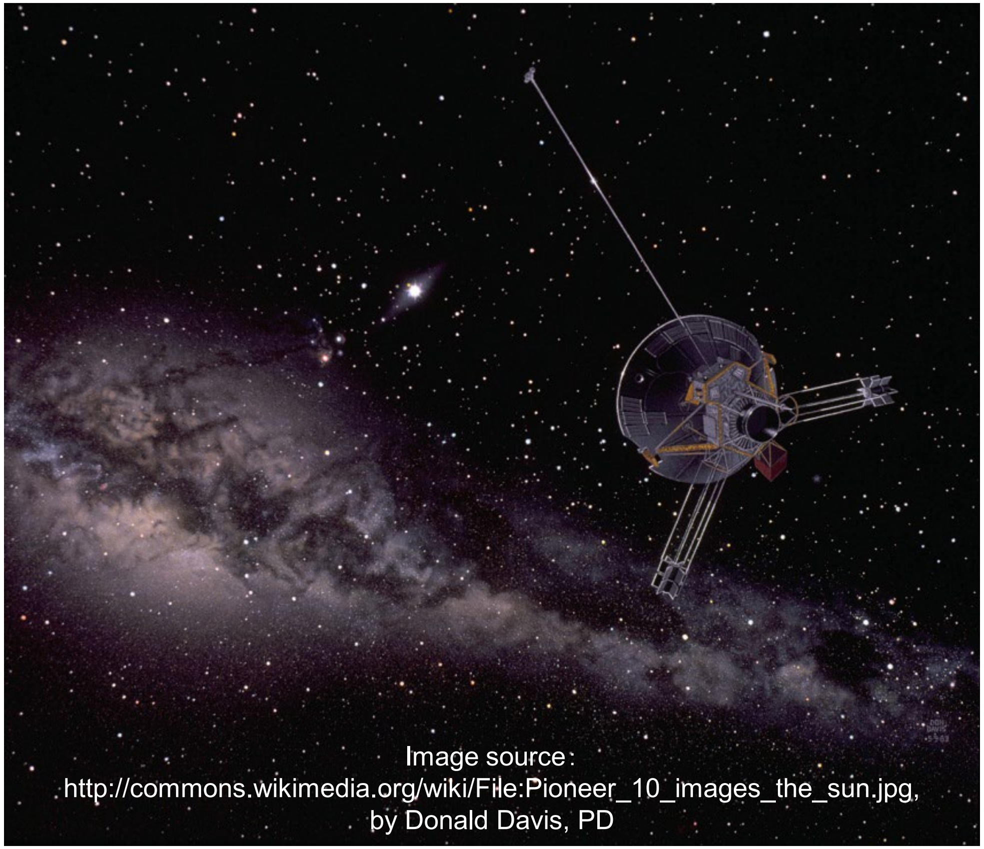
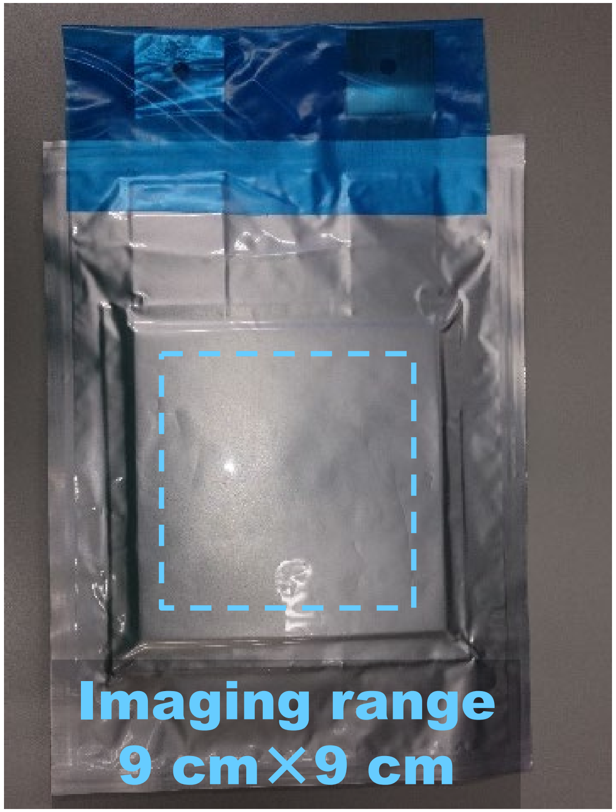


Image source:
http://commons.wikimedia.org/wiki/File:Pioneer_10_images_the_sun.jpg,
by Donald Davis, PD

Neutron scattering analytical imaging of automotive lithium ion batteries

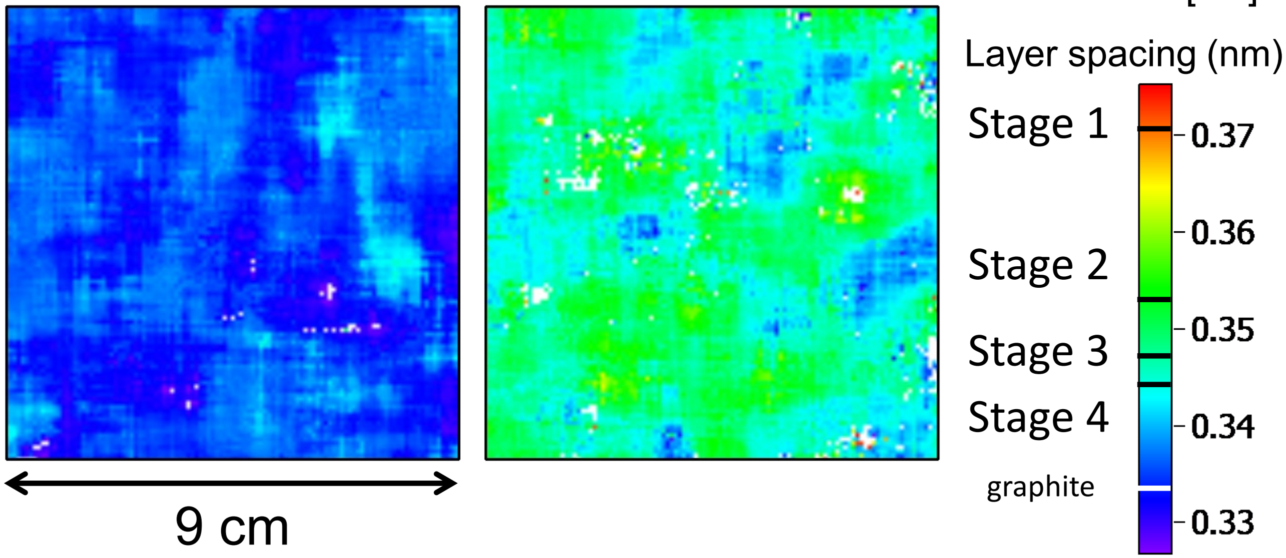
Hokkaido University - Toyota Motor Corporation

Battery photo



Before charging **After charging**

d_{002} crystal layer spacing of graphite anode material [nm].

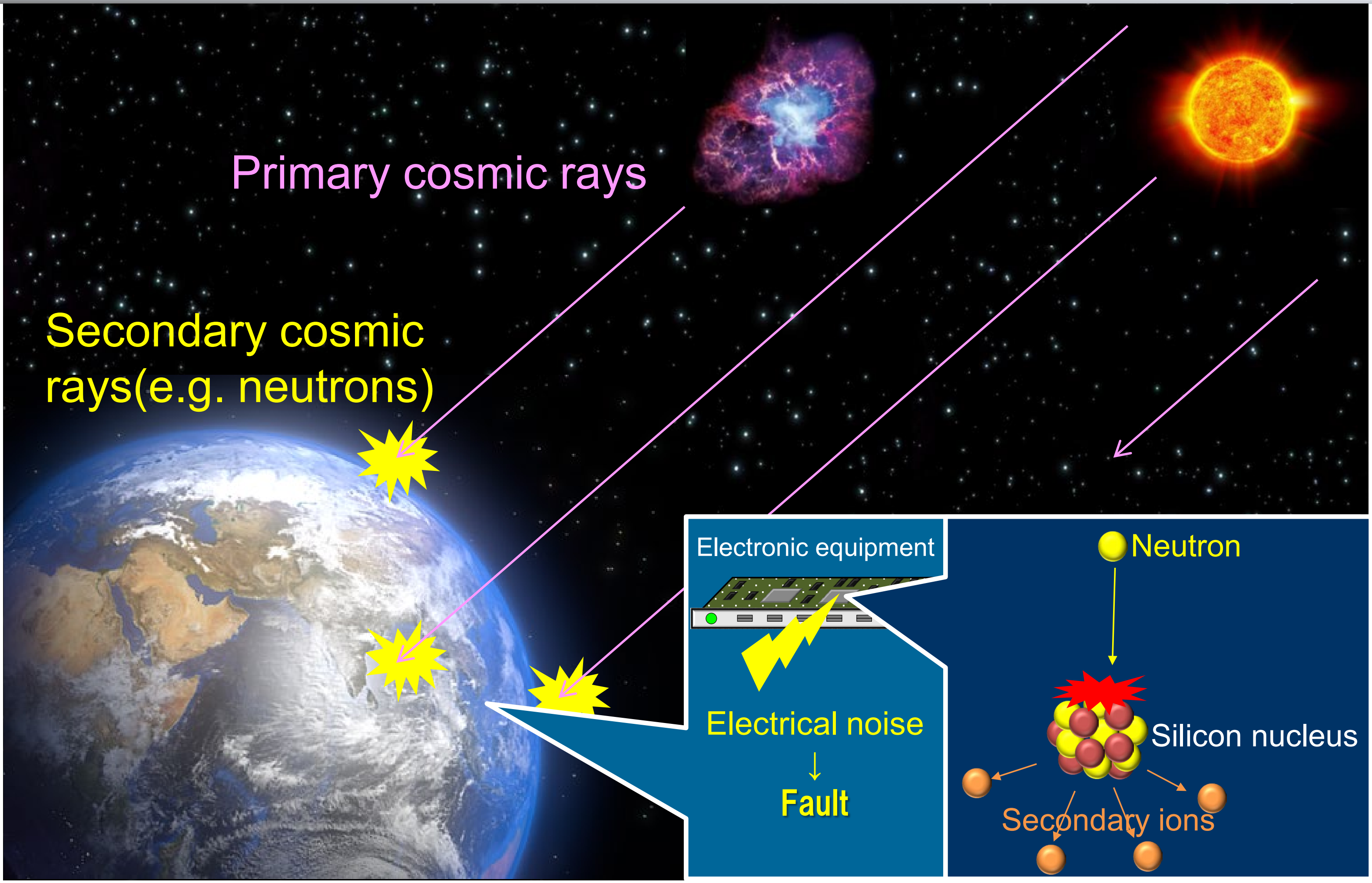


Neutrons penetrate the cathode and electrolyte to analysis of the crystal structure of the anode only

[Layer spacing expanded by charging (penetration of Li ions into the graphite layer)]. Differences exist depending on the location within the product.

Prevention of large-scale communication network failures caused by cosmic rays using accelerator beams

Joint research between Hokkaido University and NTT



High-energy neutrons generated by the accelerator are absorbed by electronic equipment, and failures are intentionally generated to estimate the frequency of failures in nature, etc.

Experiments on countermeasures against cosmic ray neutron soft errors in transmission systems for information and communication networks

Detoxification of pests

Melon fly extermination operation in Okinawa

- The larvae of the cucurbit fly feed on the contents of vegetables and are highly fecund, which can be devastating to vegetable production.
- In the 1970s, an eradication campaign was initiated in Okinawa to prevent the melon fly from invading mainland Japan.
- One of the most effective strategies was the use of a Co-60 gamma radiation source to irradiate large numbers of melon fly larvae, rendering them sterile, and then releasing them into the habitat in large numbers to suppress their reproduction.
- By 1993, the eradication of the melon fly had been successfully achieved. The number of flies used exceeded 50 billion.

画像出典：http://commons.wikimedia.org/wiki/File:Melon_fly_(Bactrocera_cucurbitae)_03.jpg

Food Irradiation

- Purpose: extension of shelf life of food products, sterilization, insecticides, etc.
- In Japan, the Shihoro Agricultural Cooperative in Hokkaido irradiates potatoes only with Co-60 gamma rays (To prevent germination).

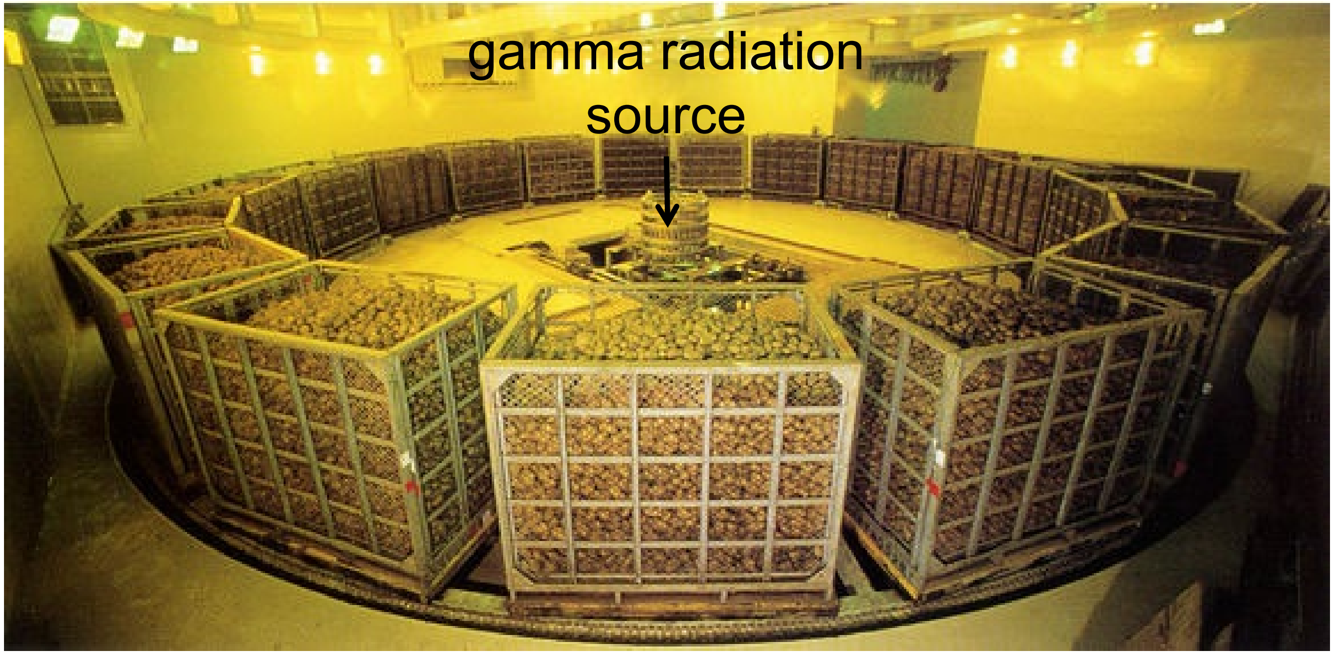
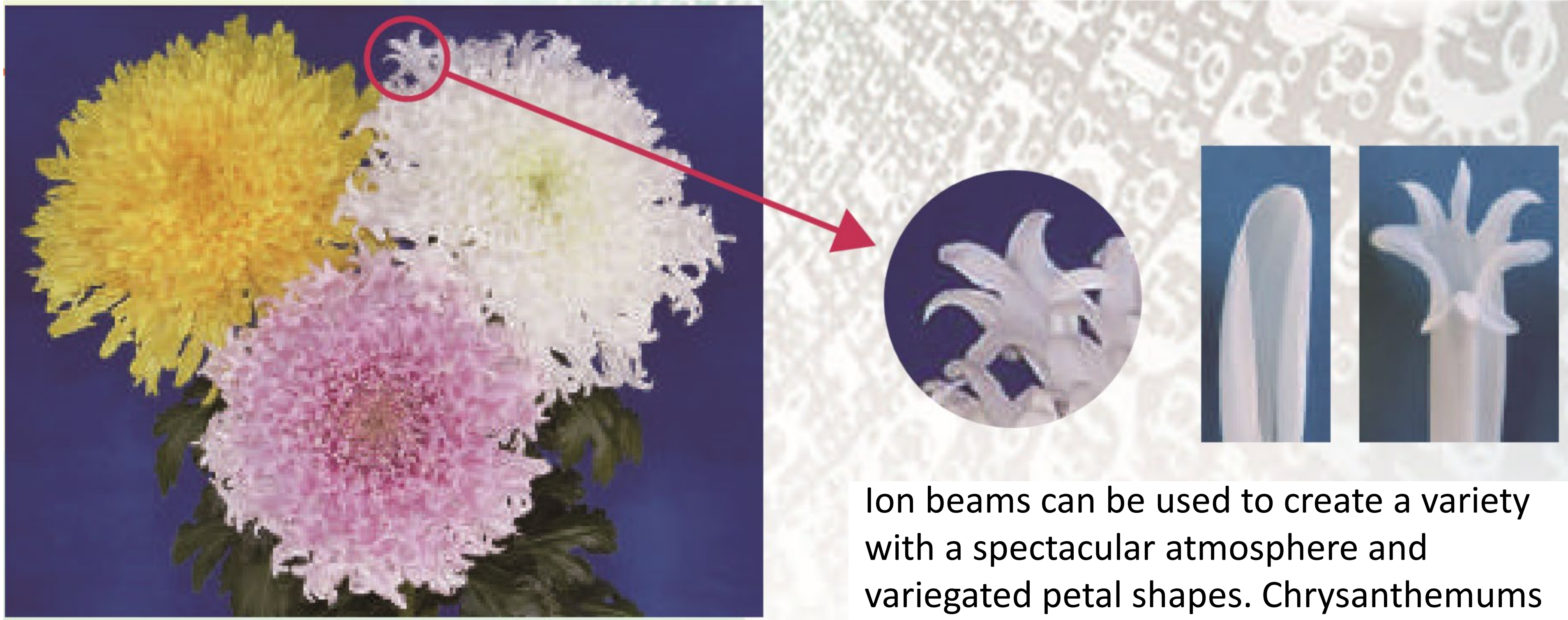


Image source：http://www.ies.or.jp/publicity_j/mini_hyakka/21/mini21.html

Flower breeding (Application of mutation)

Famous for his work with TIARA (Takasaki Ion Accelerator)

Ion beams with strong DNA-damaging effects are irradiated to increase the probability of genetic mutations related to color and shape.



A new variety of chrysanthemum with gorgeous petals

Ion beams can be used to create a variety with a spectacular atmosphere and variegated petal shapes. Chrysanthemums bred in this way are expected to open up new applications such as celebrations and arrangements.

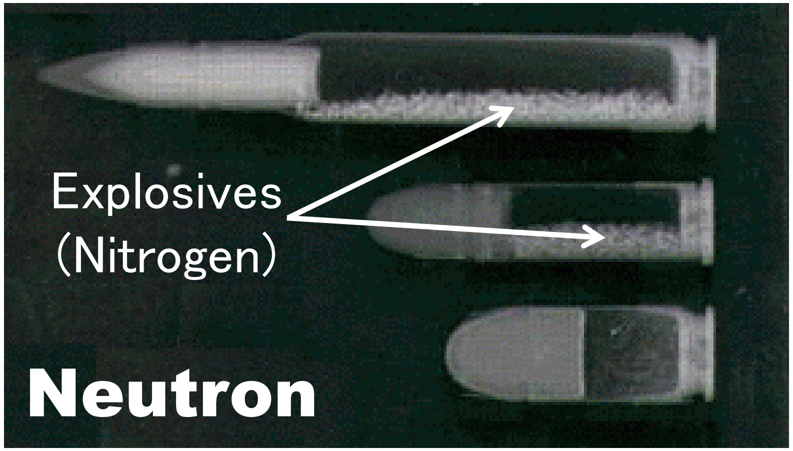
Image source: "Quantum Beam" for a family

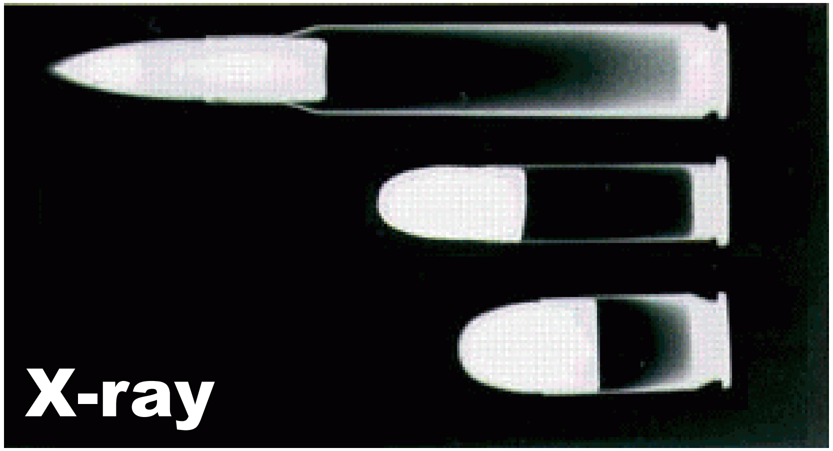
Transmission imaging using neutron beams

Feature ①: High sensitivity to some light elements (hydrogen, lithium, etc.)

Feature ②: Easily permeable to heavy metals within a few centimeters

⇒ Used for visualization of oil behavior inside engines, etc.





Visualization of objects invisible by X-ray imaging (Light elements in heavy metals)

画像出典:ATOMICA「犯罪捜査における放射線利用」

Neutron imaging provides clairvoyance to cultural properties

Transparency of the inside of a bronze sutra tube

for detection of organic materials such as paper and cloth inside a metal container

neutrons are more advantageous than X-rays!

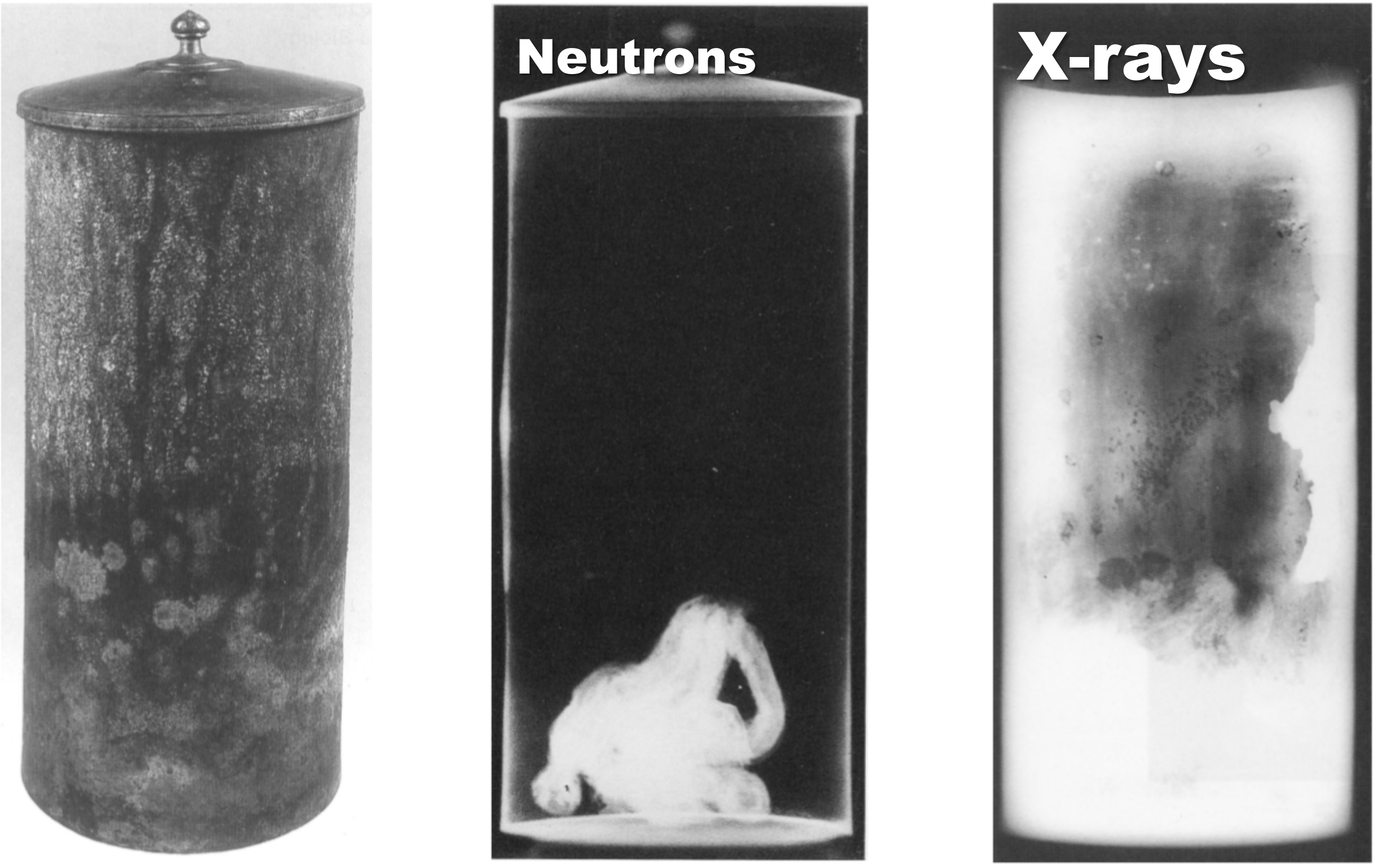


Image source: "Fundamentals and Applications of Neutron Imaging Technology," Japan Radioisotope Association

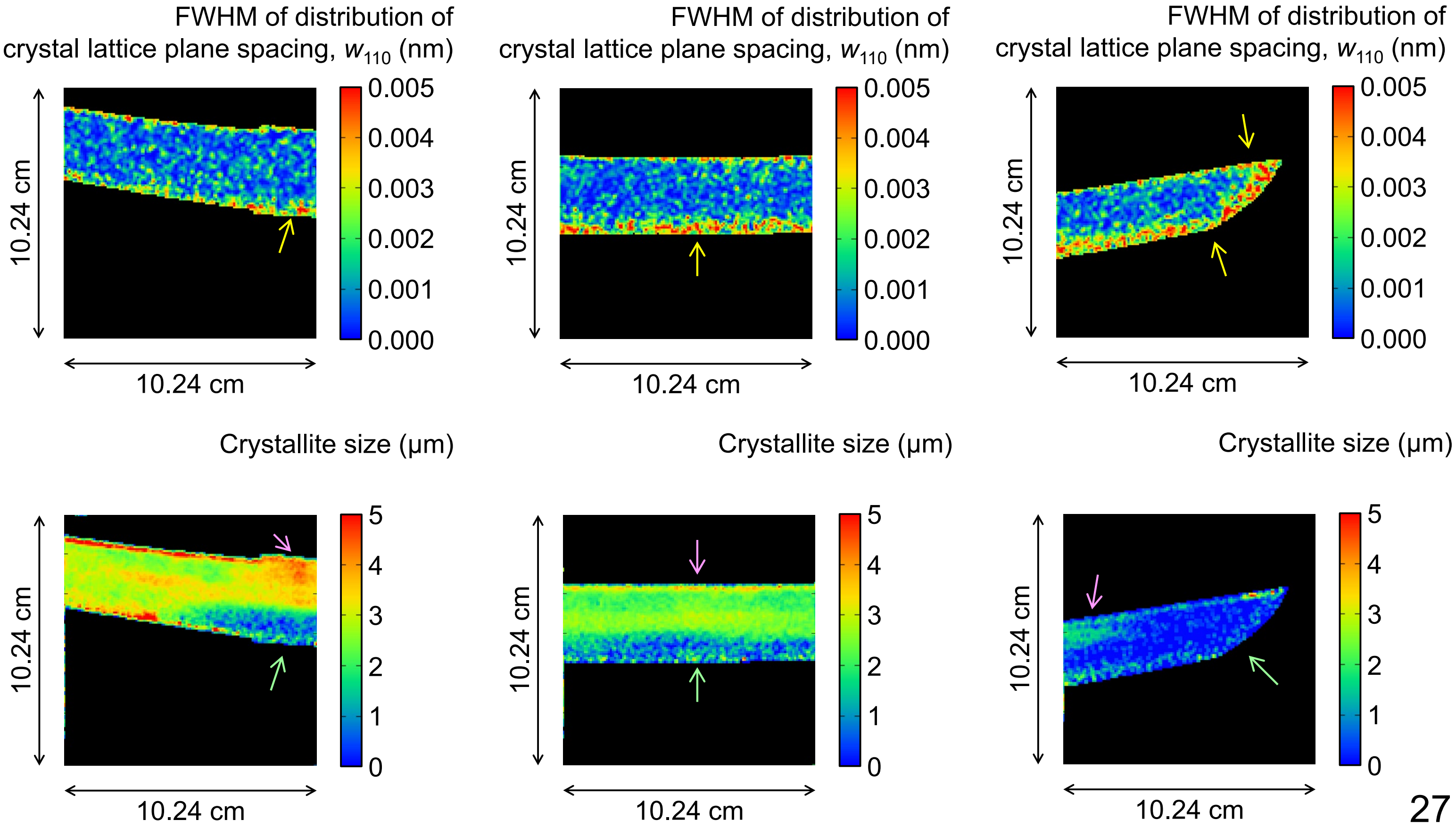
Analysis of Japanese swords without breaking them

International joint research between Japan and Italy, including Hokkaido University

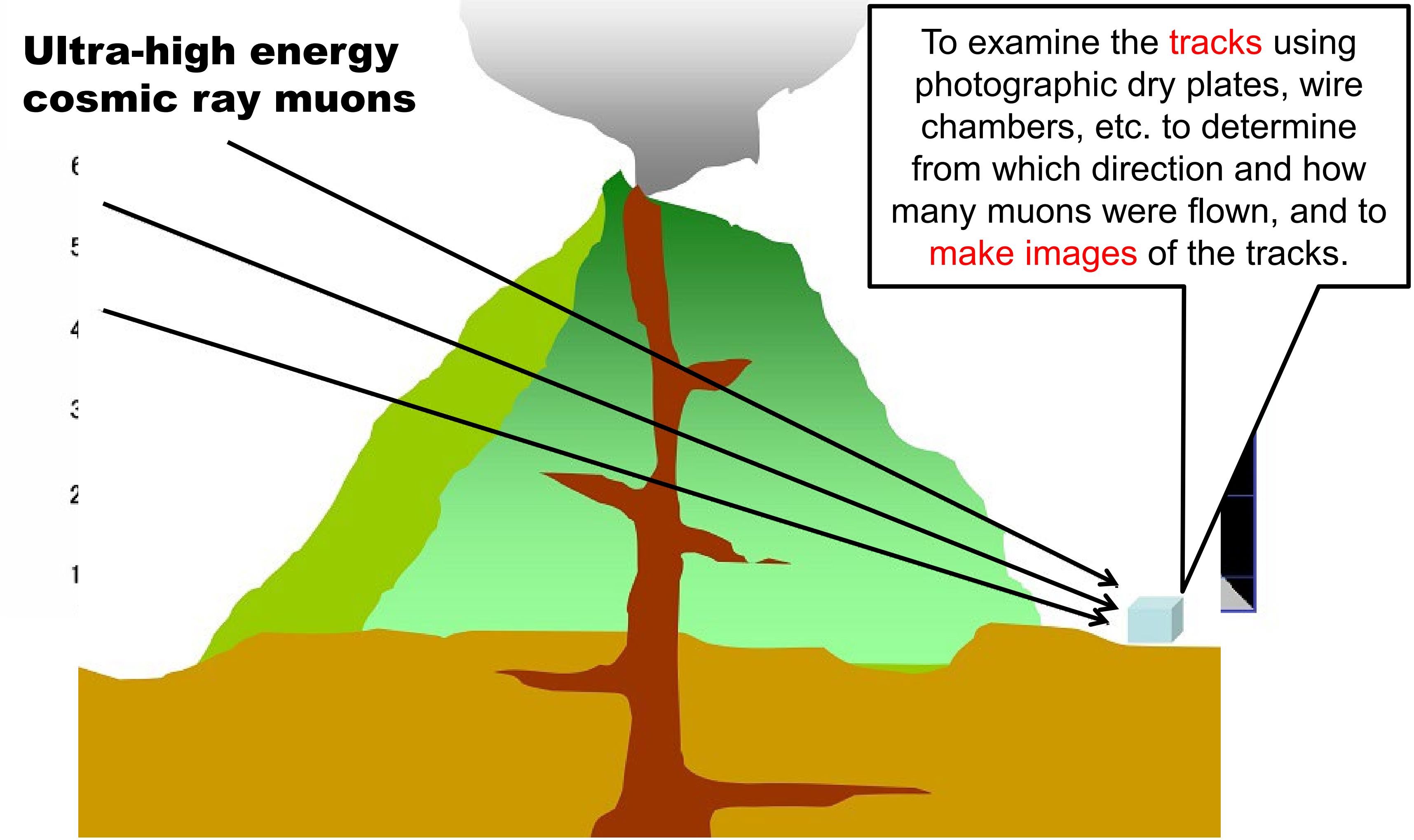
Scattering of low-energy neutrons to **see** heat-treatment-induced strain in the blade edge of Japanese swords and the presence of giant crystal grains in the blade.

Strain size⇒

Crystal grain size ⇒



Volcano imaging using cosmic ray muons



Ultra-high energy cosmic ray muons

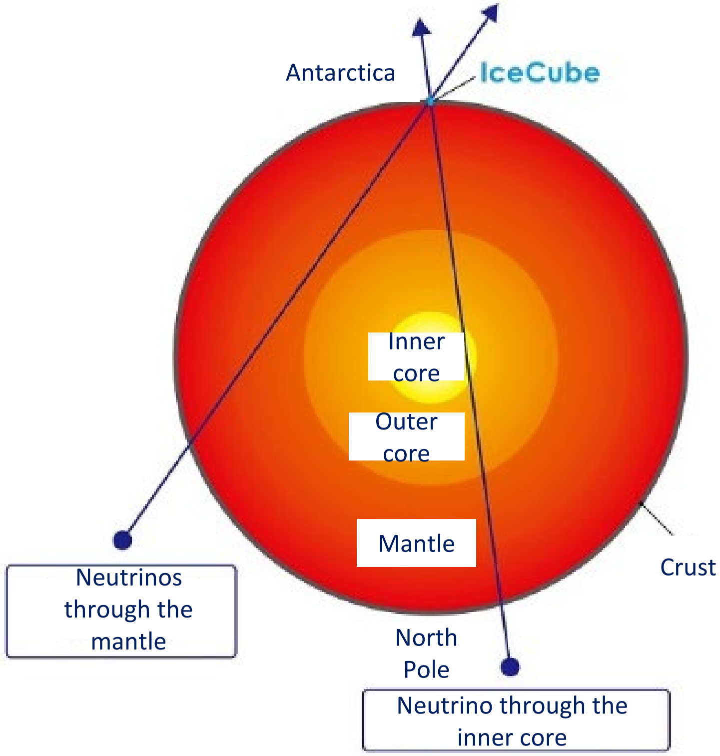
To examine the tracks using photographic dry plates, wire chambers, etc. to determine from which direction and how many muons were flown, and to make images of the tracks.

Enables visualization and analysis of the interior of very large structures

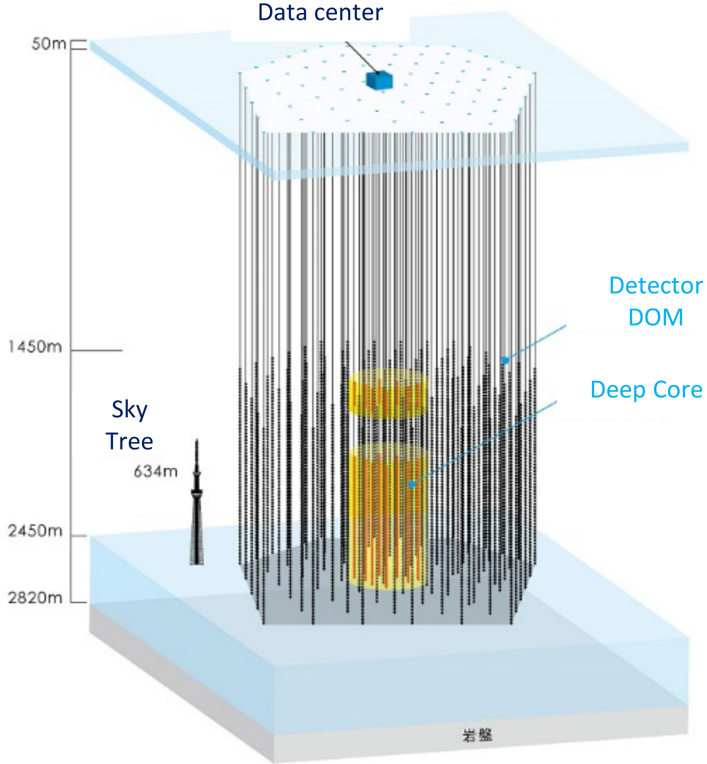
image source：http://www.u-tokyo.ac.jp/ja/utokyo-research/feature-stories/a-window-into-the-earths-interior/

Don’t we enable "x-ray the earth" with radiation (elementary particles) that have a higher ability to penetrate matter?

We are doing! Earth Imaging with Cosmic Ray Neutrinos



IceCube, a super-sized detector in the Antarctic icecap(like a supersized version of Kamiokande)



The day is near when we will actually be able to see through the internal structure of the Earth!

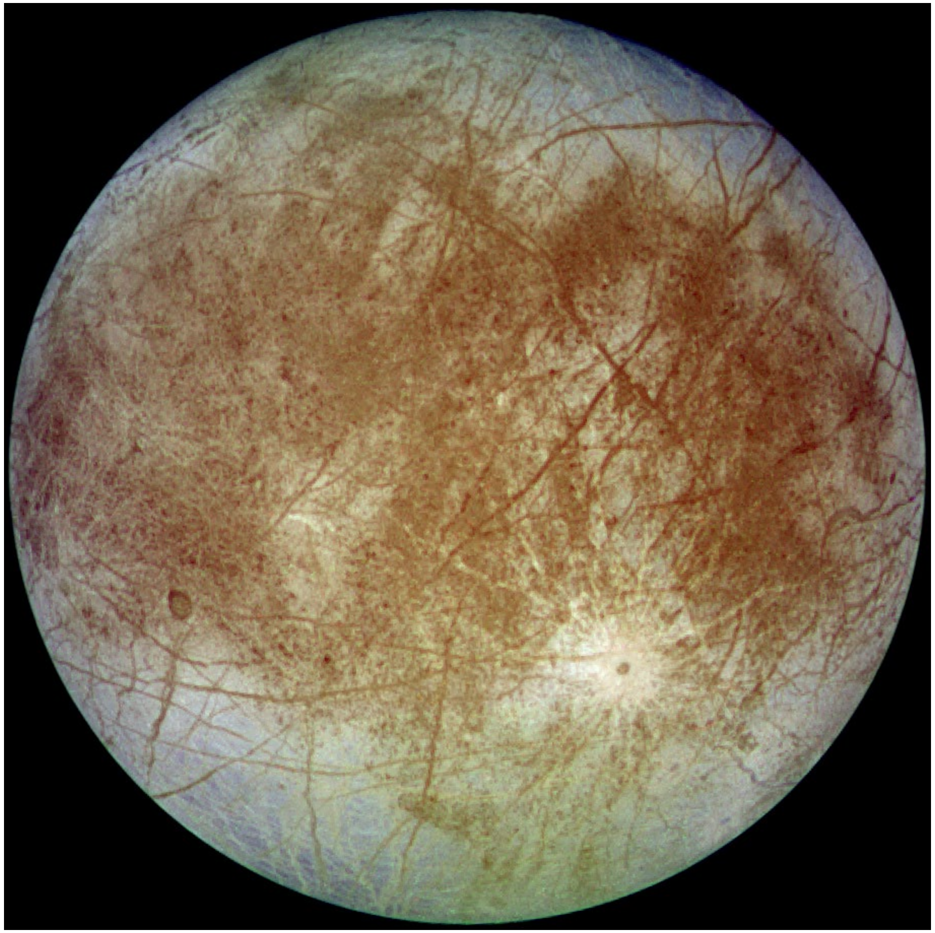
image source：http://www.u-tokyo.ac.jp/ja/utokyo-research/feature-stories/a-window-into-the-earths-interior/

IceCube

Image source：https://www.pri.org/stories/2018-03-18/new-book-recounts-amazing-history-icecube-neutrino-observatory, https://www.livescience.com/9164-world-largest-neutrino-observatory-built-south-pole.html

Research on the effects of cosmic rays on cosmic planetary materials

Europa, a satellite of Jupiter



Europa" environment simulation experiment using the electron accelerator at Hokkaido University

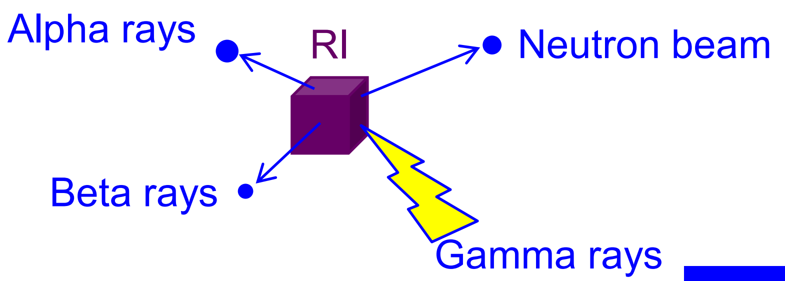
Collaboration with Exploration and Observation Unit, Planetary Space Group, Department of Space Science, Faculty of Science

Particle accelerators (Using the electron linear accelerator at Hokkaido University as an example)

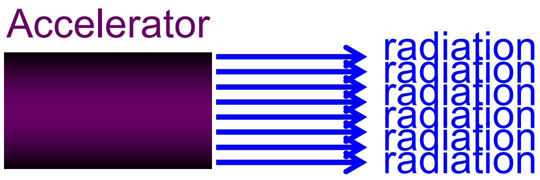
Purpose of accelerator: Generation of high-energy, high-intensity, and highly directional radiation flux

① By supplying a flux (Quantum beam) of high intensity and high directionality,

Radiation generated by radioactive materials

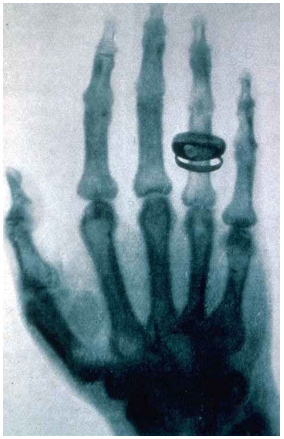


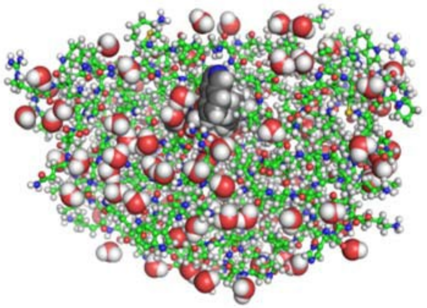
Radiation generated by accelerators



Accelerating particles are used as they are, or secondary particles are generated and used（**a wide variety of radiation that can be generated**）.

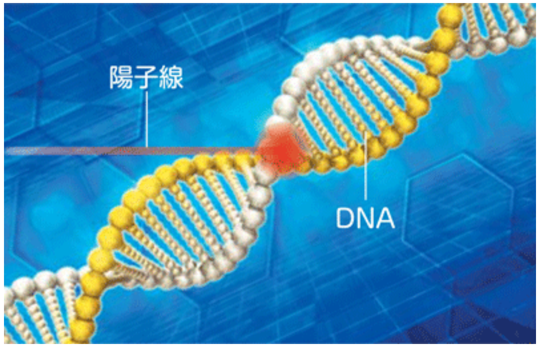
② "**Seeing**", "**changing**", and "**curing**" matter.





Atomic arrangement analysis
atomic motion analysis

Plant and semiconductor breeding
sterilization of medical instruments



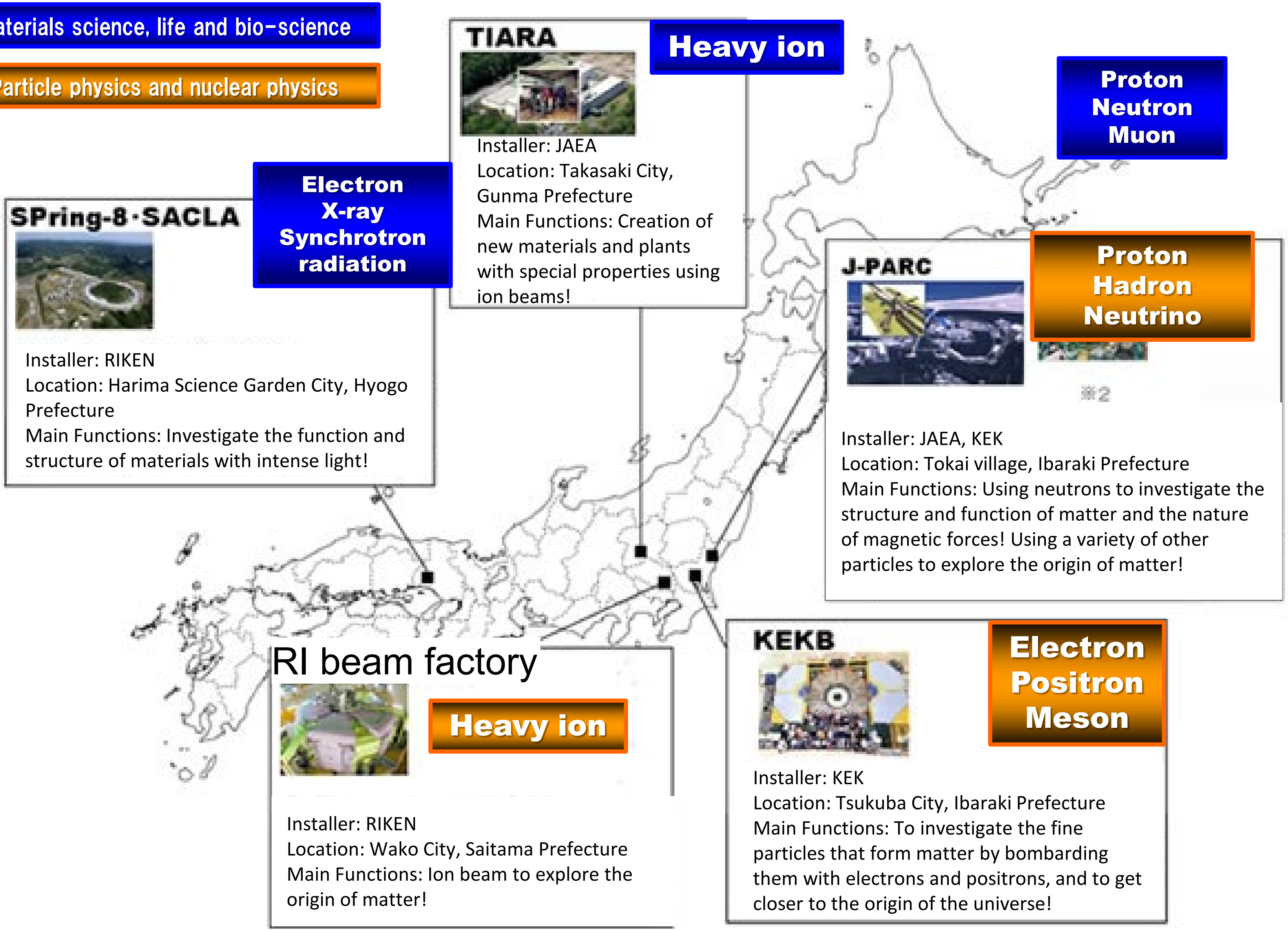
Cancer treatment

Japan’s large quantum beam facilities (Accelerators)

http://www.mext.go.jp/a_menu/shinkou/ryoushi/

Materials science, life and bio-science

Particle physics and nuclear physics



TIARA

Heavy ion

Installer: JAEA
Location: Takasaki City, Gunma Prefecture
Main Functions: Creation of new materials and plants with special properties using ion beams!

Proton
Neutron
Muon

SPring-8·SACLA

Electron
X-ray
Synchrotron radiation

Installer: RIKEN
Location: Harima Science Garden City, Hyogo Prefecture
Main Functions: Investigate the function and structure of materials with intense light!

J-PARC

Proton
Hadron
Neutrino

※2

Installer: JAEA, KEK
Location: Tokai village, Ibaraki Prefecture
Main Functions: Using neutrons to investigate the structure and function of matter and the nature of magnetic forces! Using a variety of other particles to explore the origin of matter!

RI beam factory

Heavy ion

Installer: RIKEN
Location: Wako City, Saitama Prefecture
Main Functions: Ion beam to explore the origin of matter!

KEKB

Electron
Positron
Meson

Installer: KEK
Location: Tsukuba City, Ibaraki Prefecture
Main Functions: To investigate the fine particles that form matter by bombarding them with electrons and positrons, and to get closer to the origin of the universe!

Location of LINAC facility at Hokkaido University

Faculty of Engineering, Hokkaido University
Electron Accelerator and Neutron
Experimental Facility

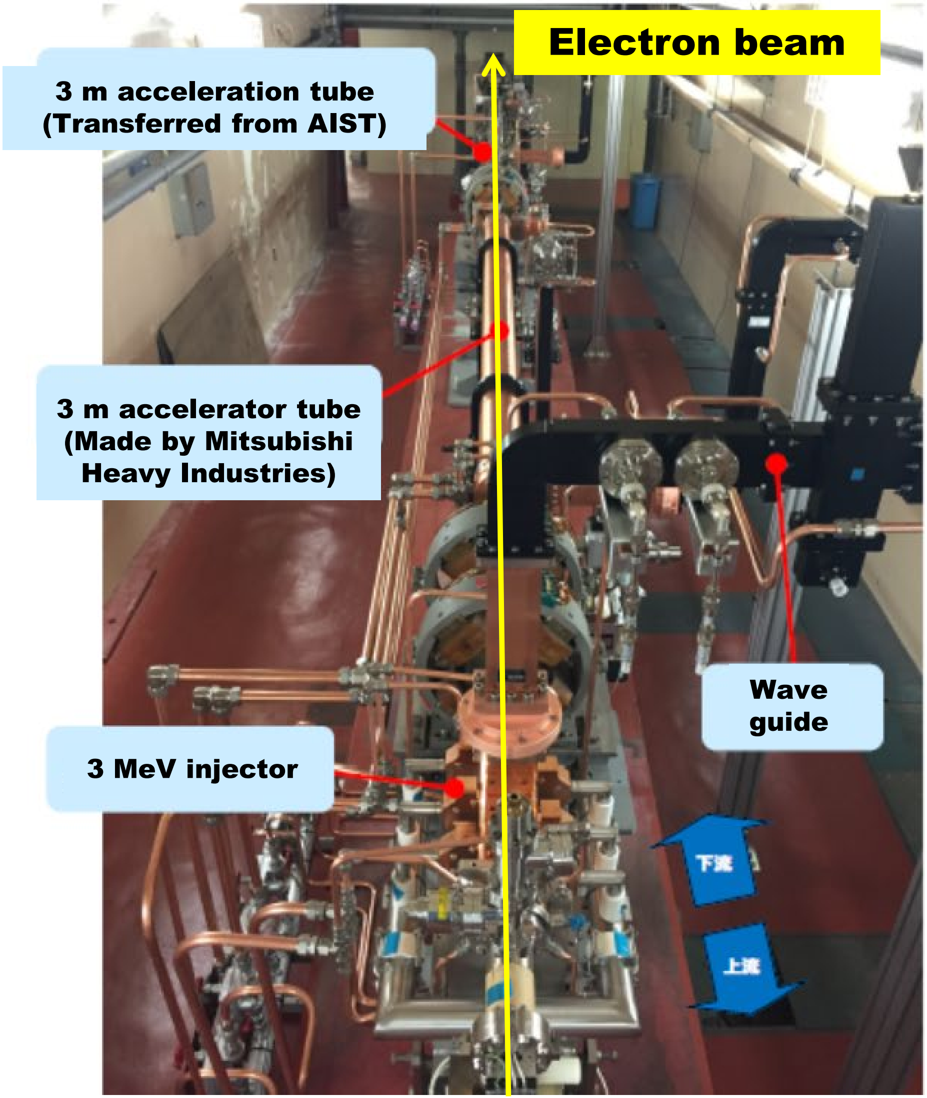
Exterior view of the LINAC facility at Hokkaido University

One of the leading radiation controlled areas on campus. One of the largest accelerator facilities on campus, along with the Proton Therapy Center at Hokkaido University Hospital.

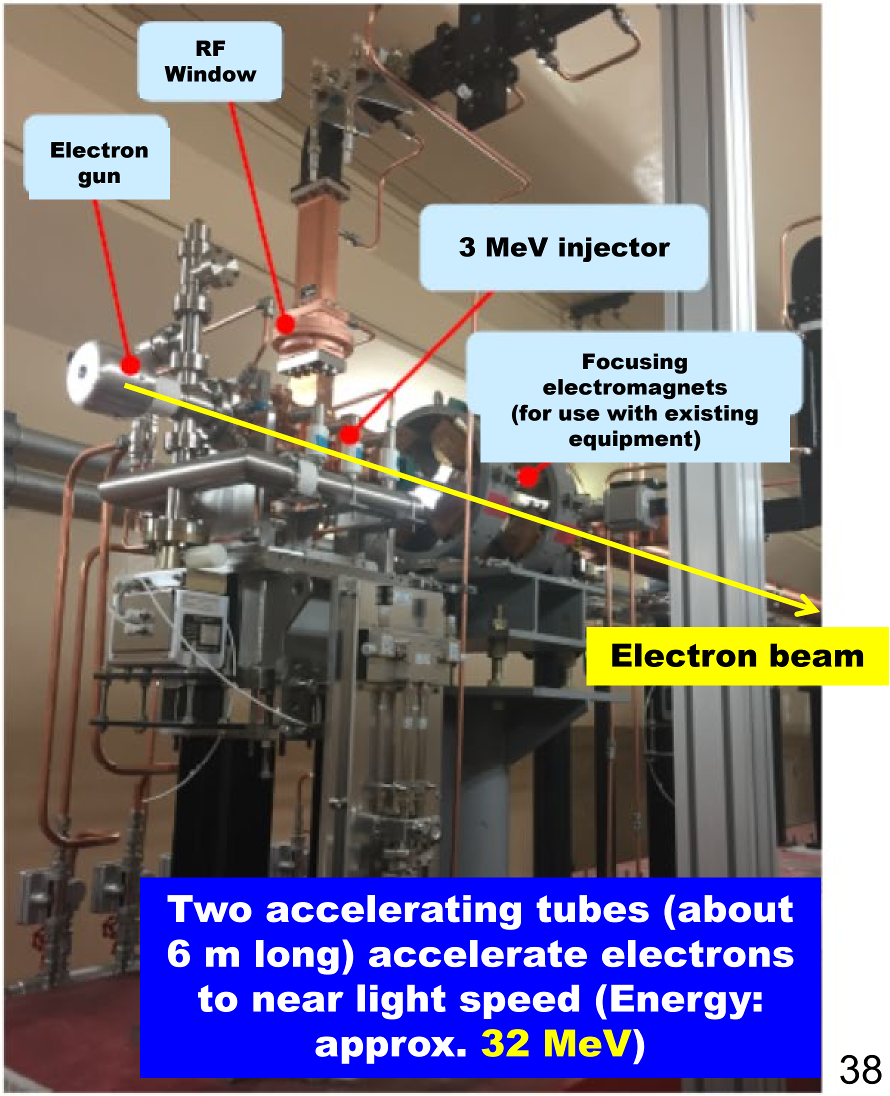
Control room in the basement: Accelerator operation, remote control of experimental equipment, meetings, etc.

Electron LINAC（Linear Accelerator）: Electron linear accelerator

Overhead view of accelerator room



Around the injector

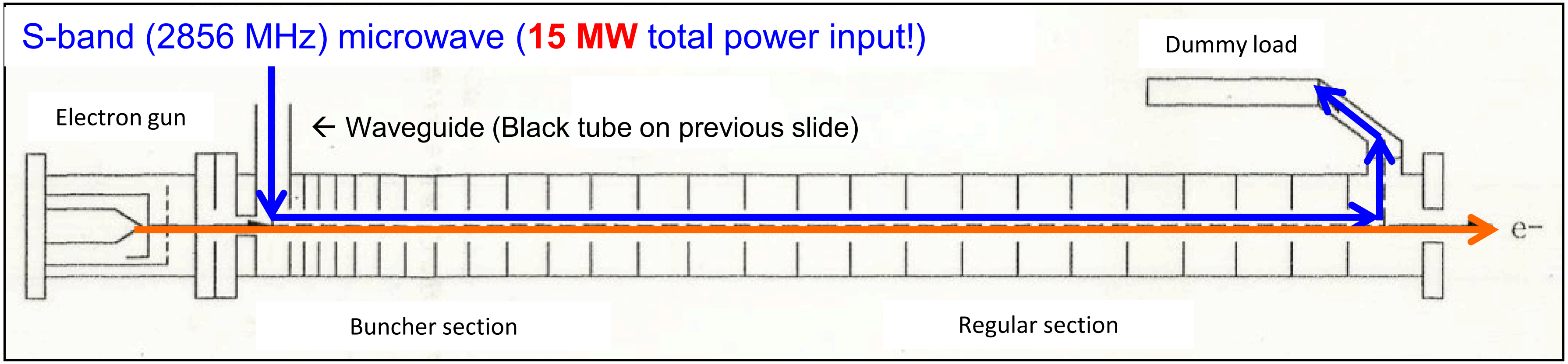


Two accelerating tubes (about 6 m long) accelerate electrons to near light speed (Energy: approx. 32 MeV)

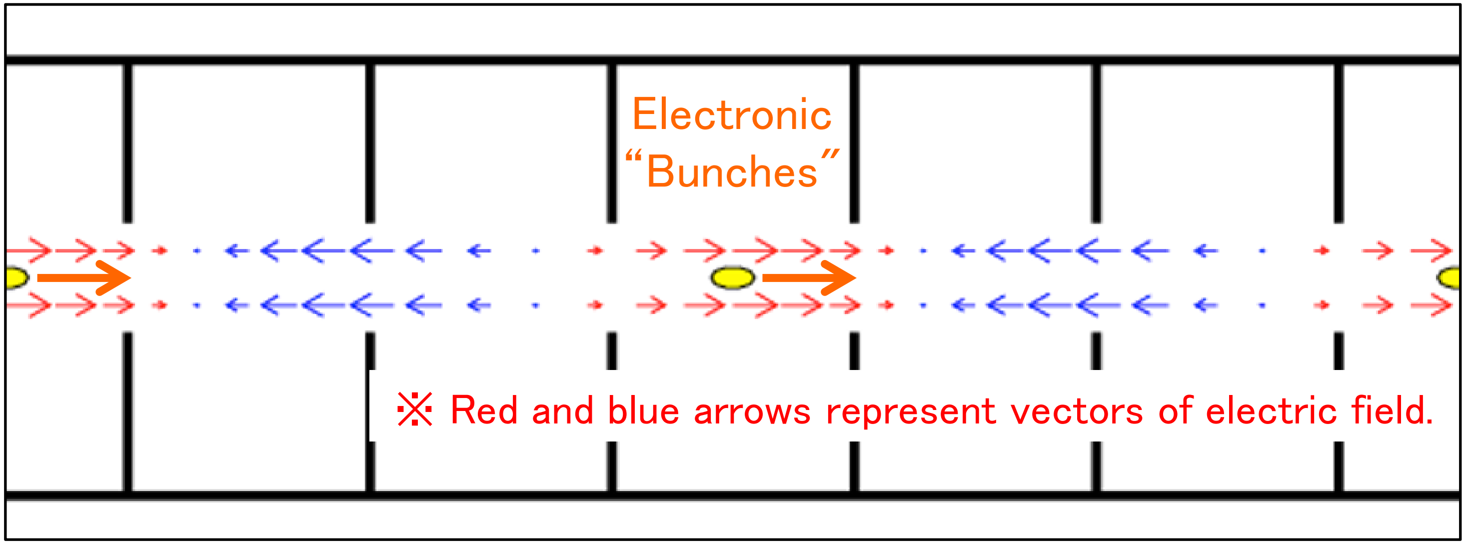
Principle of electron acceleration

(Difficult to do with an electrostatic field) A method to generate an electric field with a potential difference of 32 MV.

Utilizes the fact that electrons interact with electric and magnetic fields. Uses electromagnetic waves（Radio waves（Microwaves））.


S-band (2856 MHz) microwave (**15 MW** total power input!)

A traveling microwave wave is created in the accelerator tube. Electrons ride the traveling wave.



※ Red and blue arrows represent vectors of electric field.

Electronic LINAC performance: Summary

Overall performance (Current)

Electron kinetic energy: 32 MeV

- Electron beam time-averaged current: 50 μA
- Electron beam power: 1.6 kW (= Energy x Current)

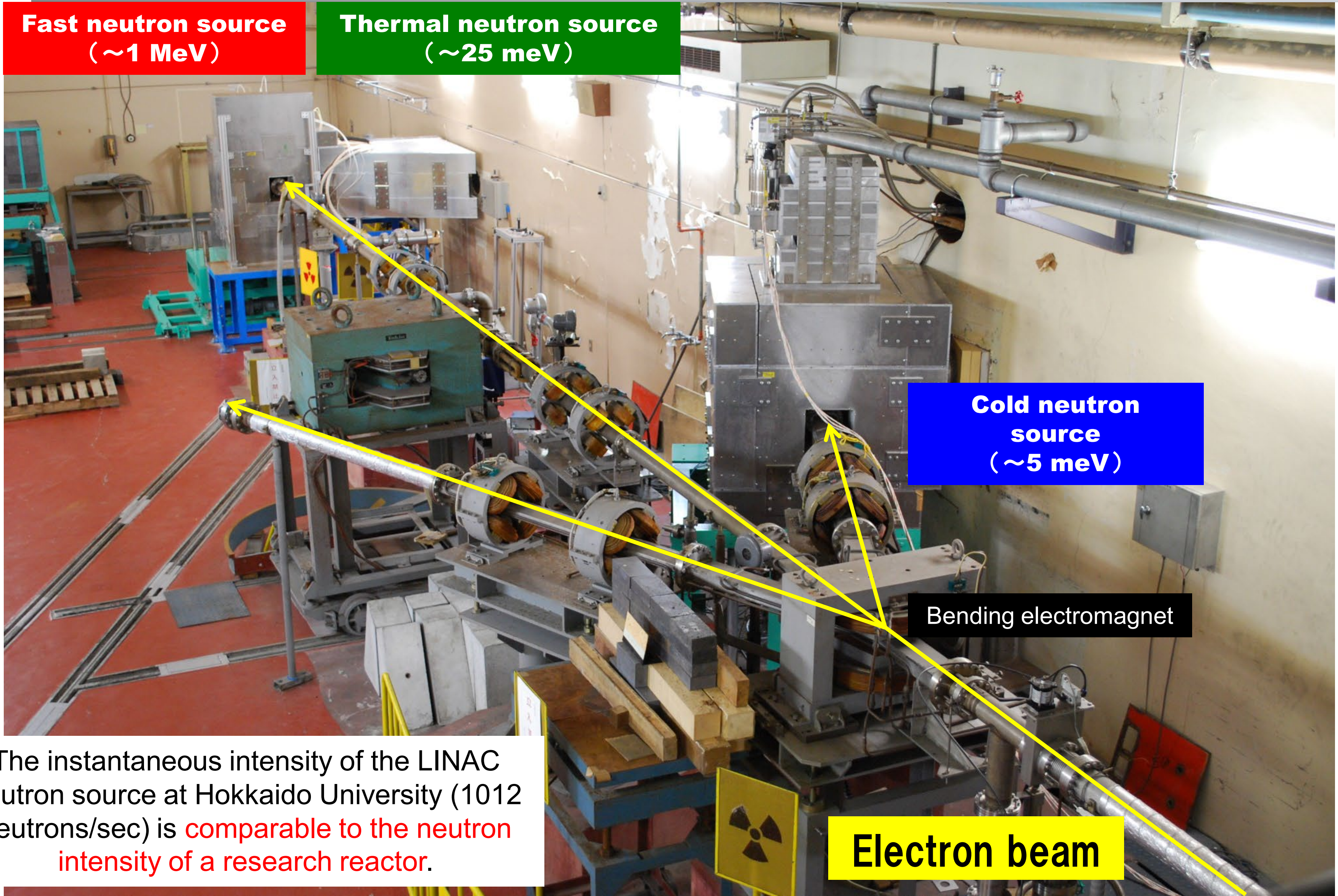
Performance of pulse operation (Current)

- Electron pulse width: 4 μs
- Pulse repetition rate: 50 pulses/second

At the maximum, the power (Current value) is doubled!

Annual operating performance: approx. 160 days

Using high-energy and high-intensity electron beams, a large number of neutrons are produced by nuclear reactions!



Fast neutron source
（~1 MeV）

Thermal neutron source
（~25 meV）

Cold neutron source
（~5 meV）

Bending electromagnet

Electron beam

The instantaneous intensity of the LINAC neutron source at Hokkaido University (1012 neutrons/sec) is comparable to the neutron intensity of a research reactor.

Why do we need an accelerator?

- ✓ Different types of radiation interact with matters with different reaction cross sections, depending on their energy.
- ✓ The structure and motion of matters can be investigated by analyzing various particles that cause various interactions.

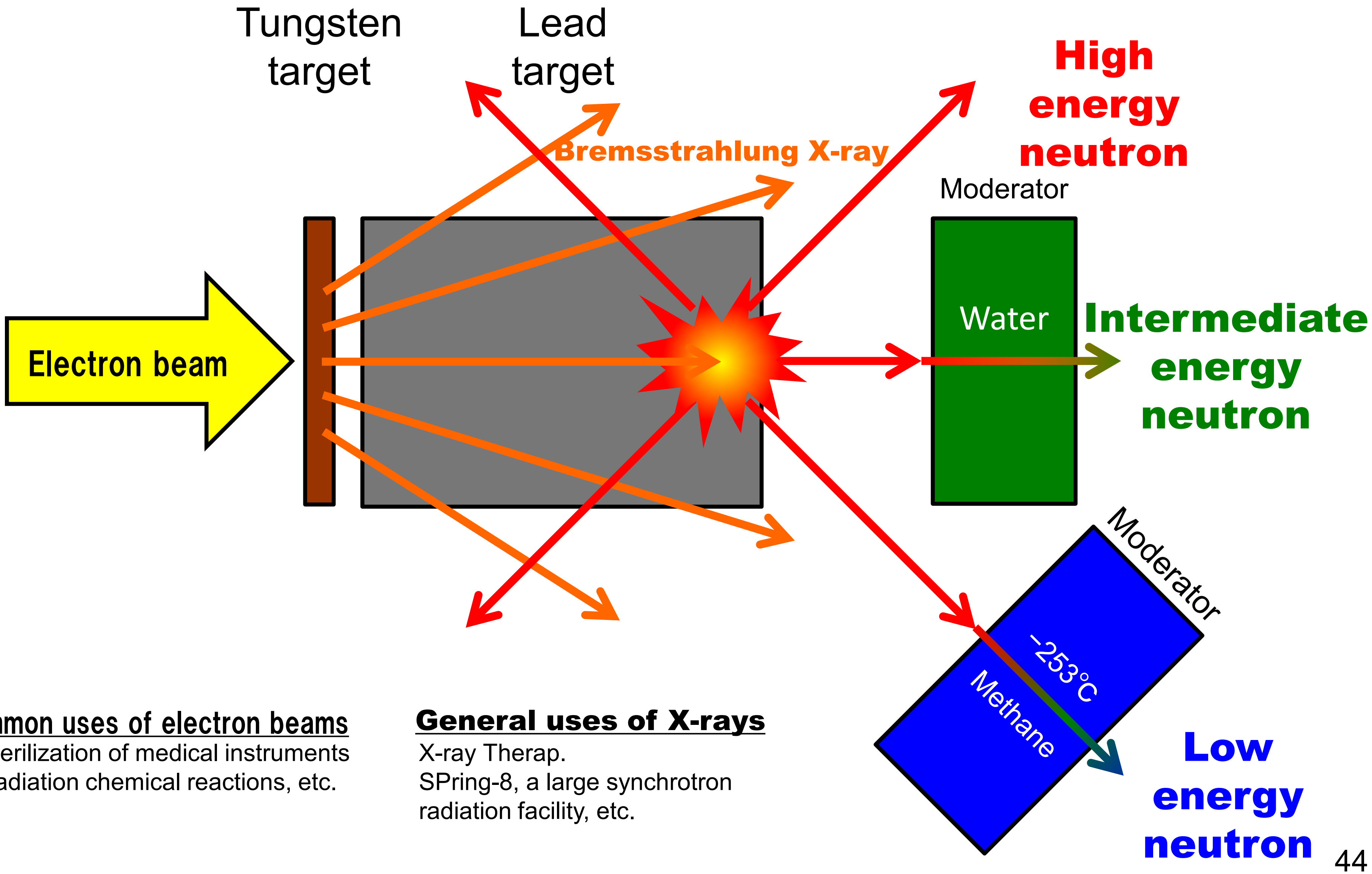
↓

- ✓ The use of primary particles with higher energy (Acceleration) will expand the range of applications for “utilizing a wide energy band (Electron beams and X-rays)” and “efficiently producing secondary and tertiary particles (Electron → X-ray → Neutron･Positron, Proton → Neutron).
- ✓ In order to "capture interactions in a shorter time" and "capture smaller reactions," the incident beam intensity must be increased.

- ● **In terms of controlling the type, energy, intensity, and shot time of radiation, accelerators are effective in meeting the above conditions.**
- ● Large accelerator facilities are designed for high intensity and high energy.
- ● Small accelerator facilities are designed for ease of use and popularization.

Particle accelerator-based neutron sources (Focusing on pulsed neutron sources)

Neutron generation and energy adjustment method (When the primary beam is an electron beam)



Tungsten target

Lead target

Bremsstrahlung X-ray

High energy neutron

Moderator

Water

Intermediate energy neutron

Electron beam

Moderator

−253°C

Methane

Low energy neutron

Common uses of electron beams

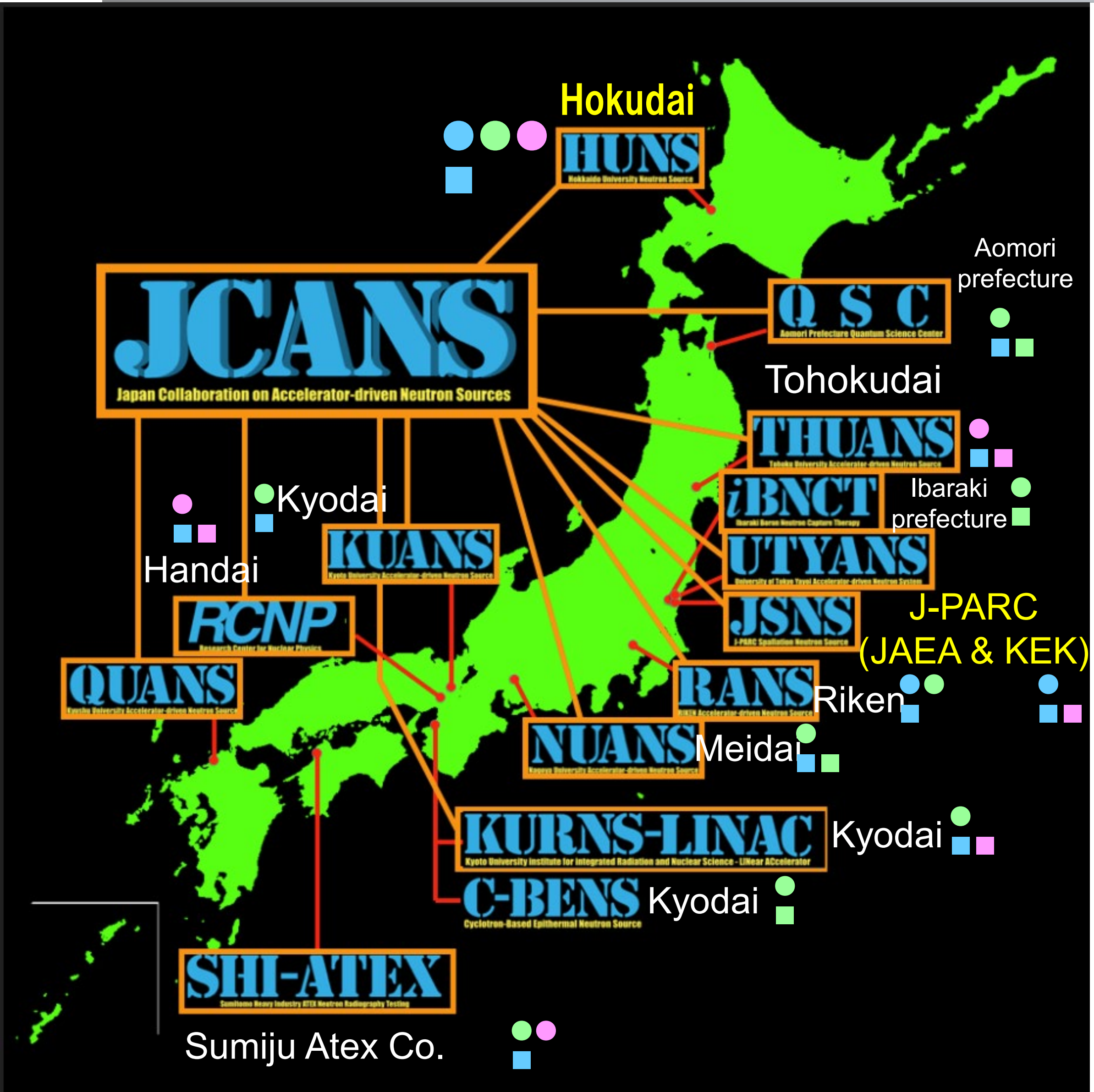
- Sterilization of medical instruments
- Radiation chemical reactions, etc.

General uses of X-rays

- X-ray Therap.
- SPring-8, a large synchrotron radiation facility, etc.

Major accelerator-driven neutron experimental facilities in Japan

Image from the website of the Japan Council of Accelerator Neutron Sources (JCANS)



● Cold neutron source

● Thermal neutron source / Epithermal neutron source

● Fast neutron source

■ Materials, life and industrial applications

■ Medical sciences

■ Particle and nuclear physics

Other,

- **National Institute of Advanced Industrial Science and Technology (AIST)** ● ■
- Other, accelerator neutron sources
- Other, reactor neutron sources
- Other, laser neutron source
- Other, RI neutron source
- …

Various neutron sources and their applications in universities, national laboratories, and private companies

Major pulsed neutron imaging facilities in Japan

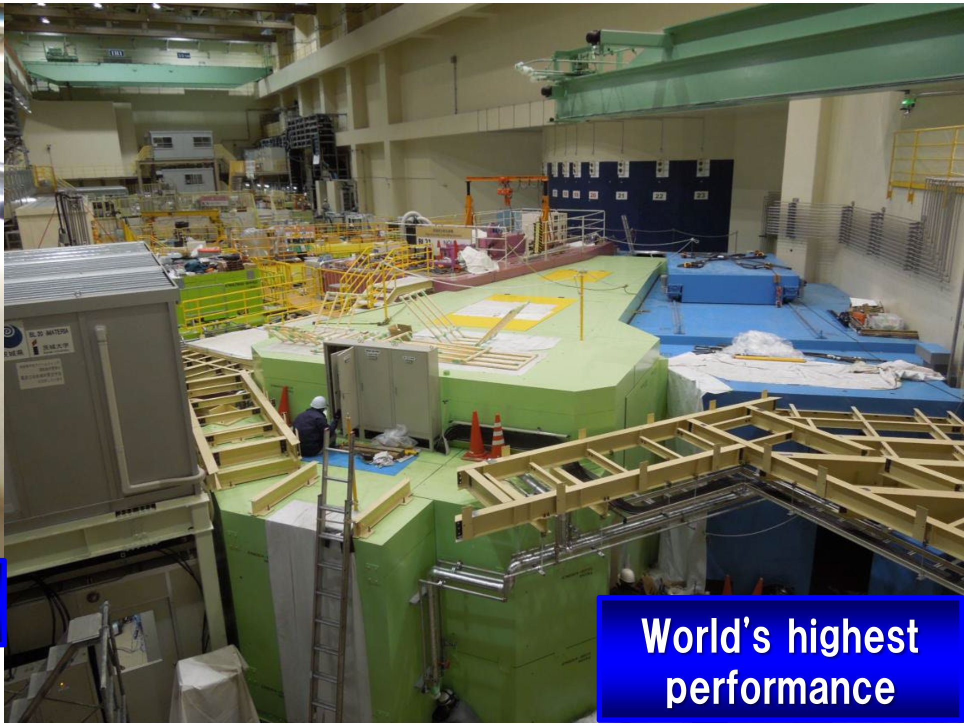
Faculty of Engineering, Hokkaido University HUNS

On demand

https://www.eng.hokudai.ac.jp/labo/QBMA/**LINAC**/

Neutron flux (max): 10^4 n/cm²/s
Wavelength resolution (min):1%
Wavelength bandwidth (max): < 1.3 nm
Beam size (max): 10 cm x 10 cm
Beam divergence angle: 1/60 rad

J-PARC MLF BL22 RADEN（Mother-of-pearl）



World's highest performance

Neutron flux (max): 10^8n/cm²/s
Wavelength resolution (min): 0.20
Wavelength bandwidth (max): < 1.76 nm
Beam size (max): 30 cm x 30 cm
Beam divergence (min): 1/7500 rad

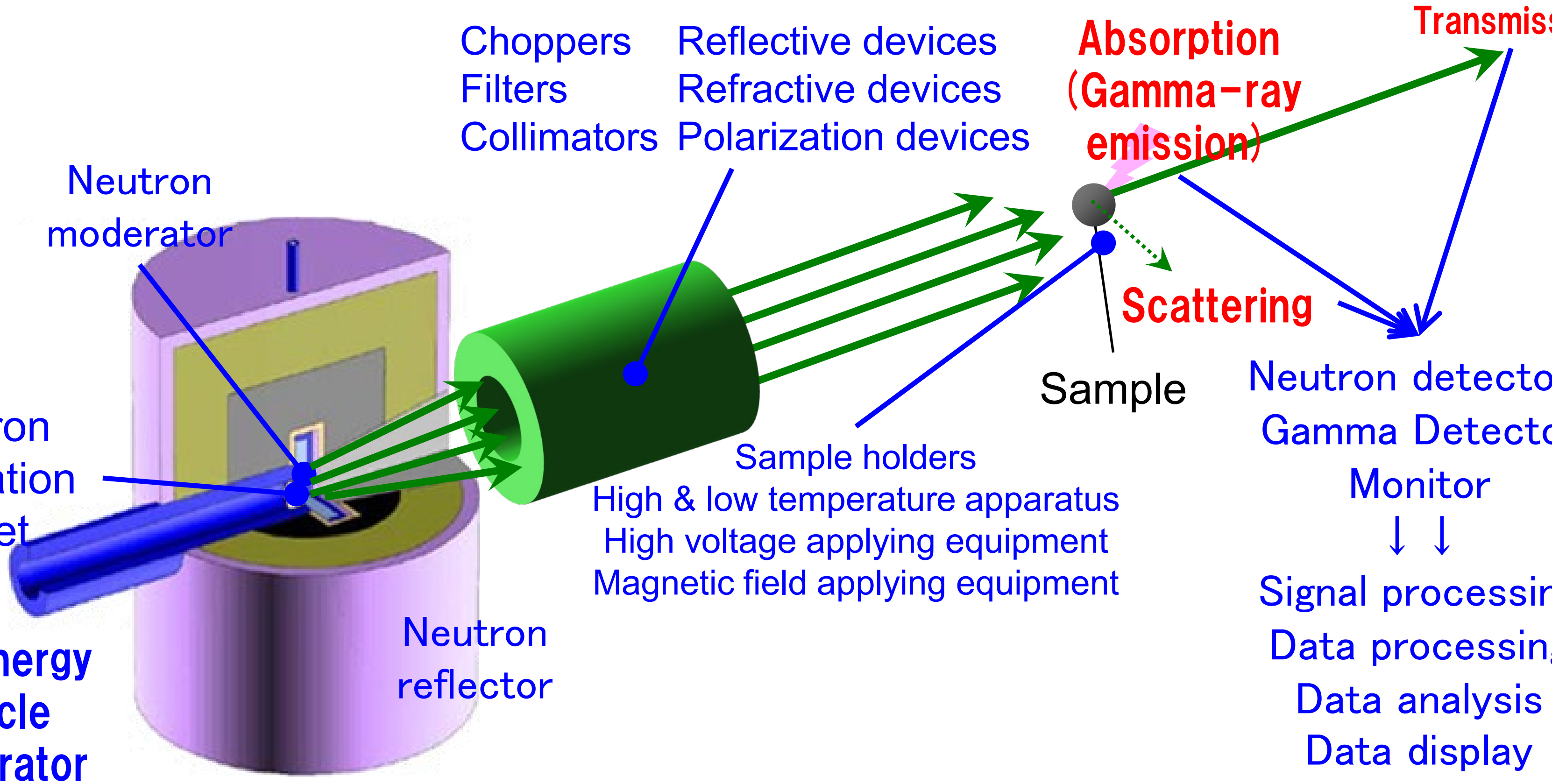
In addition to these, **AISTANS** is being maintained at AIST.

Neutron beamline configuration

Neutron generation

Neutron transport

Neutron utilization



Neutron moderator

Neutron generation target

High energy particle accelerator (Proton and Electron)

Neutron reflector

Choppers
Filters
Collimators

Reflective devices
Refractive devices
Polarization devices

Sample holders
High & low temperature apparatus
High voltage applying equipment
Magnetic field applying equipment

Absorption (Gamma-ray emission)

Transmission

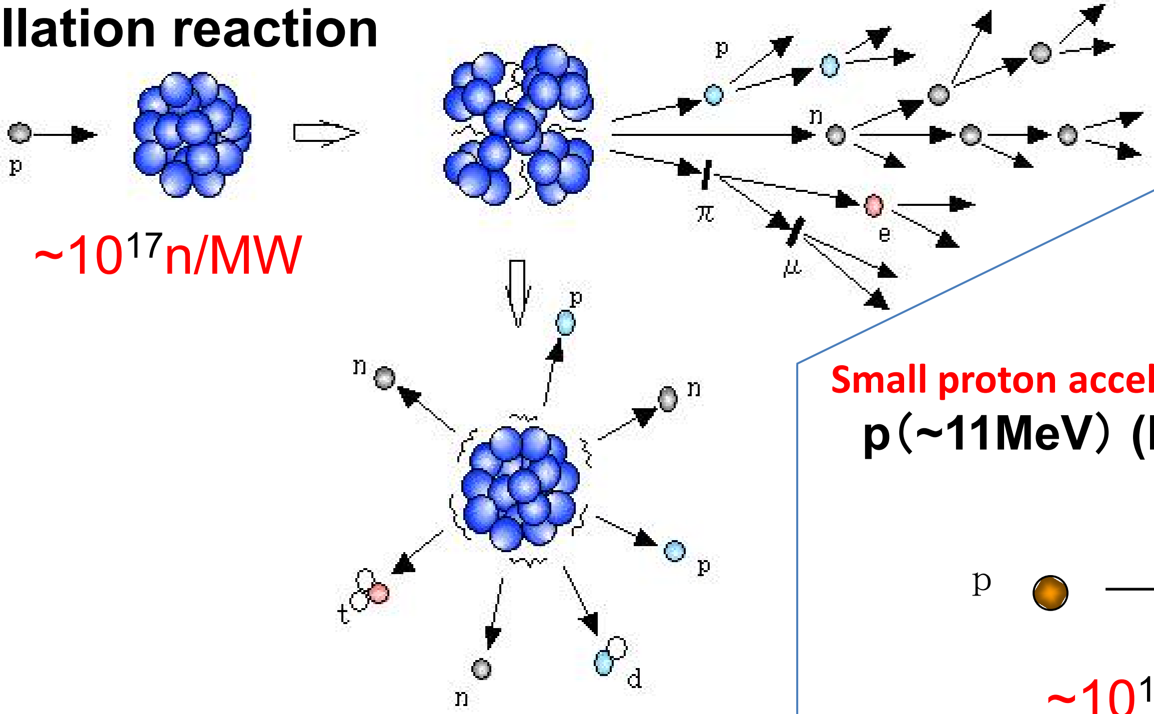
Scattering

Sample

Neutron detectors
Gamma Detector
Monitor
↓ ↓
Signal processing
Data processing
Data analysis
Data display

Large proton accelerator (more than several hundred MeV)

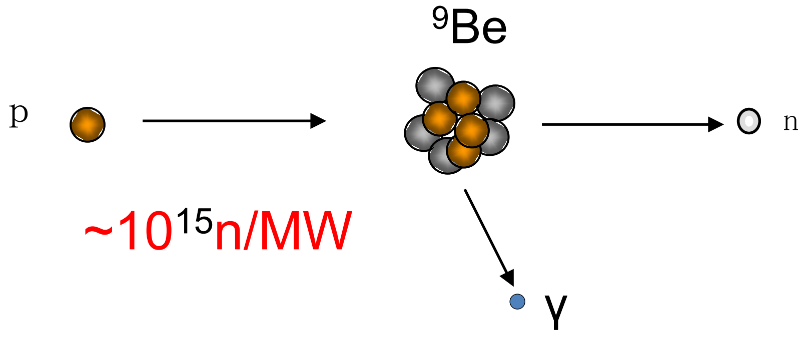
Spallation reaction



~10^{17}n/MW

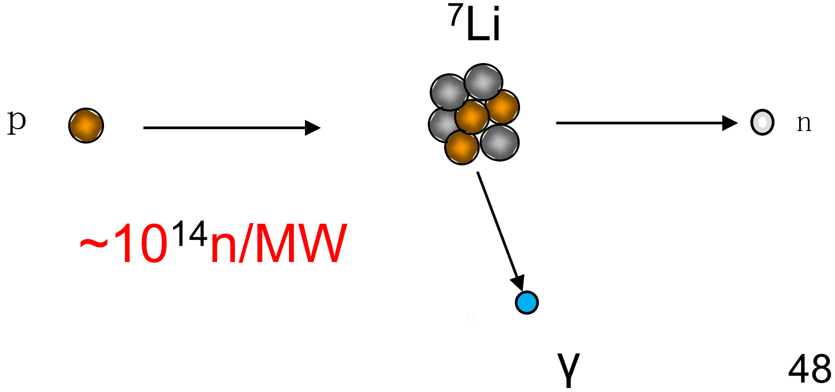
Small proton accelerators (up to several tens of MeV)

p（~11MeV）(Be,n) reaction



~10^{15}n/MW

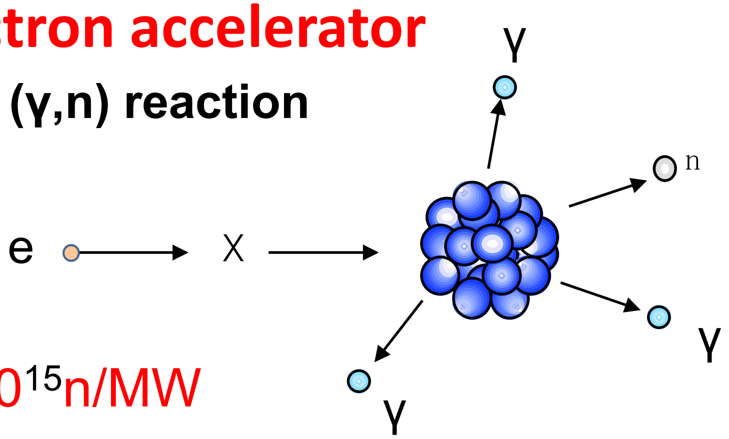
p（~3MeV）(Li,n) reaction



~10^{14}n/MW

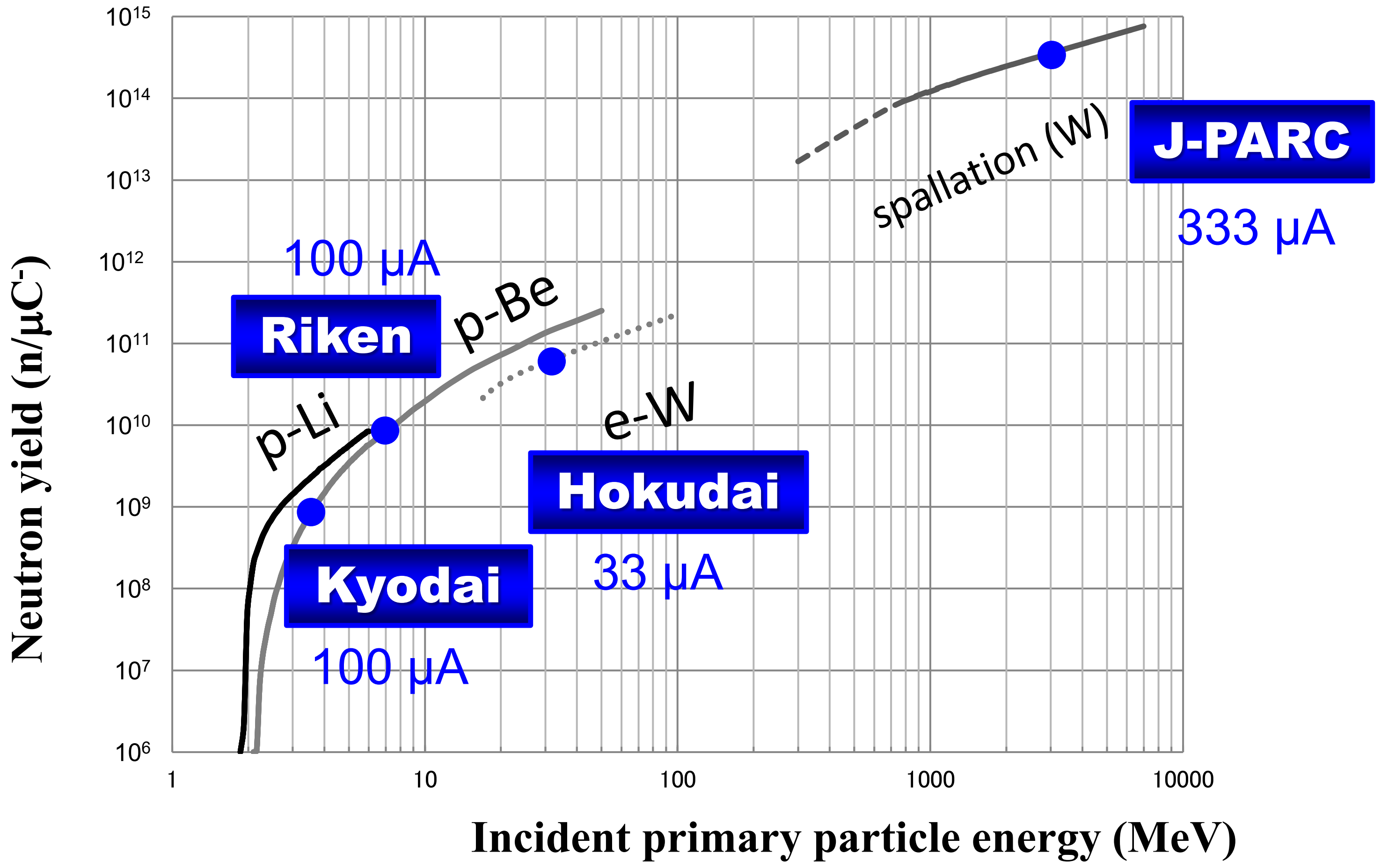
Electron accelerator

(e,γ) (γ,n) reaction



~10^{15}n/MW

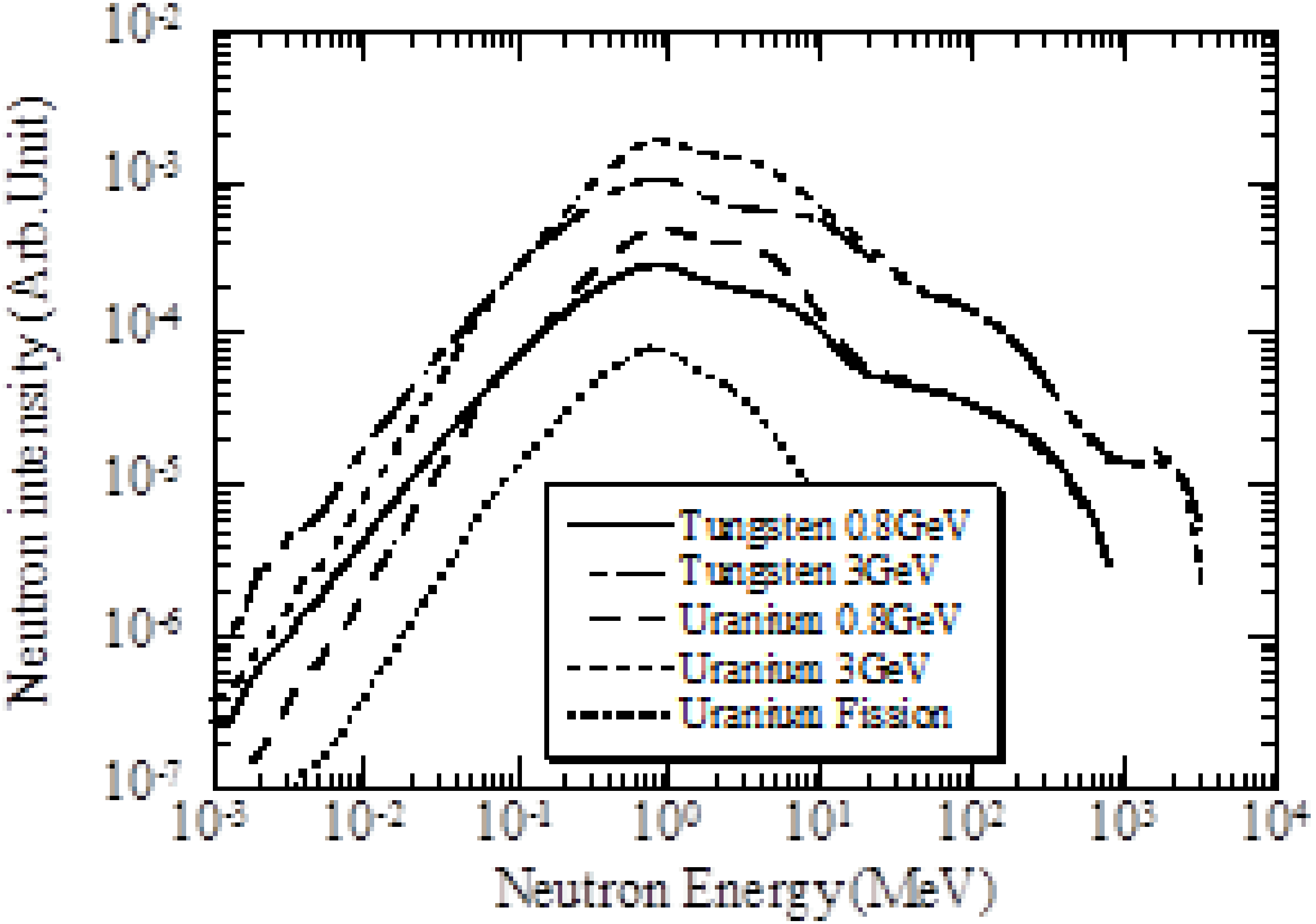
Energy dependence of the number of neutrons produced per number of primary particles for each reaction



The appropriate neutron-production reaction depends on the specifications of the accelerators

Neutron energy spectrum

Generated neutron spectrum peaks at MeV



However, the energy required by material and life sciences is meV.

HUNS neutron source moderator system

Electron LINAC (Linear Accelerator)

- 33 MeV × 33 μA = 1 kW
- electron pulse width ：3 μs
- electron pulse repetition ：50 Hz

Billiard“ collisions with **hydrogen nuclei**, which are closest in mass to the neutron.

Neutron generation

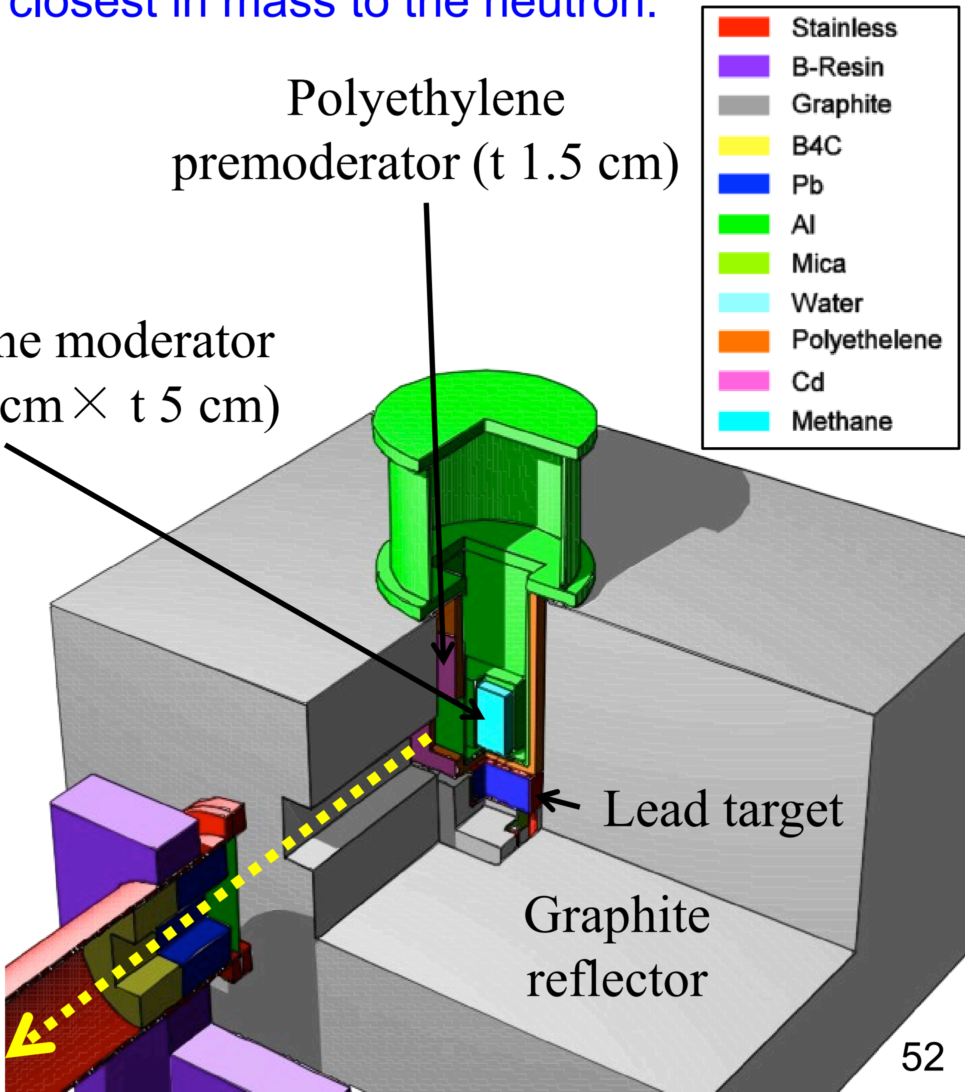
- Generation of synchrotron radiation X-rays by bremsstrahlung of electron beams
- Photonuclear reaction
- Target material: Pb
- 1.6×10^{12} per second

Neutron moderator

- Reflector-coupled moderator
  → Polyethylene pre-moderator
- 18 K solid methane moderator
- Cold neutron pulse width ：160 μs

Neutron reflector

- Graphite（$80 \times 80 \times 80$ cm^3）



Polyethylene premoderator (t 1.5 cm)

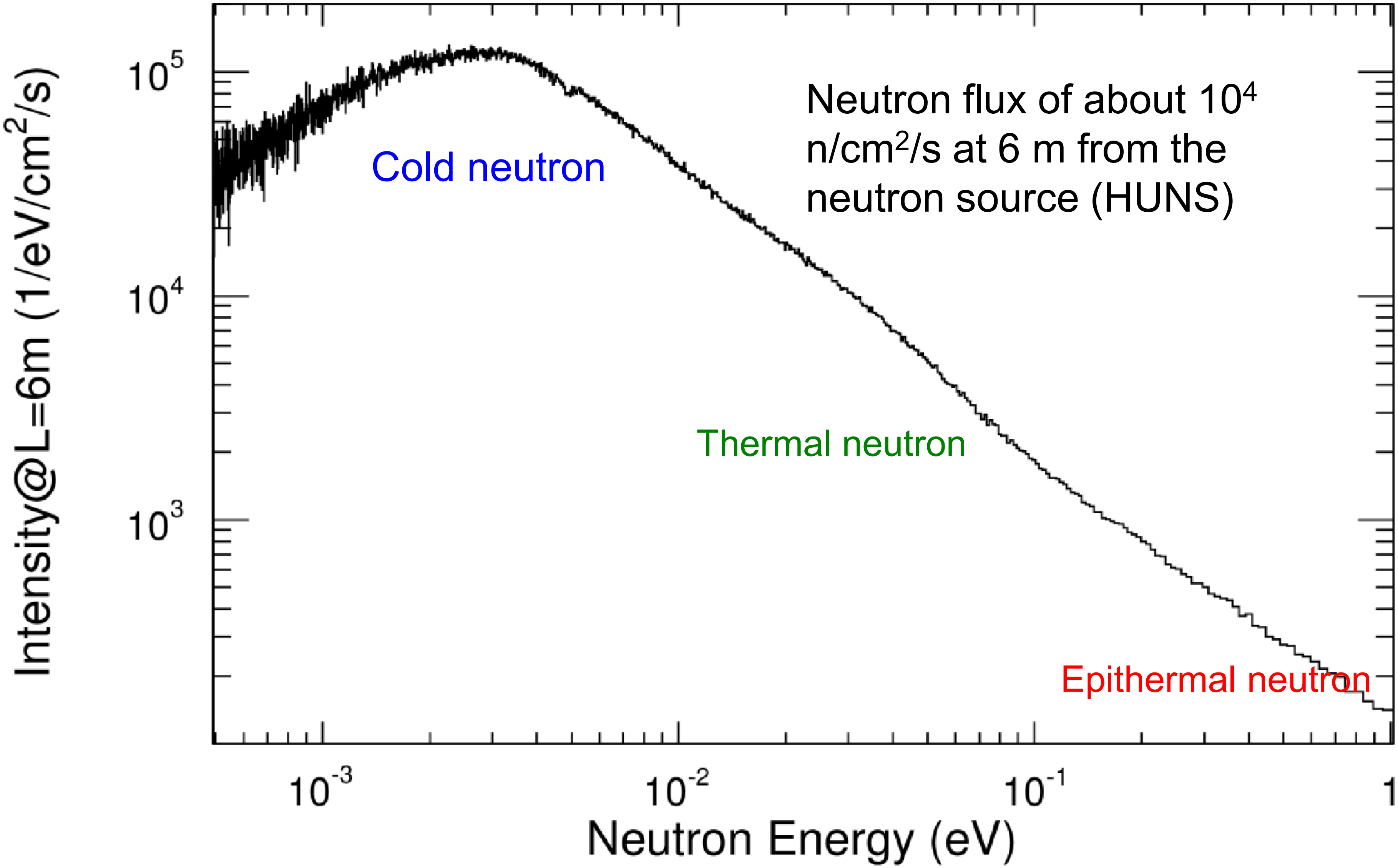
Solid methane moderator (12 cm × 12 cm × t 5 cm)

Lead target

Graphite reflector

10 cm × 10 cm beam extraction

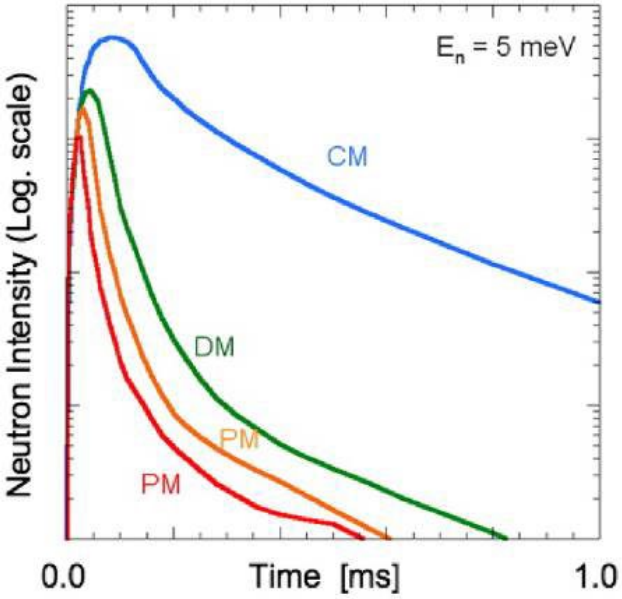
Energy spectrum of supplied neutrons



Neutron flux of about 10^4 n/cm^2/s at 6 m from the neutron source (HUNS)

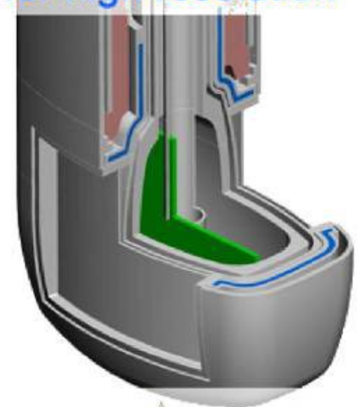
Neutron transport and detection

Neutron pulse shape changed by the type of neutron moderator (using J-PARC as an example)



Poisoned Decoupled moderator (PM)

for high resolution



Decoupled moderator(DM)

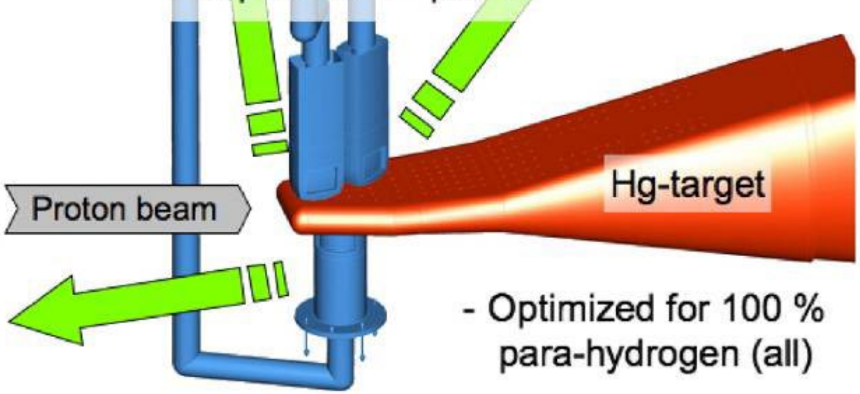
for balanced performance

- Adoption of Ag-In-Cd (AIC) alloy for high decoupling energy at 1 eV
- optimized decouple coverage for lower pulse tail
- Adoption of Cd poison

Coupled moderator (CM)

for high intensity

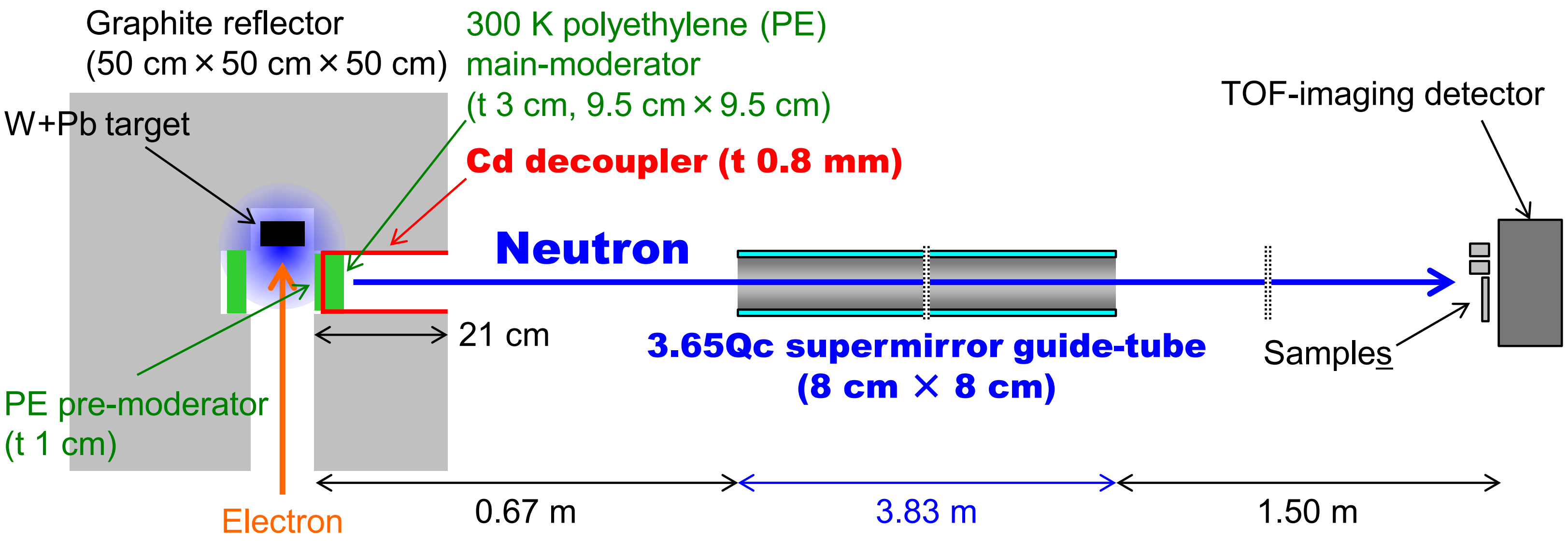
- large & cylindrical
- wide angle beam extraction

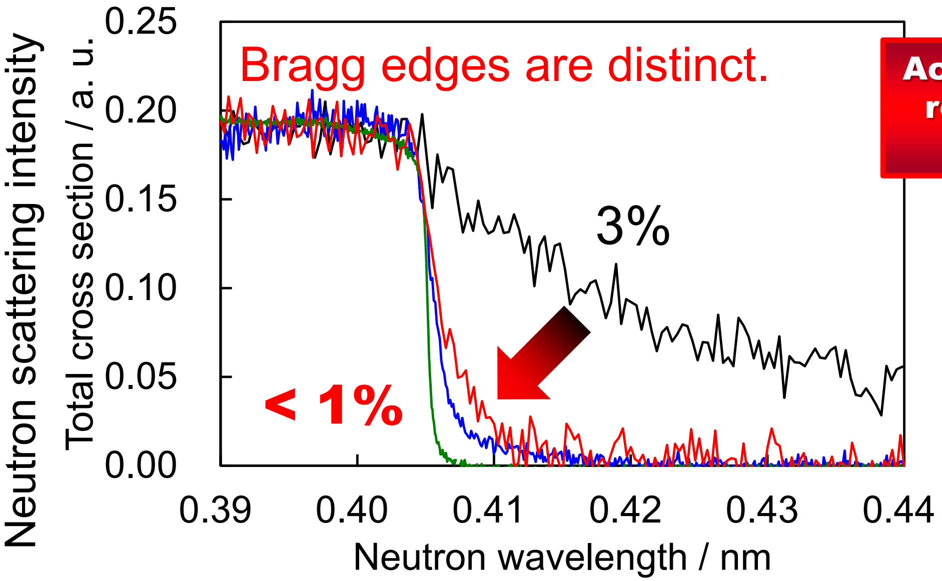


- Optimized for 100 % para-hydrogen (all)

Short pulse neutron source & neutron guide tube for higher performance HUNS instruments

H. Sato *et al.*, Phys. B **551** (2018) 452.

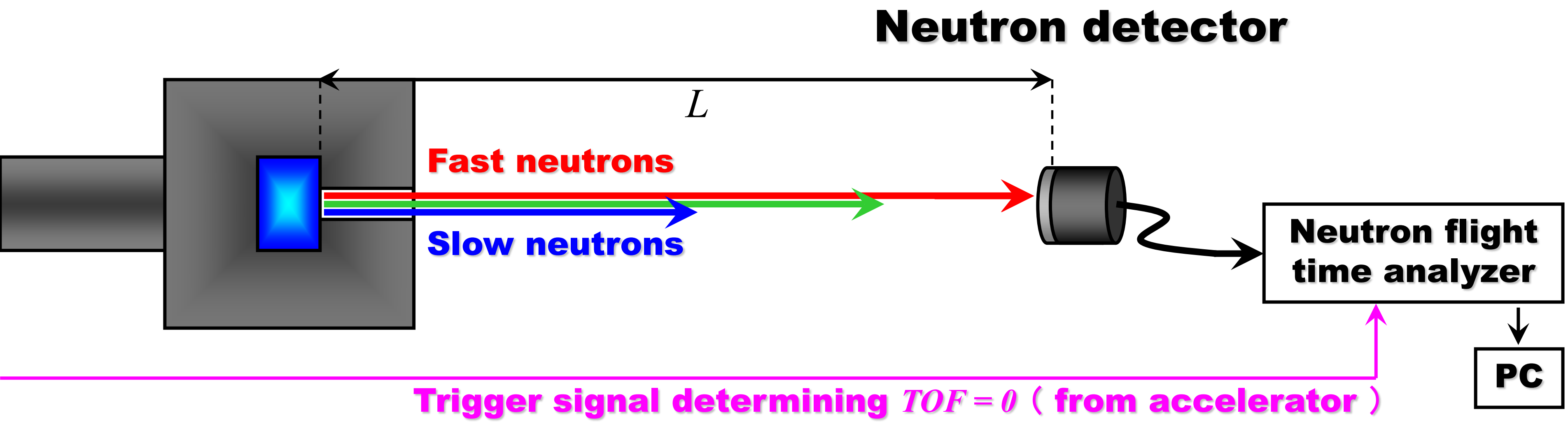




Achievement of wavelength resolution comparable to that of J-PARC

Improvement of research capability at Hokkaido University

Accelerator <u>pulsed</u> neutron source allows velocity (energy) analysis by time-of-flight (TOF) method



Neutron detector

L

Fast neutrons

Slow neutrons

Neutron flight time analyzer

PC

Trigger signal determining $TOF = 0$（ from accelerator ）

$$E=\frac{1}{2}mv^2=\frac{1}{2}m\left(\frac{L}{TOF}\right)^2=\frac{1}{2m}\left(\frac{h}{\lambda}\right)^2$$

m : Mass of neutron

v : Velocity of neutron

L : Path Length from source to detector

TOF : Elapse time from neutron generation

h : Plank’s constant

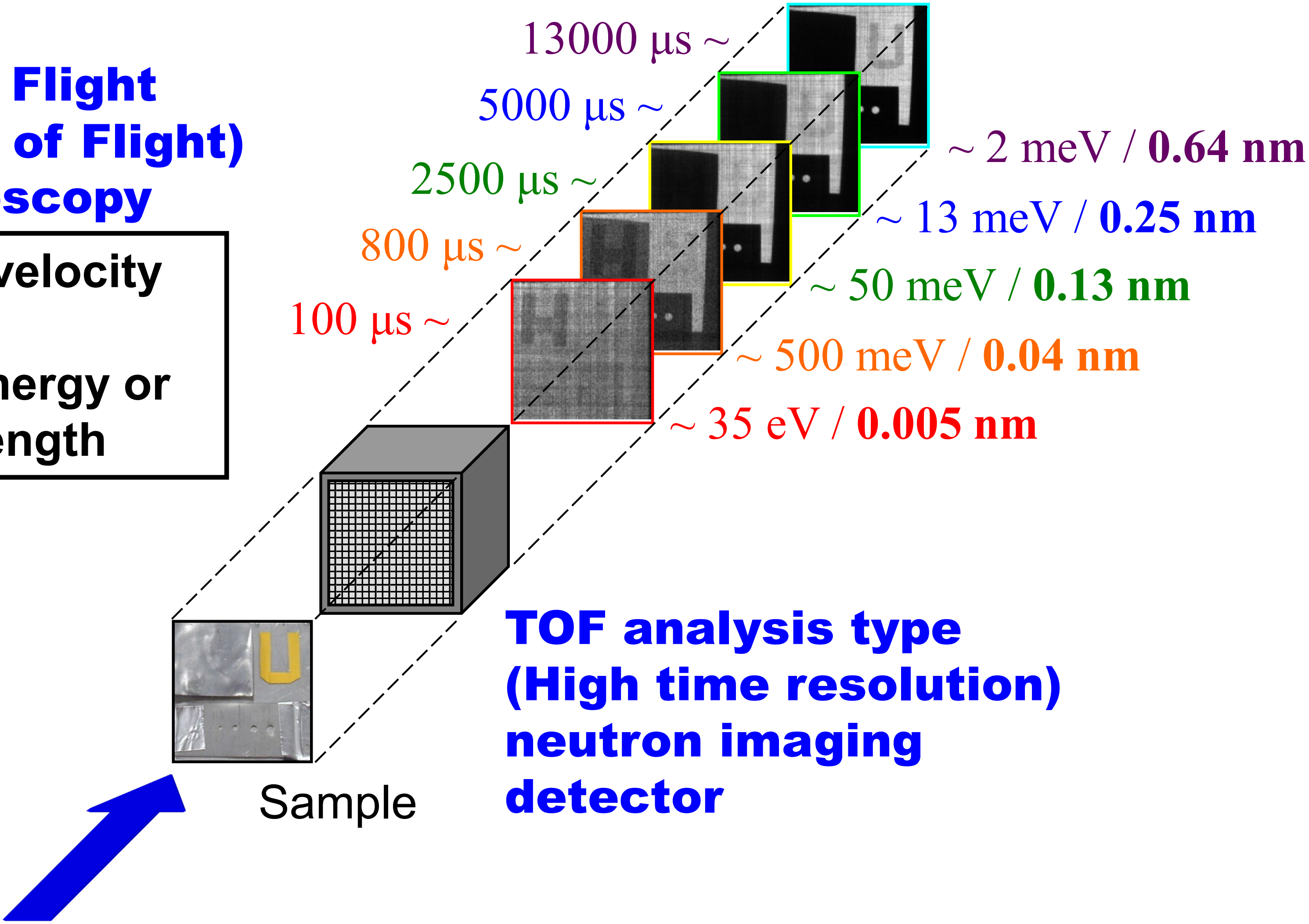
λ : Neutron wavelength

Because it's pretty slow, it can be controlled mechanically.

Wavelength-resolved neutron transmission imaging using accelerator pulsed neutron source and TOF method

Time of Flight
（TOF: Time of Flight)
spectroscopy

Neutron velocity
↓
Kinetic energy or wavelength

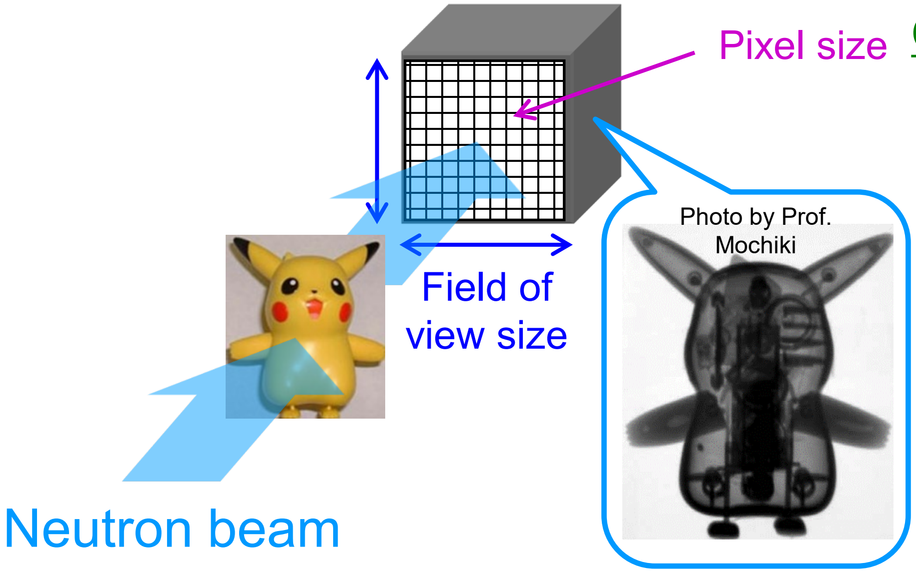


Sample

TOF analysis type (High time resolution) neutron imaging detector

Pulsed white neutron beams

Performance of TOF analytical neutron imaging detectors



GEM-type neutron imaging detector

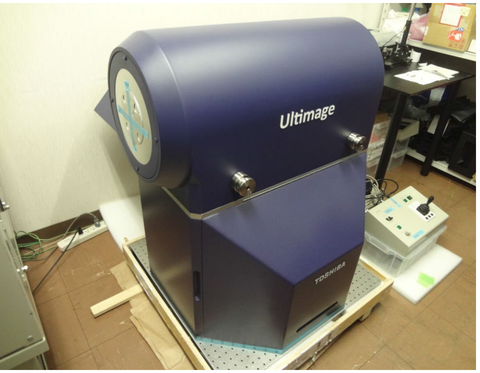
KEK Uno group

Pixel size: 800 μm

Field of vision: 10 cm × 10 cm

Mid-range spatial resolution
Large area
High reliability

High-speed camera type neutron image detector



Tokyo City University Mochiki group

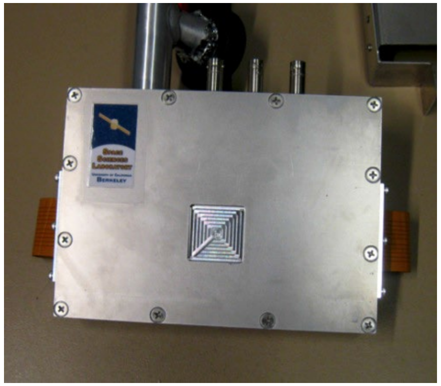
Pixel size: 520 μm

Field of vision: 13 cm × 13 cm

High spatial resolution
maximum area

Neutron color image intensifier (Toshiba) +
High-speed CMOS video camera (NAC)

MCP type neutron image detector



UC Berkeley
Dr. A. S. Tremsin

Pixel size: 55 μm

Field of vision: 2.8 cm × 2.8 cm

Ultra-high spatial resolution

Major reactions for neutron conversion & detection

$n + {}^{3}\mathrm{He}$ (0.0001% / 5333 barn) → ${}^{3}\mathrm{H} + p$

Isotope abundance　Thermal neutron absorption cross-section

$n + {}^{6}\mathrm{Li}$ (7.5% / 940 barn) → ${}^{3}\mathrm{H} + \alpha$

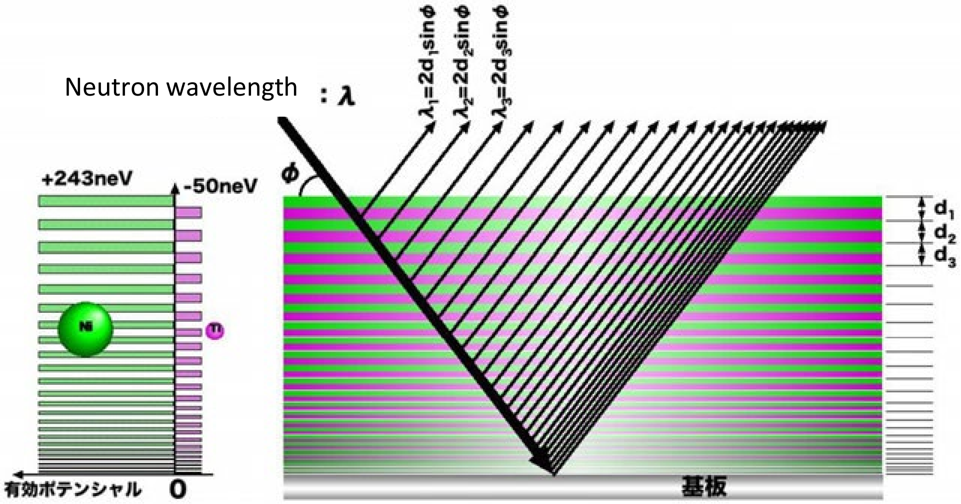
$n + {}^{10}\mathrm{B}$ (20% / 3835 barn) → ${}^{7}\mathrm{Li} + \alpha$

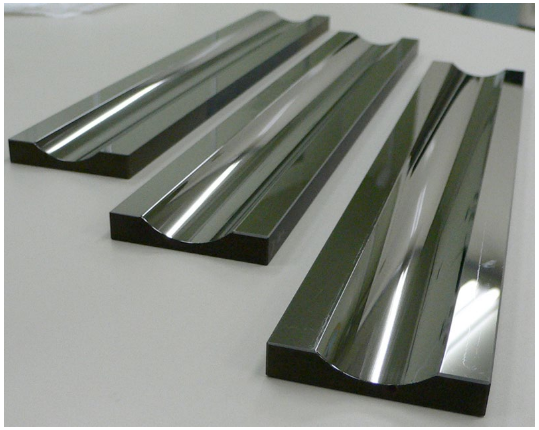
$n + {}^{155}\mathrm{Gd}$ (15% / 61100 barn) → ${}^{156}\mathrm{Gd} + \gamma + e^{-}$ (IC)

$n + {}^{157}\mathrm{Gd}$ (16% / 259000 barn) → ${}^{158}\mathrm{Gd} + \gamma + e^{-}$ (IC)

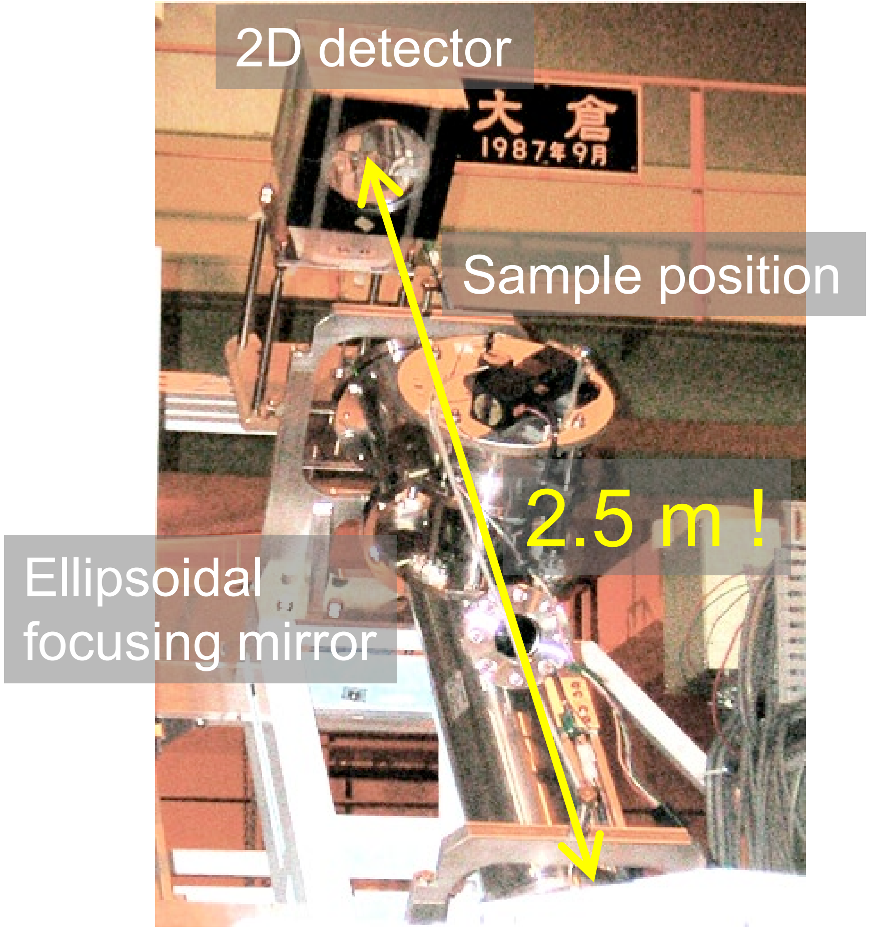
Neutron supermirror

Reflection of neutrons over a wide wavelength range by Ni-Ti multilayers



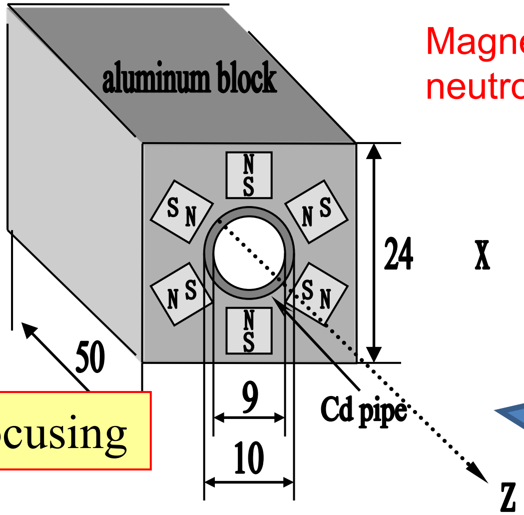


Neutron focusing and miniaturization of neutron focusing and miniaturization of equipment



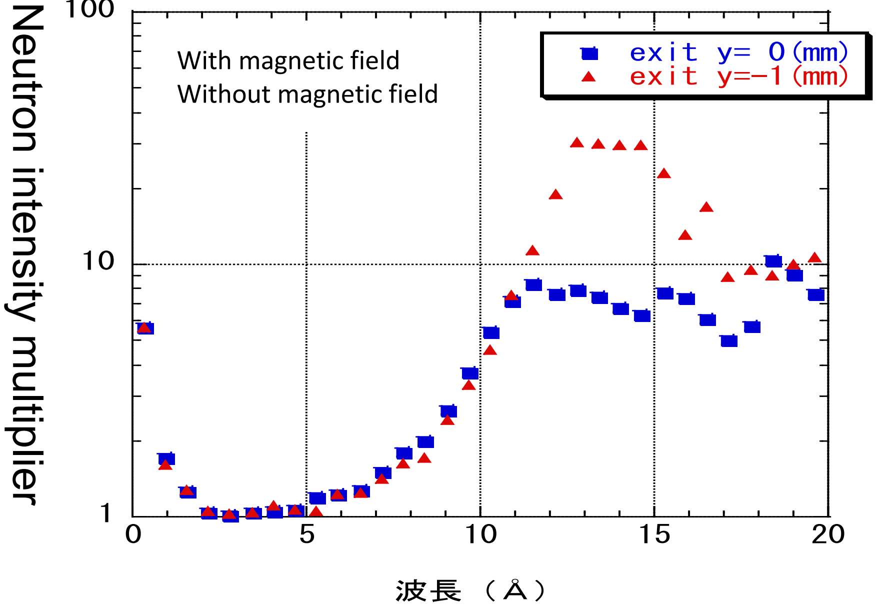
Nagneto-neutron lenses

Sextuple magnet

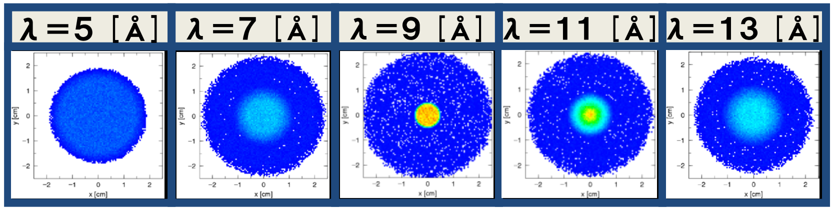


Magnetic focusing

Magnetic lensing with permanent magnets success of neutron focusing experiment at Hokkaido Univ.

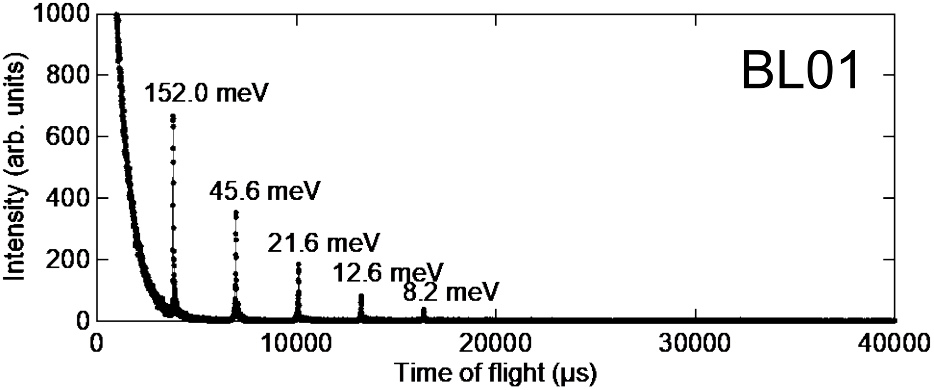


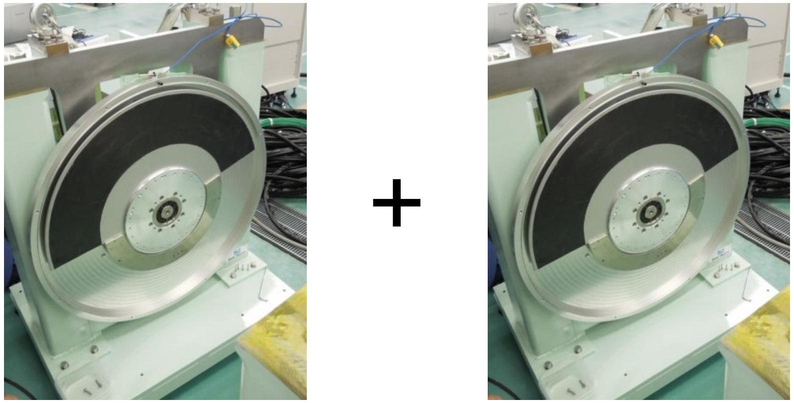
TOF synchronous (wide wavelength bandwidth type) with no chromatic aberration to the development of pulsed sextupole magnets



Other examples of neutron devices（J-PARC MLF）

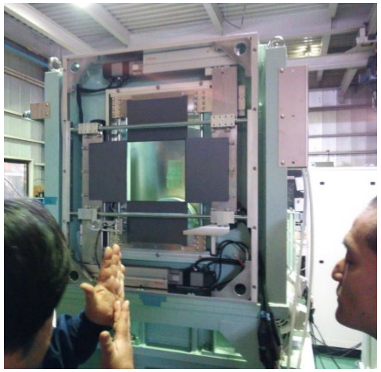
Chopper (Monochromator)

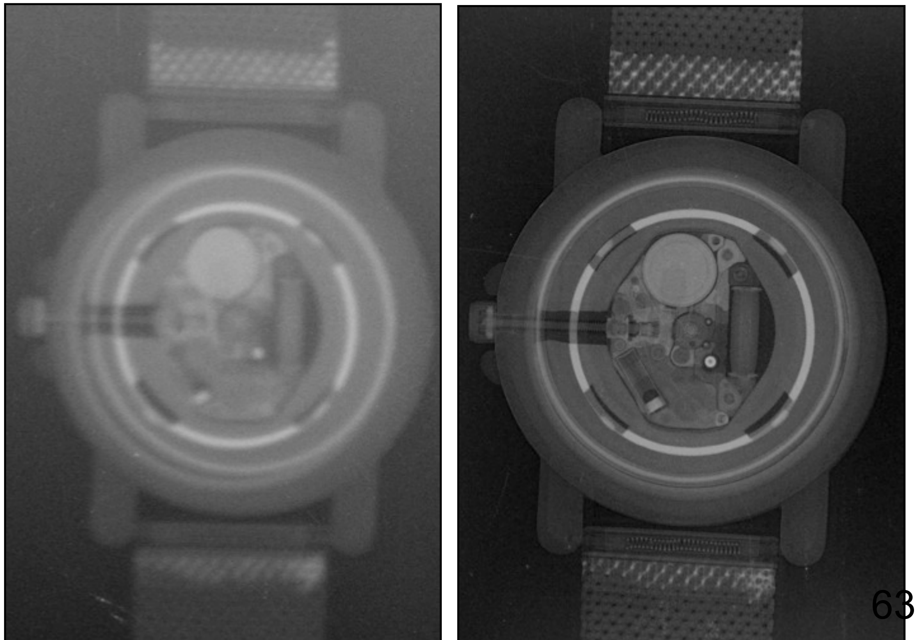




Double disk chopper:
Variable aperture area.

Collimator (Divergence angle controller)





Wo. collimator

With collimator

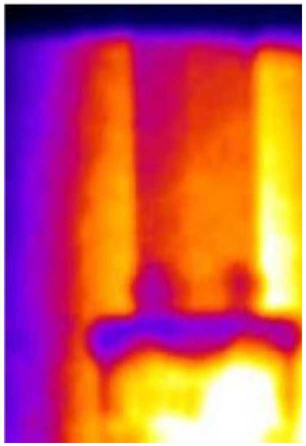
Neutron utilization

Quantum beams" in mechanical engineering!

Some specific examples will be presented later.

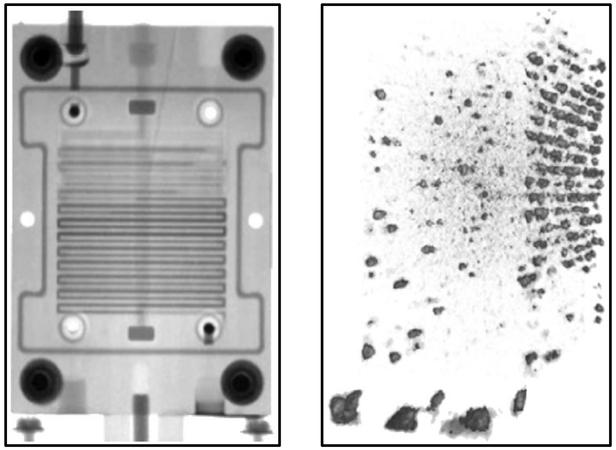
- **Space engineering**
- **Thermal and fluid engineering**
- **Materials engineering**
- **Bionics**
- **Control engineering**
- **Fuel cells, lithium batteries**
- **Nuclear reactor engineering**

Neutron imaging analysis of space propulsion thrusters by JAXA

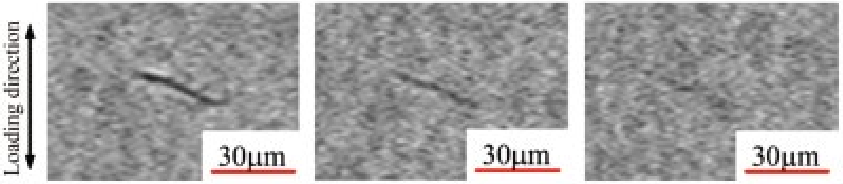


Neutron imaging analysis of oil behavior inside an engine

Water behavior in fuel cells 4D neutron imaging



Fatigue cracks in metallic materials synchrotron radiation imaging analysis of fatigue cracks in metallic materials (Prof. Nakamura)



1300N (80%) 800N (50%) 0N (0%)

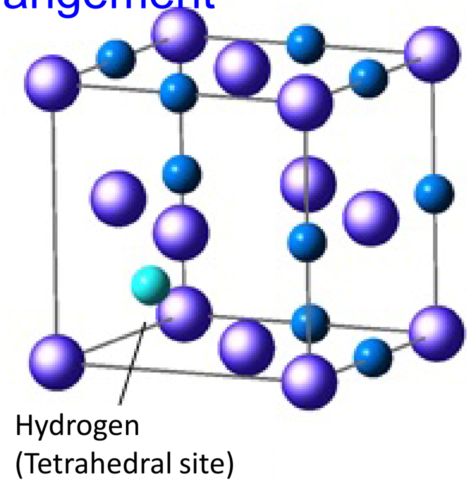
Quantum beams" in Engineering and Science!

Some specific examples will be presented later.

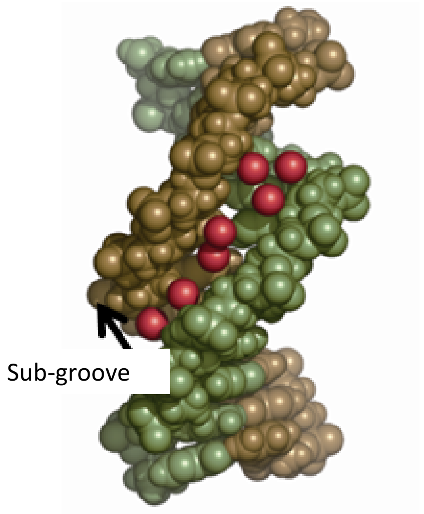
- **(Applied) Physics**
- **(Applied) Chemistry**
- **Biology**
- **Space and planetary science**

- **Materials engineering**
- **Architectural engineering**
- **Soil engineering**
- **Environmental engineering**

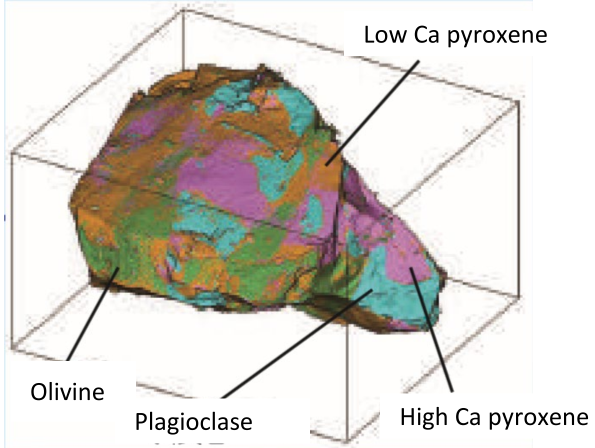
Neutron scattering atomic arrangement analysis



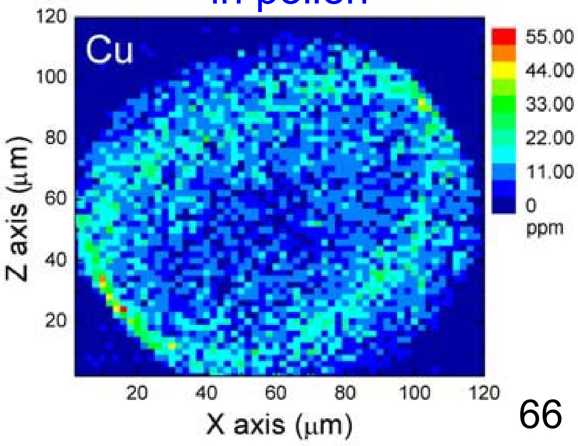
DNA structure and sub-groove hydration water



Space probe Hayabusa brought back quantum beam linkage analysis of asteroid material



By X-ray fluorescence analysis image of elements in pollen

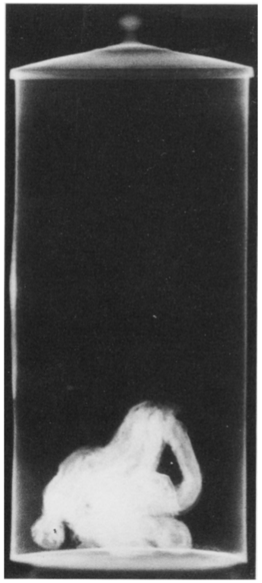


The "quantum beam" is relevant to (almost) all faculties at Hokkaido University!

Some specific examples will be presented later.

- **Literature (Cultural property)**
- **Law (Forensic science)**
- **Medicine and dentistry (Treatment and diagnosis)**
- **Pharmaceutical sciences (Drug discovery / pharmaceutical)**
- **Agricultural science**

Neutron imaging analysis of paper in a sutra tube

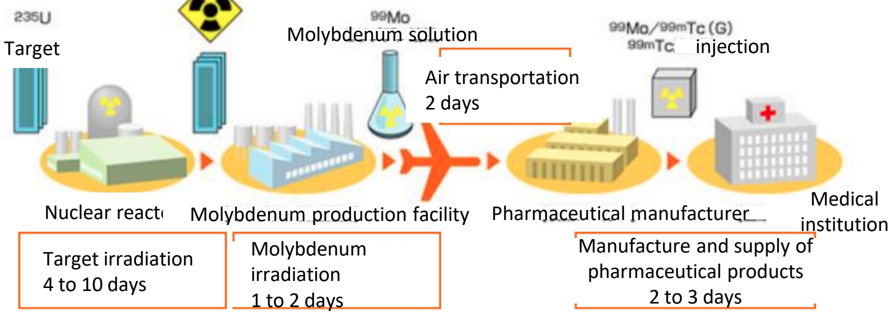


Neutron-based pharmaceuticals of radiopharmaceuticals for medical diagnostic SPECT

Process of 99mTc preparation

$^{99}\text{Mo} \Rightarrow {}^{99m}\text{Tc} \Rightarrow {}^{99}\text{Tc} \Rightarrow$

65.94hr 6.015hr 2.1×10^5y



Air transportation
2 days

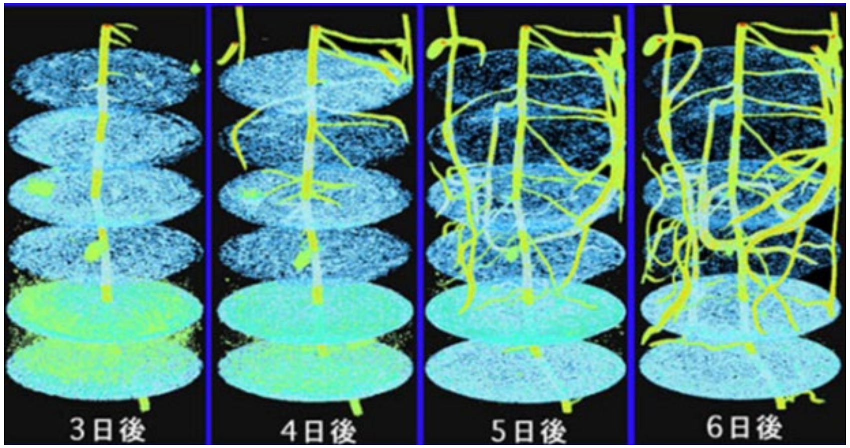
Nuclear react
Molybdenum production facility
Pharmaceutical manufacturer
Medical institution

Target irradiation
4 to 10 days

Molybdenum irradiation
1 to 2 days

Manufacture and supply of pharmaceutical products
2 to 3 days

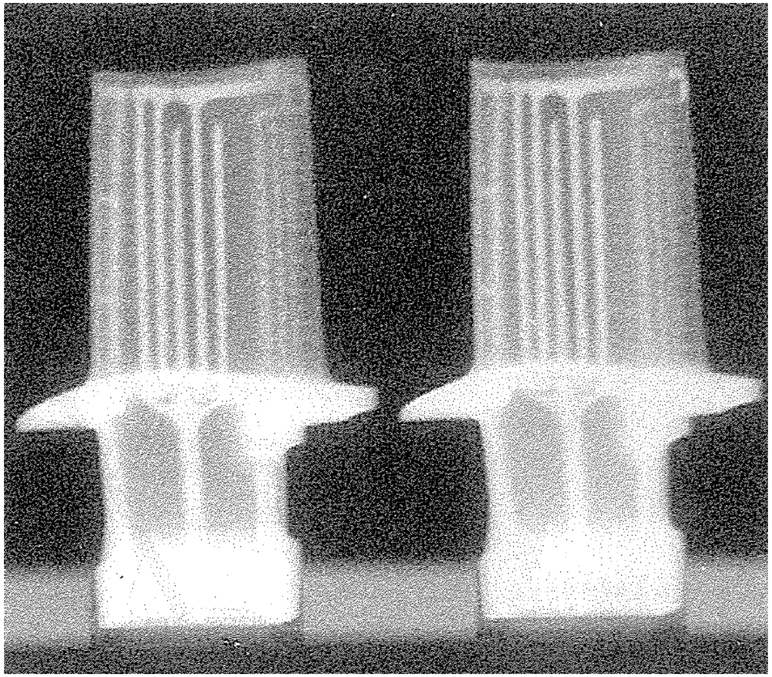
4D neutron imaging analysis of root growth in soil

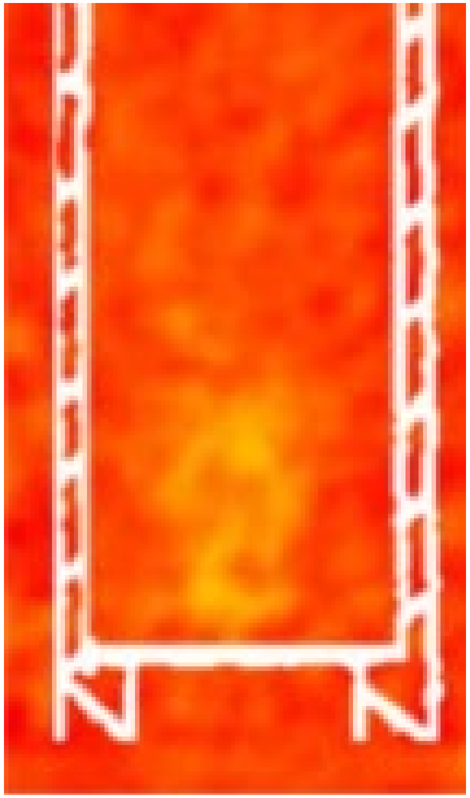


Non-destructive testing and development research of aerospace equipment by neutron imaging

Turbine blade

Satellite thrusters



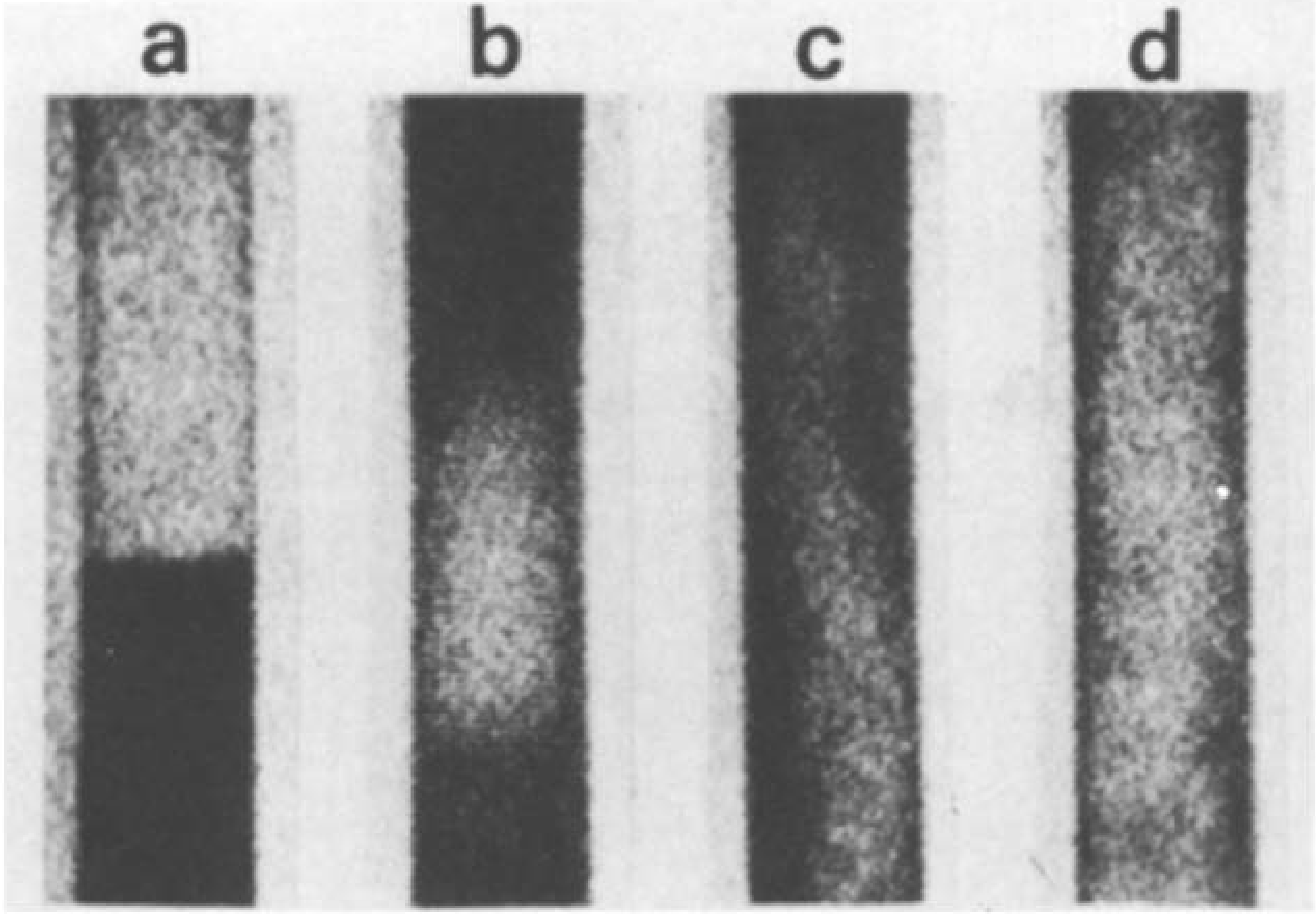


At the proton cyclotron facility of Sumiju Atex Co, inspection of all space equipment by neutron imaging

Two-phase flow studies by neutron imaging

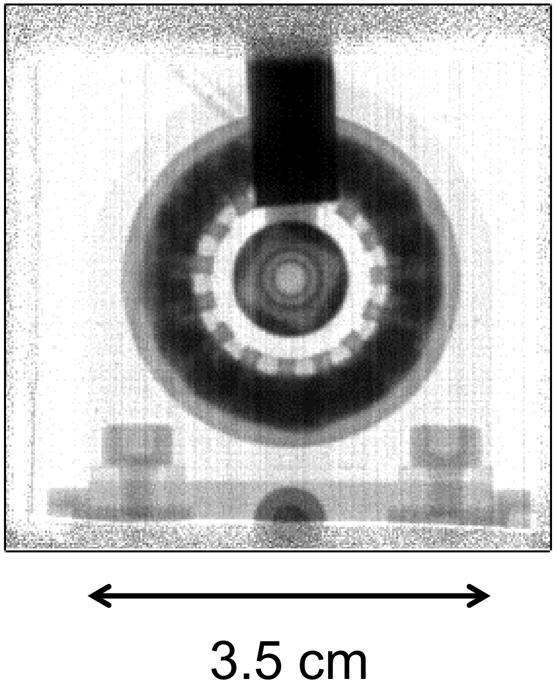
Visualization of water/nitrogen gas two-phase flow in stainless steel pipe

(a) Half water, (b) Slag flow, (c) Bubble flow, (d) Annular flow



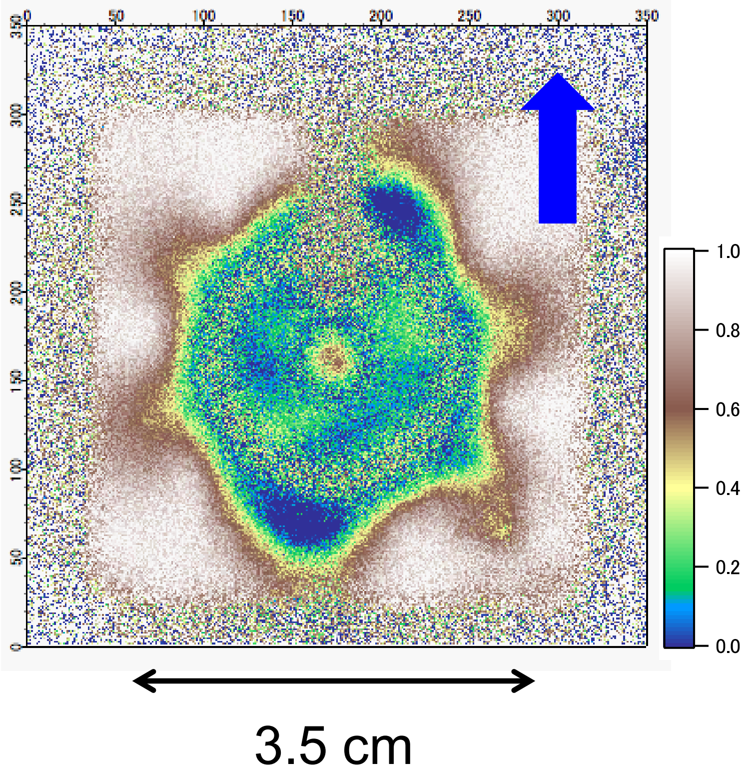
Time-dependent neutron magnetic field imaging analysis of electric vehicle motors

Neutron stroboscopes



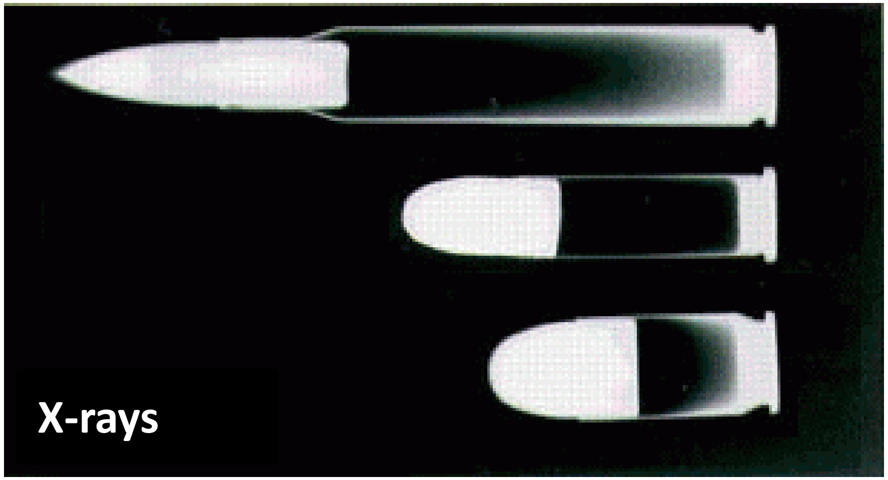
Stroboscope of magnetic field

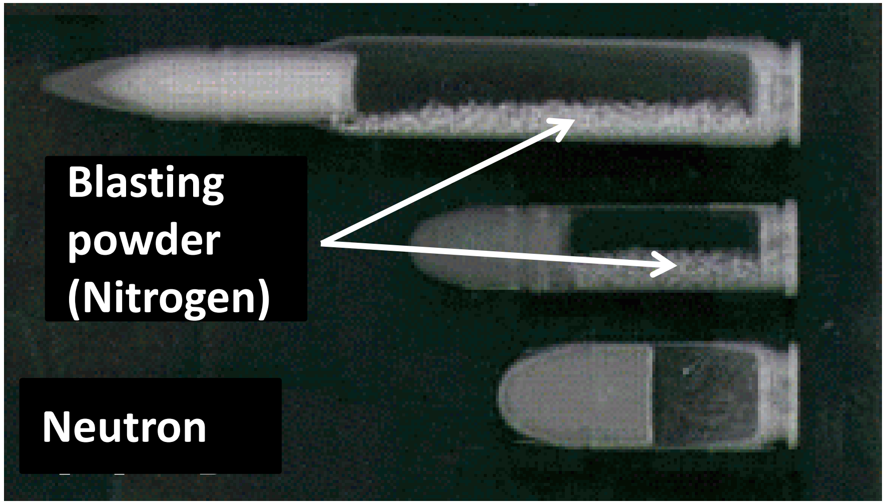
(Visualize the magnetic field in the vertical direction)



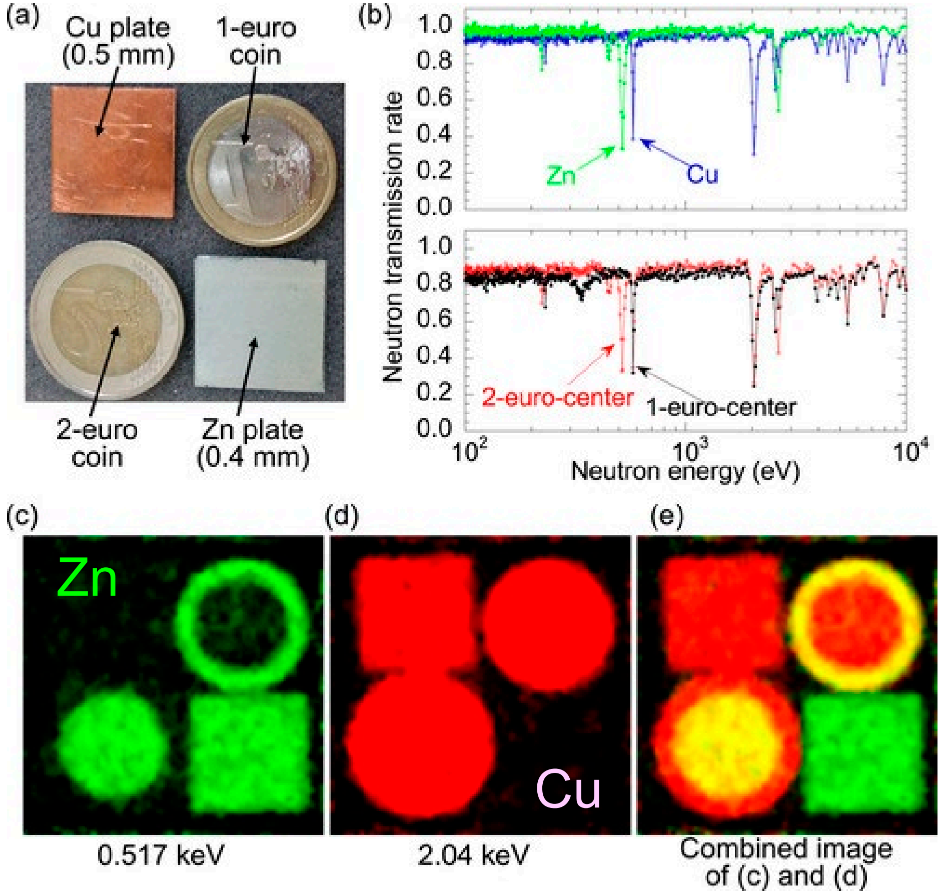
SDGs／Cutting-edge research toward a carbon-neutral society

Neutron imaging analysis for forensic applications



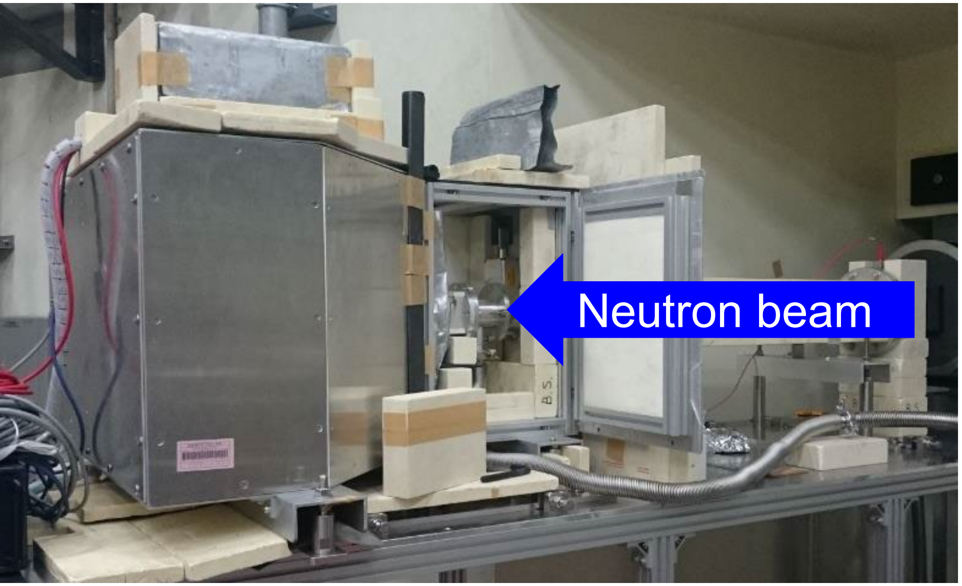


Using neutron resonance absorption, currency inspection by elemental analysis

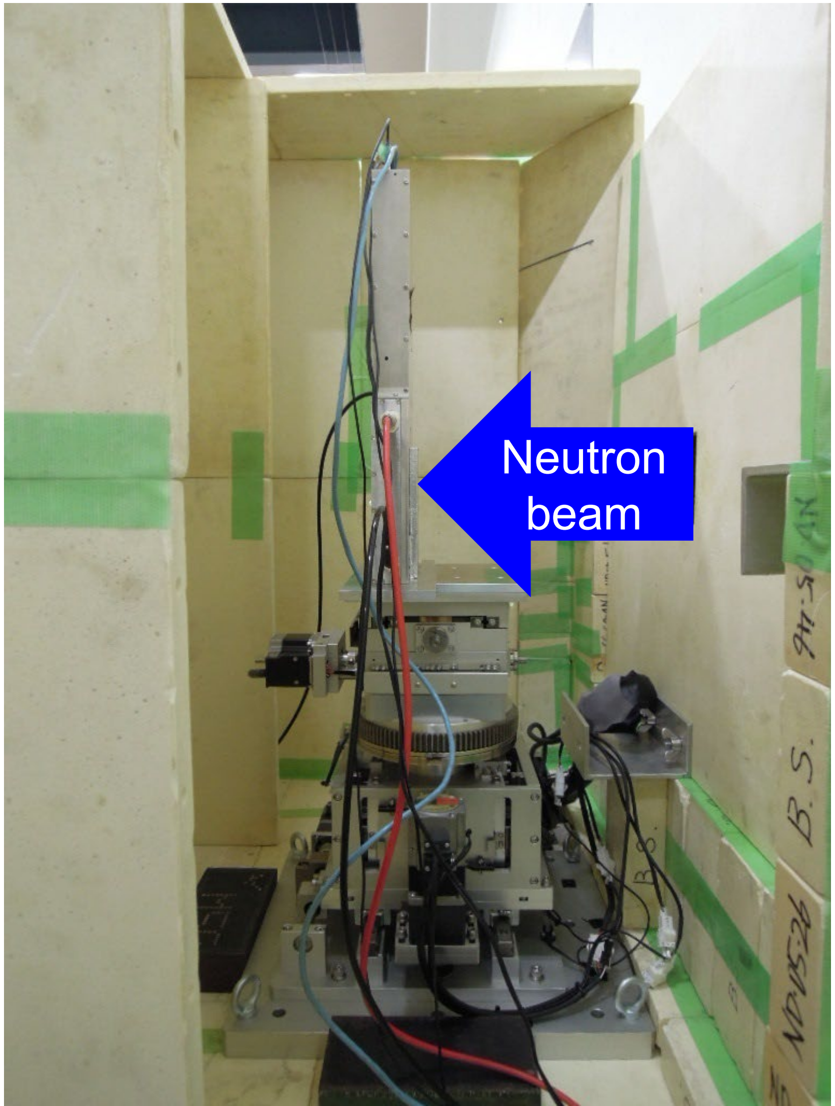


Equipment installed in the neutron laboratory at Hokkaido University

Small-angle neutron scattering instrument

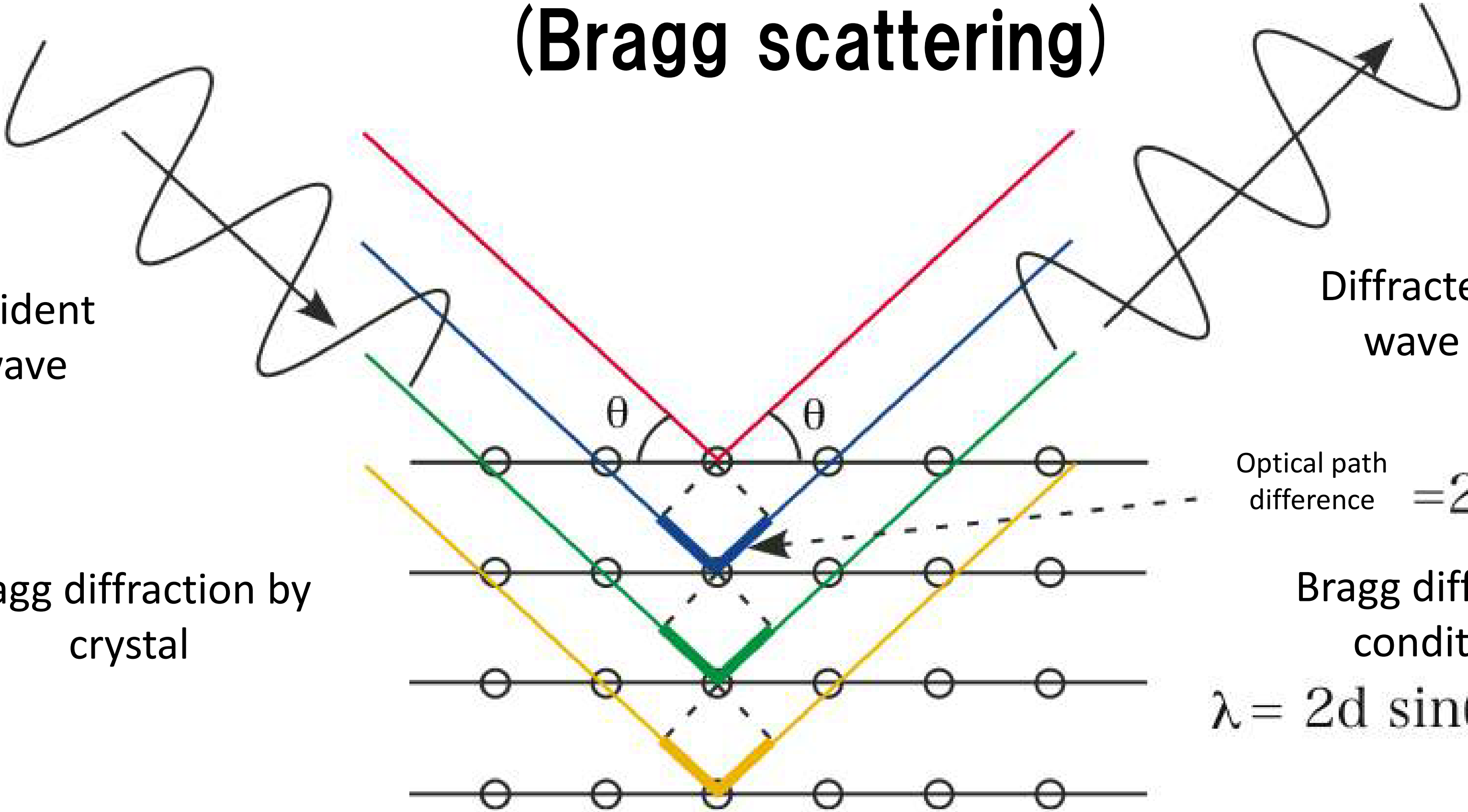


Neutron imaging system



We obtain microscopic information on the atomic level of matter by analyzing the momentum change of neutrons reacting with the matter!

Example: crystal structure analysis using diffraction (Bragg scattering)



Incident wave

Diffracted wave

Optical path difference $=2d\sin\theta$

Bragg diffraction by crystal

Bragg diffraction conditions

$\lambda = 2d\sin\theta$

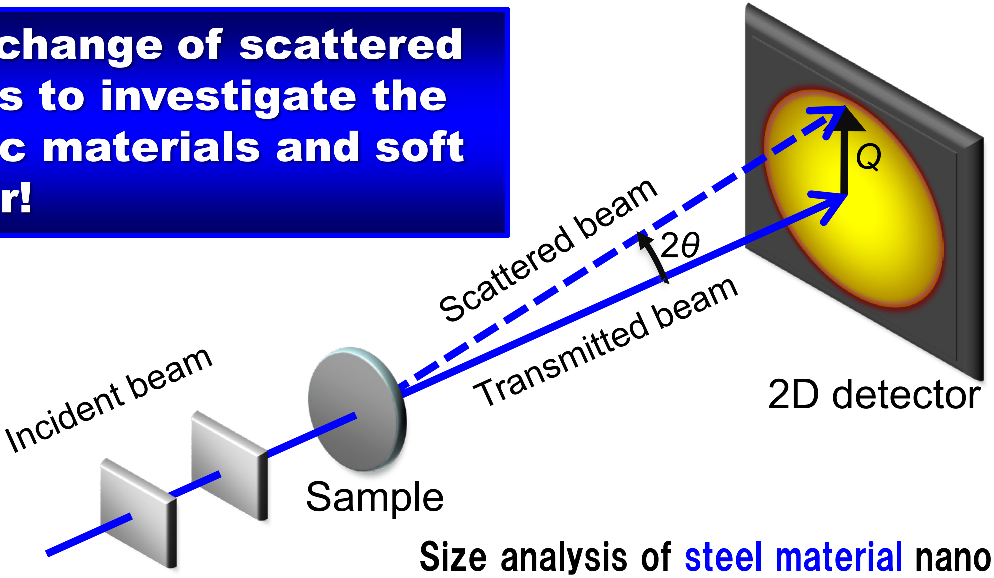
http://bb.phys.se.tmu.ac.jp/~bb/pukiwiki/index.php?Neutron_xray_1

When you want to know the face spacing d of a crystal lattice, you can examine the wavelength λ and the scattering angle 2θ (i.e., the momentum change) of the neutrons that caused the diffraction phenomenon (intensification).

Nano-structural analysis of steel, food, and building materials by small-angle neutron scattering

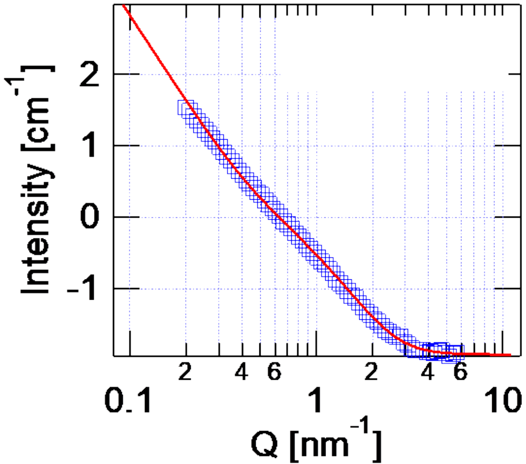
Quantum beam materials engineering laboratory A (Ohnuma laboratory)

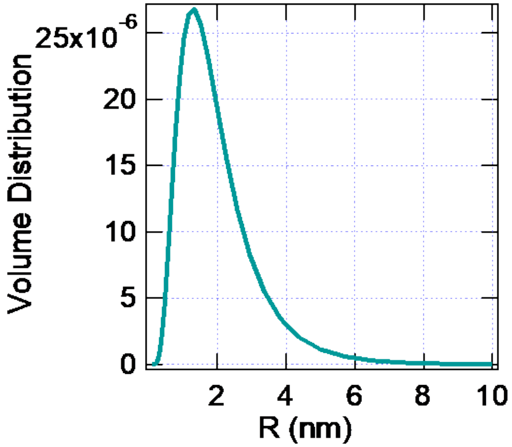
Analyze the momentum change of scattered neutrons at small angles to investigate the nanostructure of metallic materials and soft matter!



Size analysis of steel material nanoprecipitates, analysis of aggregation state of food proteins, nano-structure analysis of building materials (cement), etc.

Example of measured SAXS data

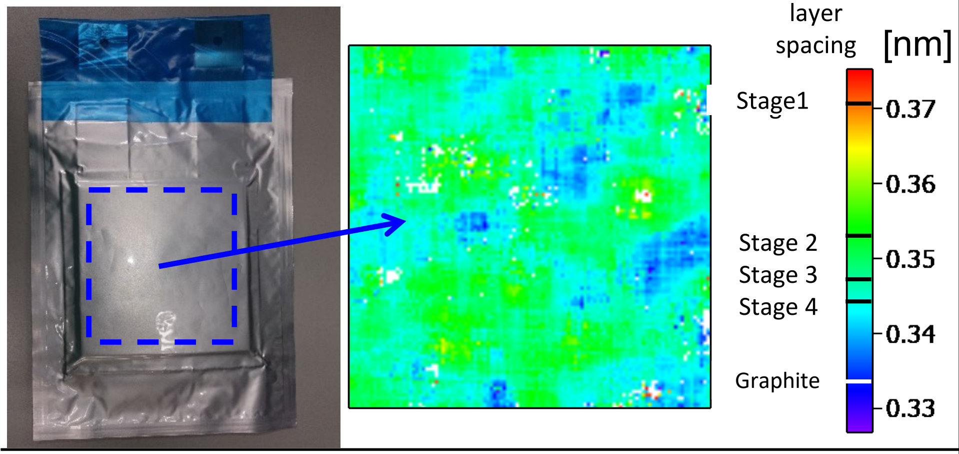




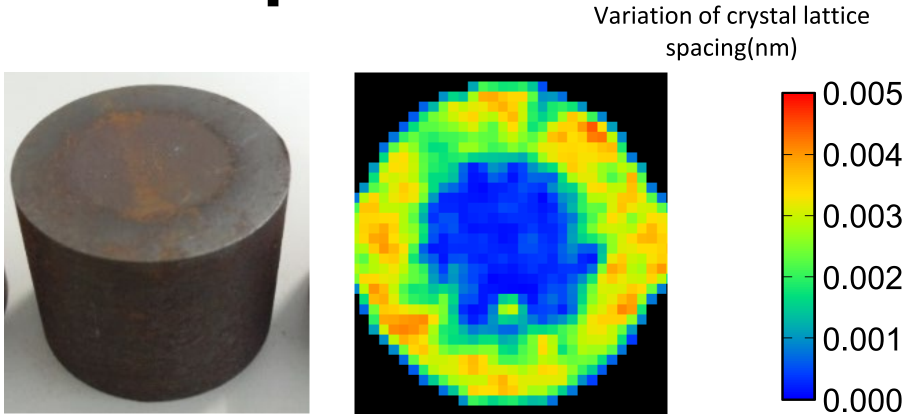
Extensive analysis of automotive parts and cultural assets by neutron imaging

Neutron beam science and engineering laboratory (Kamiyama-Sato laboratory)

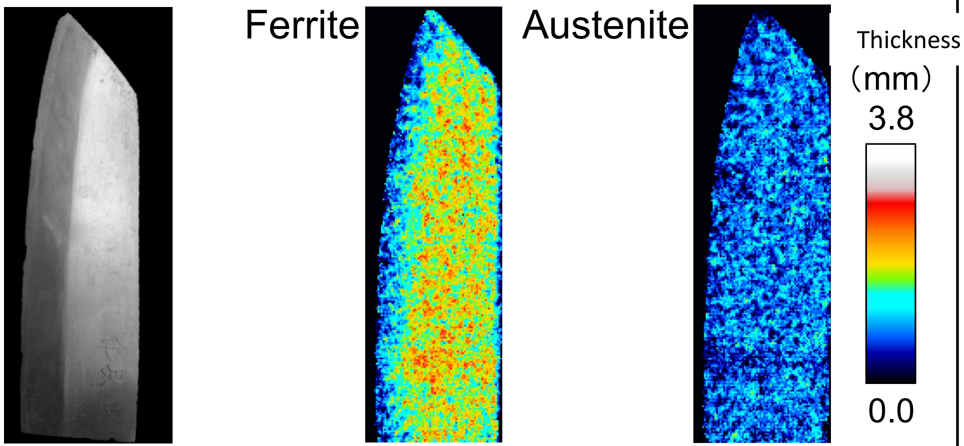
Visualization of interlayer distance of graphite anode material for **Lithium ion batteries**



Visualization of strain in heat-treated parts of **crankshafts**

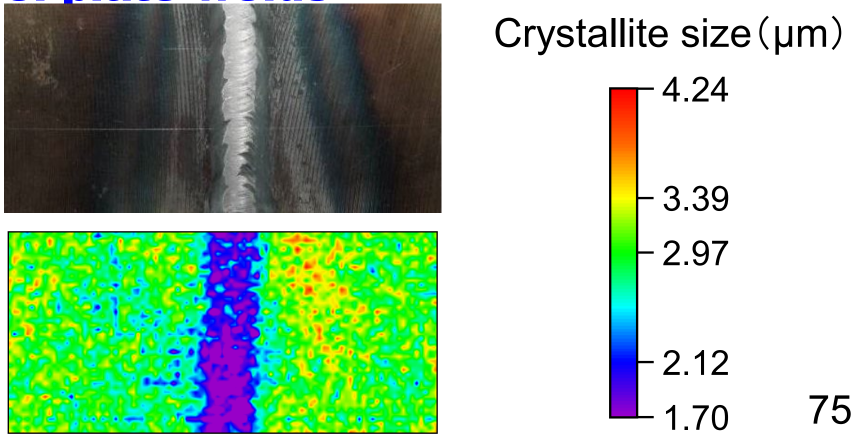


Visualization of the amount of different metals present in **Japanese knives**



Visualization of crystallite size in **steel plate welds**

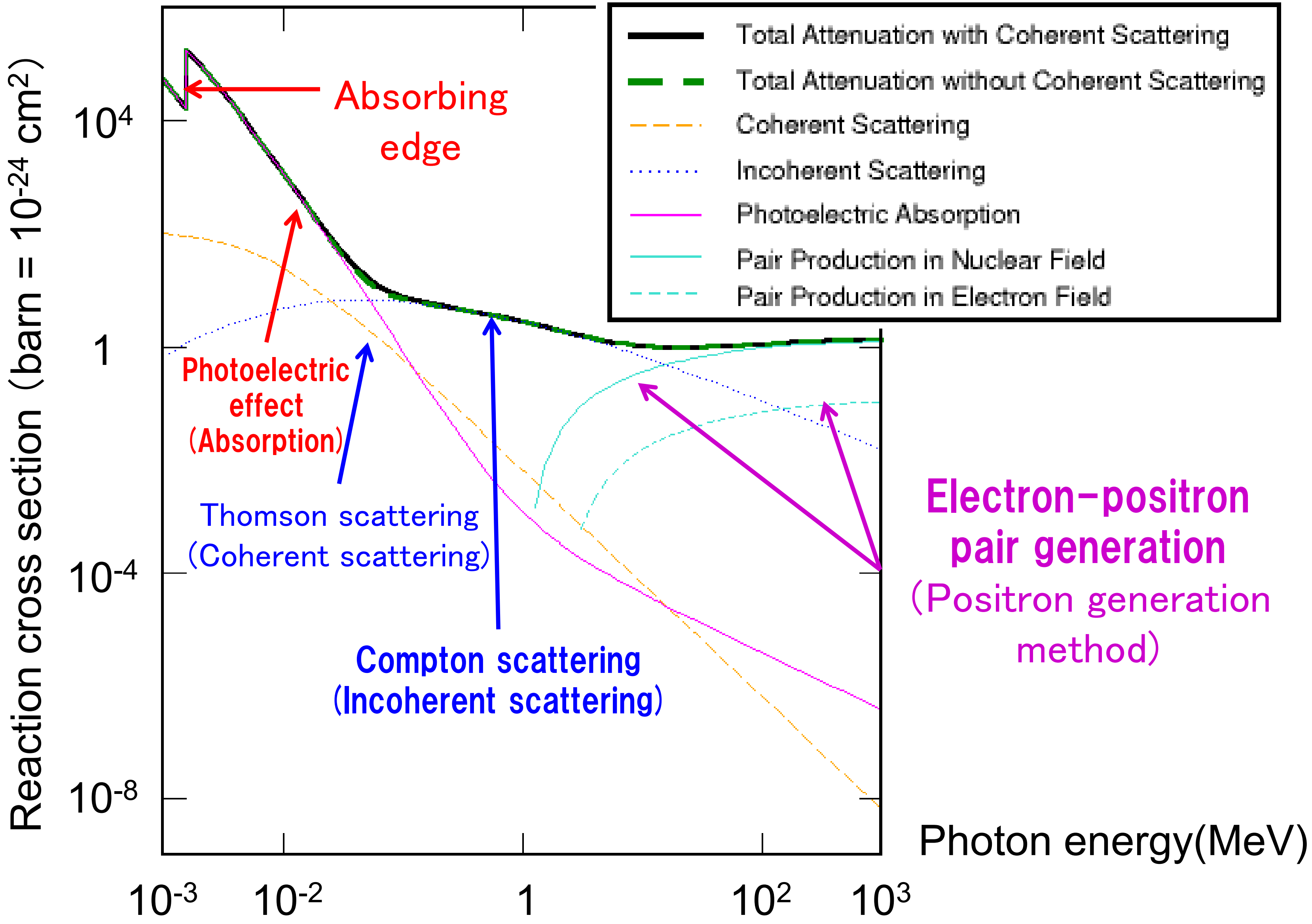
※ Applied to the study of **rails** and **Japanese swords, etc.**



Reaction cross section between neutrons and nuclei

Total cross section of X-rays and one Al atom（Scattering·Absorption）

http://www.nist.gov/pml/data/xcom/index.cfm



Absorbing edge

Photoelectric effect （Absorption）

Thomson scattering （Coherent scattering）

Compton scattering （Incoherent scattering）

Electron-positron pair generation

（Positron generation method）

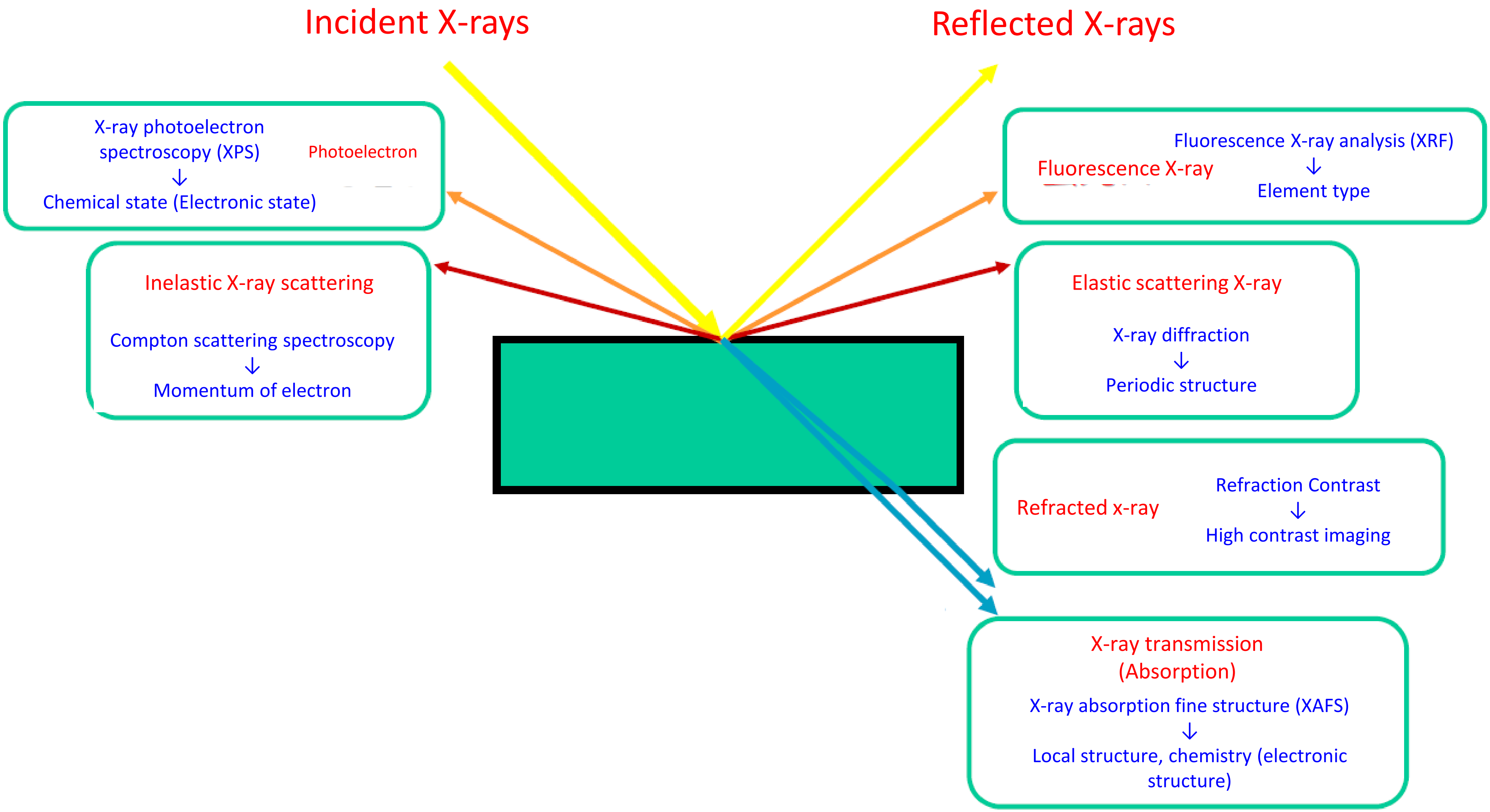
Interaction of γ-rays with matter

- Photoelectric effect: K-orbital or L-orbital electrons absorb all of the energy of the γ-ray and are ejected.
- Compton scattering: Orbital electrons that collide with γ-rays acquire part of the γ-ray energy.
- Electron pair production: When the γ-ray energy exceeds 1.022 MeV, electron-positron pairs are produced.

- Due to these interactions, a γ-ray incident on a matter can give all (or part) of its energy to the matter.

Material analysis using X-ray-matter interaction

http://www.sci.u-hyogo.ac.jp/material/x-ray_optics/kago/presentations/



Incident X-rays

Reflected X-rays

X-ray photoelectron spectroscopy (XPS)
↓
Chemical state (Electronic state)

Photoelectron

Inelastic X-ray scattering

Compton scattering spectroscopy
↓
Momentum of electron

Fluorescence X-ray

Fluorescence X-ray analysis (XRF)
↓
Element type

Elastic scattering X-ray

X-ray diffraction
↓
Periodic structure

Refracted x-ray

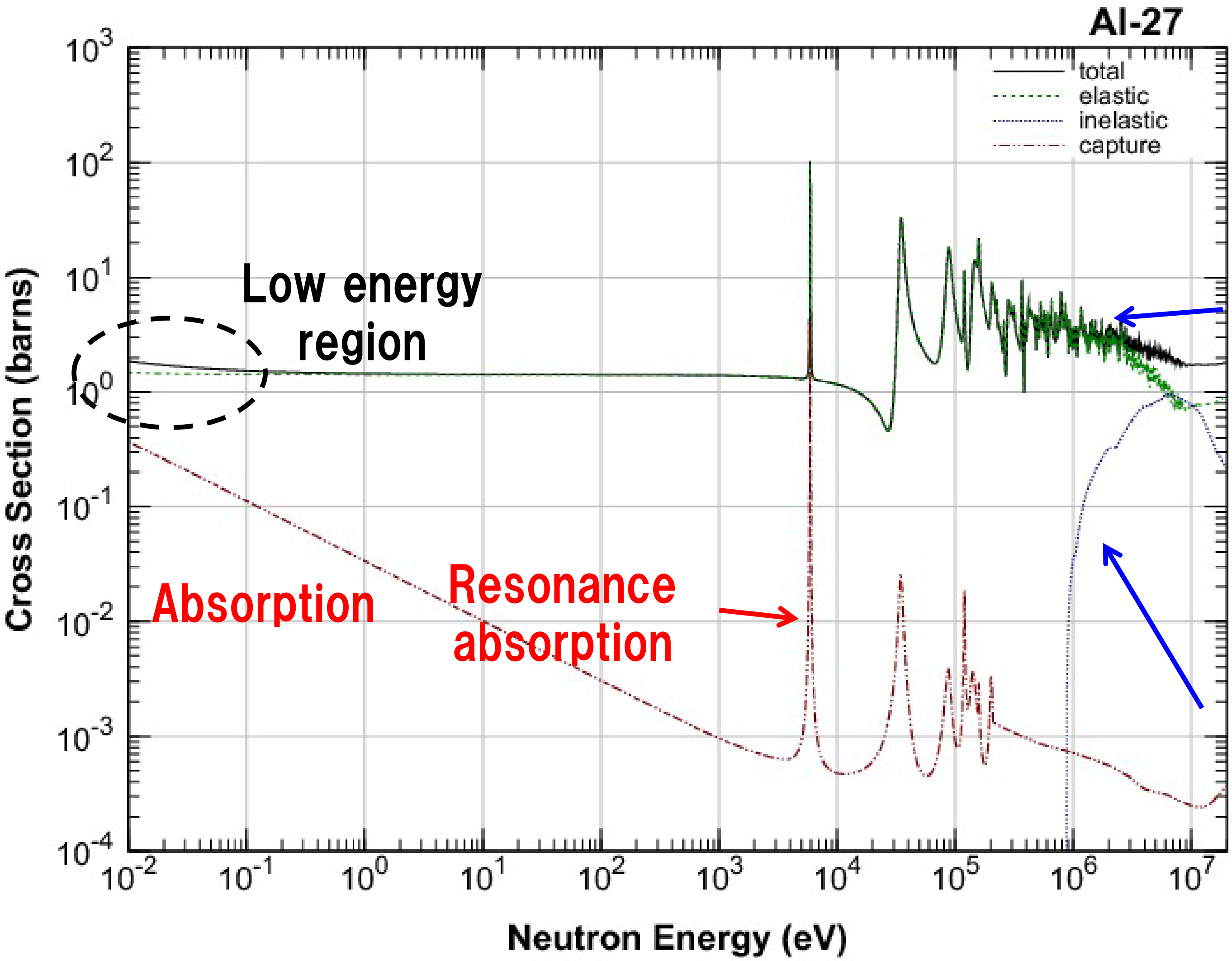
Refraction Contrast
↓
High contrast imaging

X-ray transmission (Absorption)

X-ray absorption fine structure (XAFS)
↓
Local structure, chemistry (electronic structure)

Total cross section of neutron and one Al nucleus

http://wwwndc.jaea.go.jp/jendl/j40/J40_J.html



Low energy region

Absorption

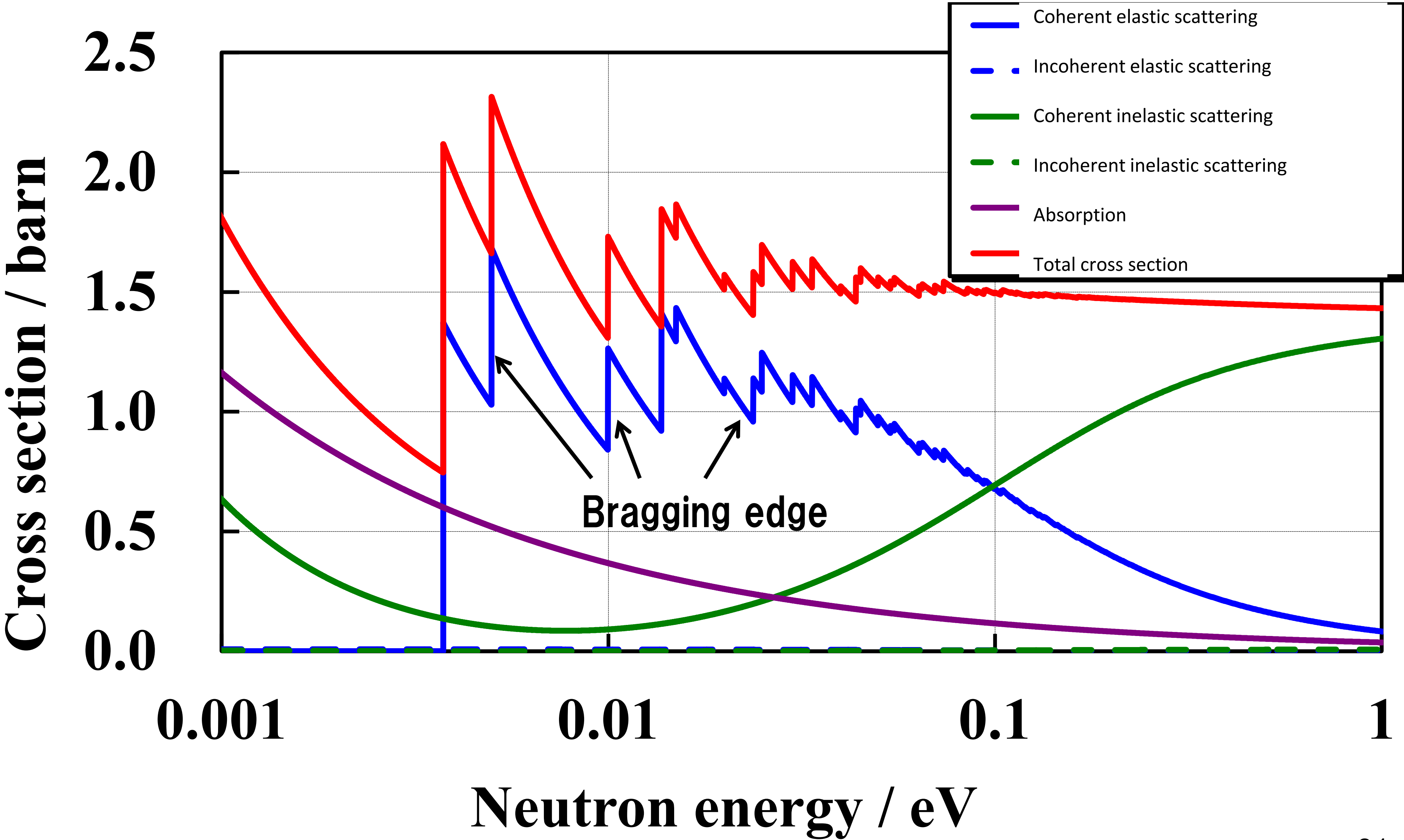
Resonance absorption

Resonance scattering

(n,p) reaction, (n,α) reaction etc. Sources of soft errors

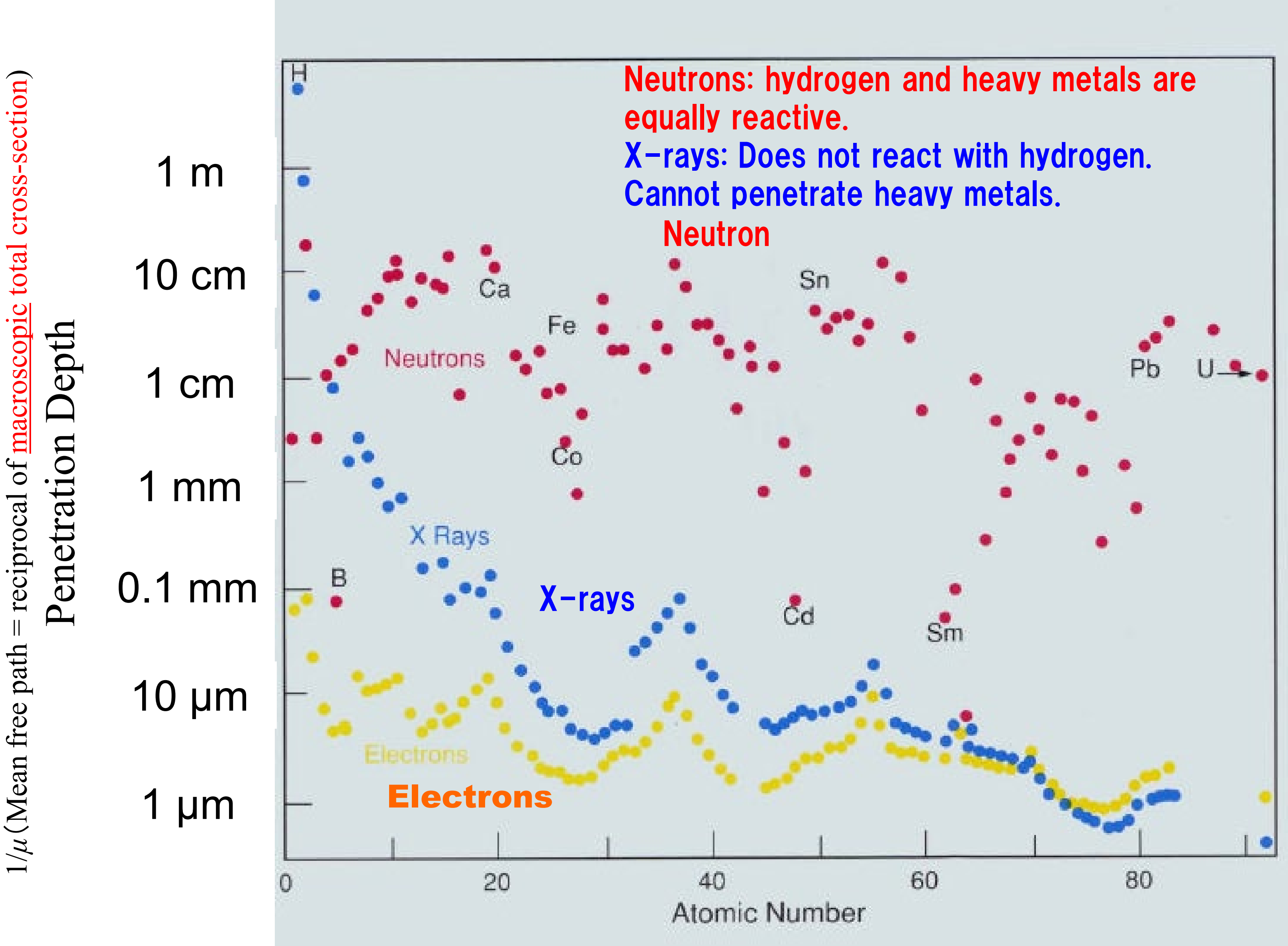
Total cross-section in low-energy region

Data of Al nuclei calculated by "RITS" code developed by Hokkaido University



Comparison of mean free paths of electrons, X-rays, and neutrons

Roger Pynn, Neutron Scattering - A Primer, Los Alamos National Laboratory



Neutrons: hydrogen and heavy metals are equally reactive.

X-rays: Does not react with hydrogen. Cannot penetrate heavy metals.

Scattering and absorption cross sections of major elements for thermal neutrons

Element	Scattering cross section（barn）	Absorption cross section（barn）	Applications in neutron engineering
H	82	0.33	Moderator (∵ light nuclei) After neutron capture, high-energy γ rays are also emitted
Li	1.37	71	Absorbers with low gamma ray effect after neutron capture
B	5.24	767	Absorber Emits alpha rays after neutron capture
Al	1.5	0.23	Transparent material (?)
Si	2.17	0.17	
Fe	11.6	2.56	Scatterer
Co	5.6	37.18	Co-60 (gamma ray emitter) formation by neutron capture
Cd	6.50	2520	Absorbing material, intense γ-ray emitter
Gd	180	49700	Absorber
Pb	11.1	0.17	Non-decelerating scatterer Excellent gamma ray shielding material

Radiation safety at neutron generating facilities (Once again, see and learn about the site firsthand!)

Principles of elemental analysis by neutron activation analysis

What is Neutron Activation Analysis?

- Neutrons are captured by various nuclei, and γ rays may be emitted from the nucleus that capture the neutron. This is called the capture reaction ((n,γ) reaction).

- Since the energy of γ rays emitted from a radionuclide has a unique value for each nuclide, the nuclide can be identified by measuring its energy.

- Based on this principle, a sample is irradiated by neutrons to nondestructively qualify or quantify the nuclides contained in the sample, which is called neutron activation analysis.

Applications of neutron activation analysis

Neutron activation analysis (elemental analysis) that raised the possibility of Napoleon's "poisoning" theory.

- When neutrons are captured in the nucleus, the matter is activated (becomes radioactive).
- Analyzing the energy of radiation (gamma rays) reveals the type of nucleus (element).
- Suitable for cultural assets because it does not break or dissolve like normal chemical analysis.

Napoleon's hair was found to contain a large amount of arsenic! Poisoning? !

Image source：http://www.revive.co.jp/magokoro/scrap_and_build/history-napoleon-revolution/

Analysis of particles brought back by Hayabusa from the asteroid "Itokawa" through quantum beam fusion collaboration

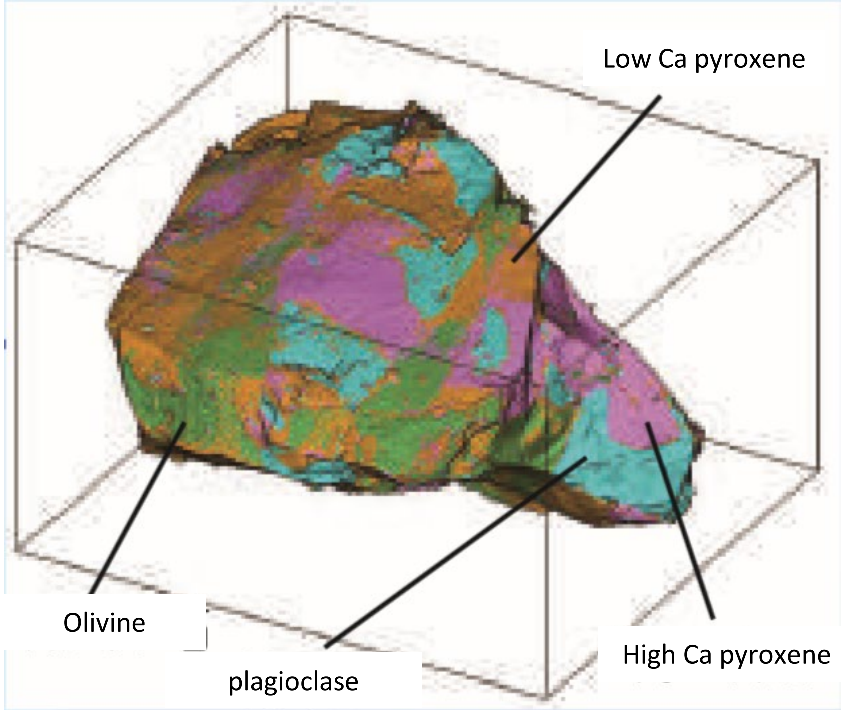


image source: "Quantum Beam" for a family

Three-dimensional distributions of olivine, pyroxene, and other minerals in dust particles from the asteroid Itokawa brought back from space were obtained.

- Analysis of 3D shape and 3D internal structure of particles by X-ray CT imaging
- Crystal structure analysis of minerals by X-ray diffraction (Bragg scattering)
- Elemental analysis by neutron activation analysis
- Identification of minerals and microstructural analysis of particles by electron microscopy

Even if one analytical method cannot give us a complete picture, by combining the power of quantum beams, we can even see the history of the formation of the solar system!

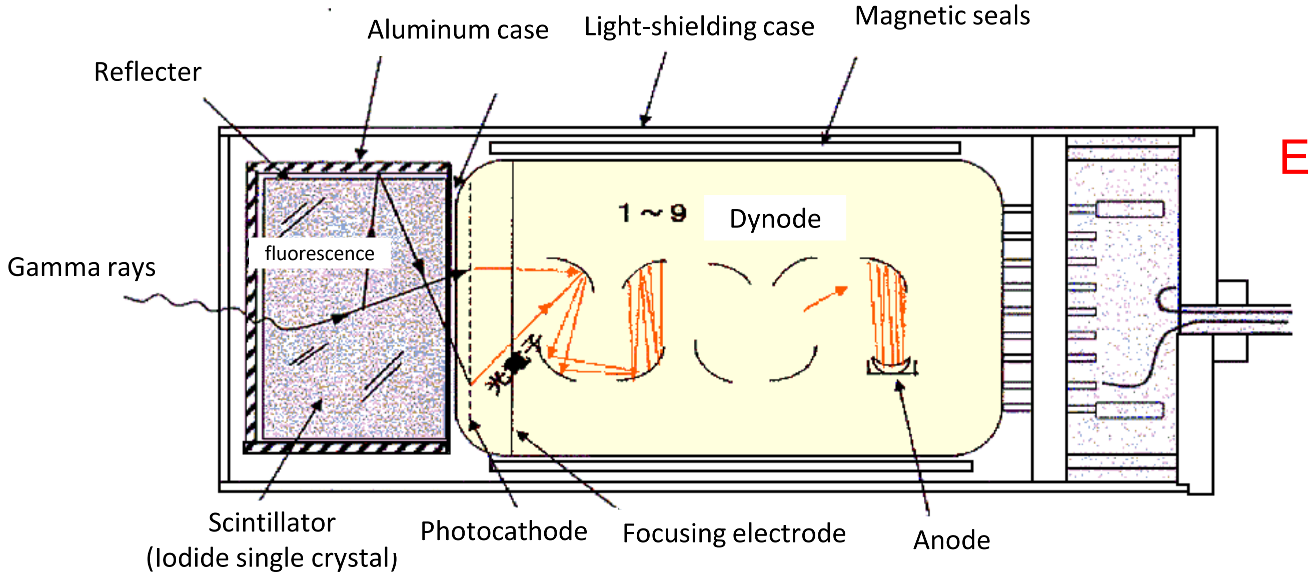
Pulse height spectrum of NaI(Tl) scintillation detector

NaI(Tl) scintillator

- As a gamma (γ) ray measurement device, a scintillation detector consisting of a sodium iodide (NaI) crystal containing a trace amount of thallium (Tl) is commonly called an NaI(Tl) scintillator.
- When a NaI(Tl) crystal is bombarded with γ-rays, the interaction (photoelectron effect, Compton scattering, and electron pair production) causes secondary electrons to excite the NaI(Tl) crystal material, and when the excited state returns to a steady state, fluorescence, called scintillation, is produced.
- This light is captured and combined with a photomultiplier tube to convert the radiation dose into a current pulse, which is used as a radiation measuring instrument.
- The intensity of the light is proportional to the energy lost by secondary electrons in the crystal, which provides information on the energy of the incident γ-rays.
- Scintillators have the advantages of (1) fast measurement due to short scintillation decay time, and (2) energy measurement from the proportional relationship between the amount of fluorescence and absorbed energy.

Operating principle of NaI(Tl) scintillator

- When γ-rays strike the NaI crystal, secondary electrons are produced and the Tl atoms emit fluorescence. Thallium (Tl), which emits fluorescence, is called an activator.
- When the fluorescence by the Tl atom hits the photocathode of the photomultiplier tube, photoelectrons are ejected due to the photoelectric effect, which is multiplied by a number of dynodes to generate a pulse current in the circuit.
- By counting the number of pulsed currents, the number of γ rays that enter the NaI crystal can be measured.



Electrical signal

Amplified by pre-amplifier & main amplifier

Structure of NaI(Tl) scintillator

- NaI(Tl) scintillator in combination with a photomultiplier tube.
- The junction surface of the NaI(Tl) scintillator and photomultiplier tube has a glass window.
- A cylindrical type with a diameter of 5 cm x height of 5 cm or a well type with large geometric efficiency is used.

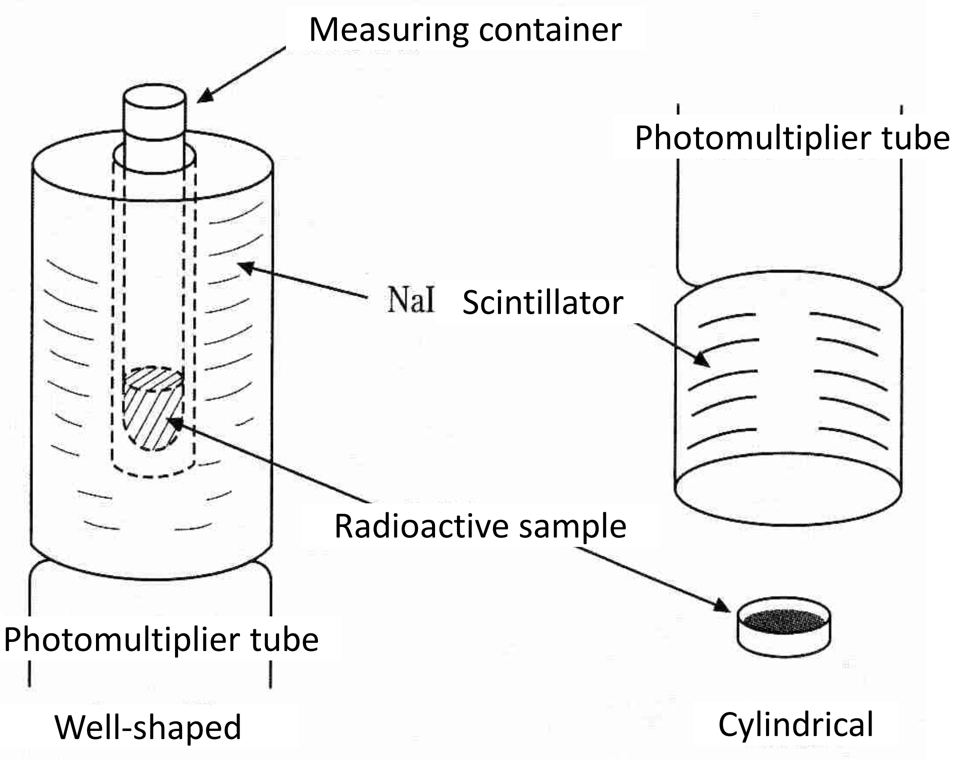


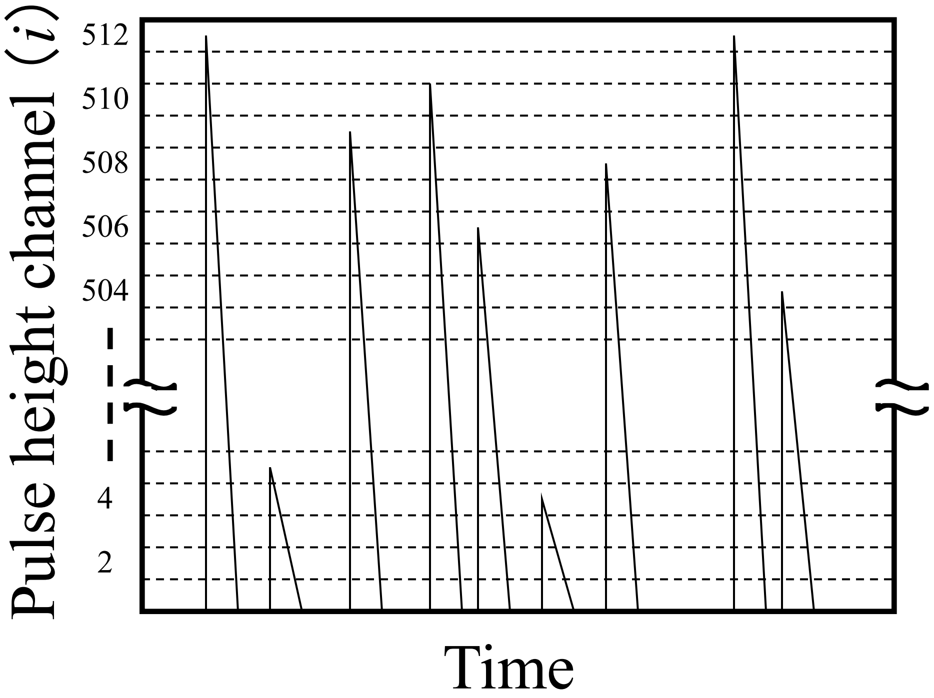
Figure 1.11 NaI(Tl) Scintillator shape

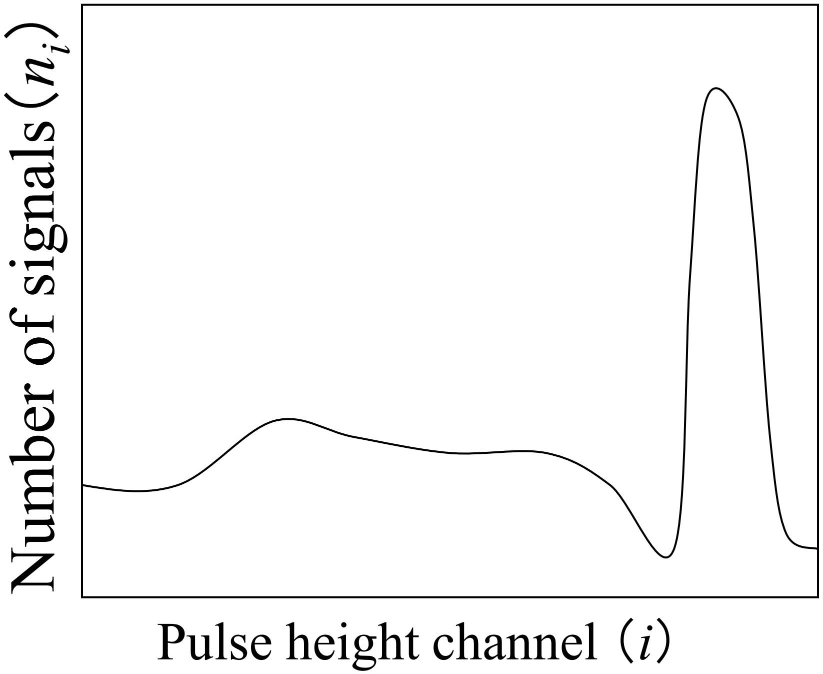
Gamma-ray energy measurement

- The energy required to emit one photoelectron at the photocathode of a photomultiplier tube is 120 eV. When a γ-ray strikes the NaI(TI) scintillator and all of its energy is transferred to the NaI crystal, a number of photoelectrons proportional to the γ-ray energy are produced.
- As a result, a pulse current with intensity proportional to the γ-ray energy flows through the circuit, and the γ-ray energy can be determined by measuring the wave height of the pulse current using a multi-channel pulse height analyzer (MCA).

MCA operation in γ-ray spectrometry

MCA: Multichannel Pulse Height Analyzer

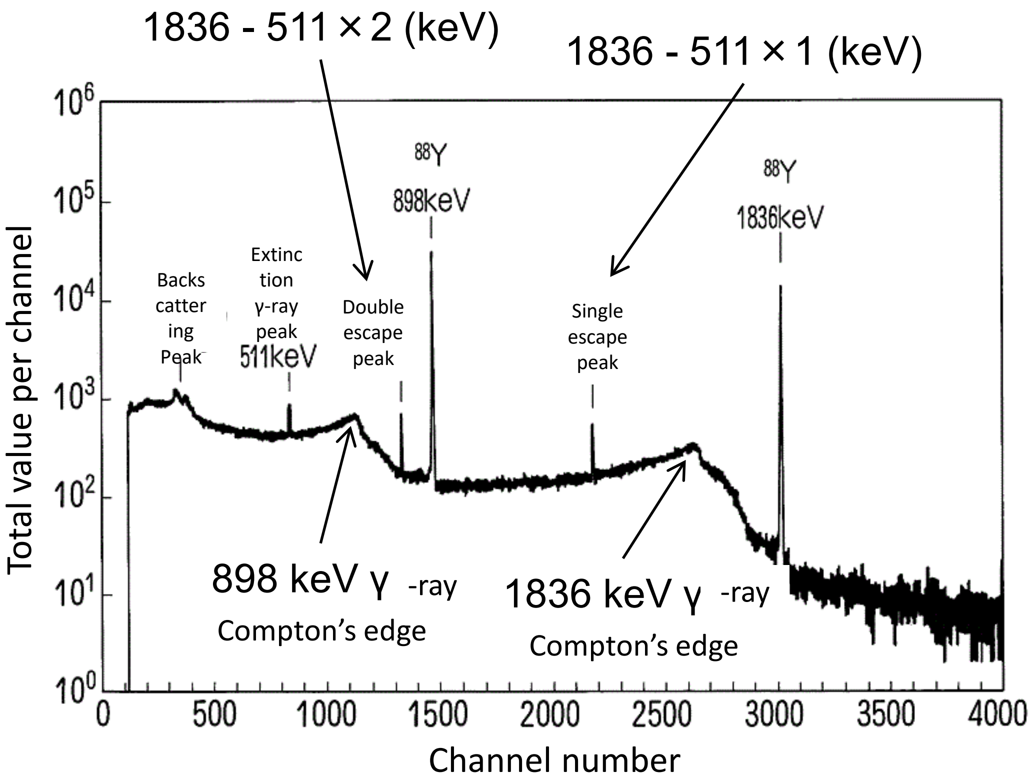




Discretize the pulse height voltage and count the number of signals（n_i）belonging to channel（i）.

Plot the number of signals（n_i）in the pulse height channel（i）

Example of γ-ray spectra from a high-energy-resolution Ge semiconductor detector（^{88}Y）



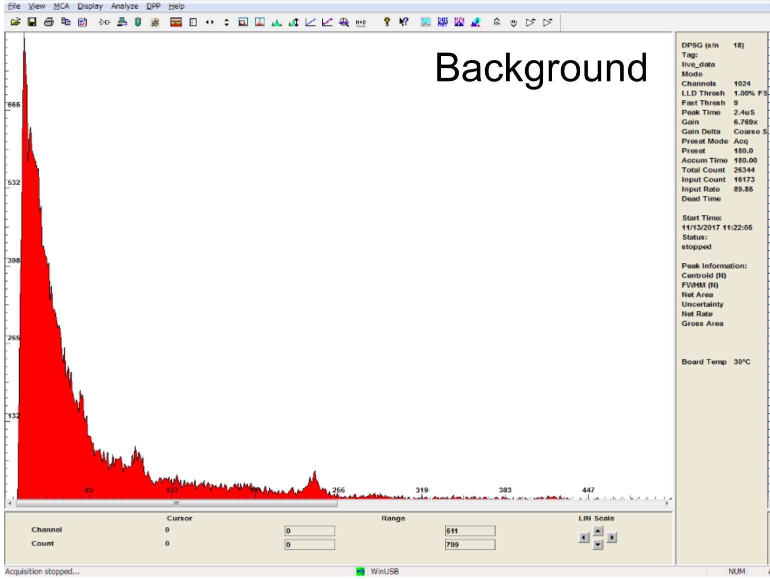
- The total absorption peaks at 1836 keV and 898 keV are caused by the photoelectric effect, where all of the γ-ray energy is absorbed by the NaI crystal.
- Since only a part of the energy of the γ-rays is transferred to the NaI crystal due to Compton scattering, etc., a continuous spectrum is measured.

Neutron irradiation experiments

- **Irradiation samples**
  - ？（Metal）
- **Electron accelerator driven conditions: 10 pps for 15 minutes**
- **Attached to polyethylene moderator and placed near neutron source**

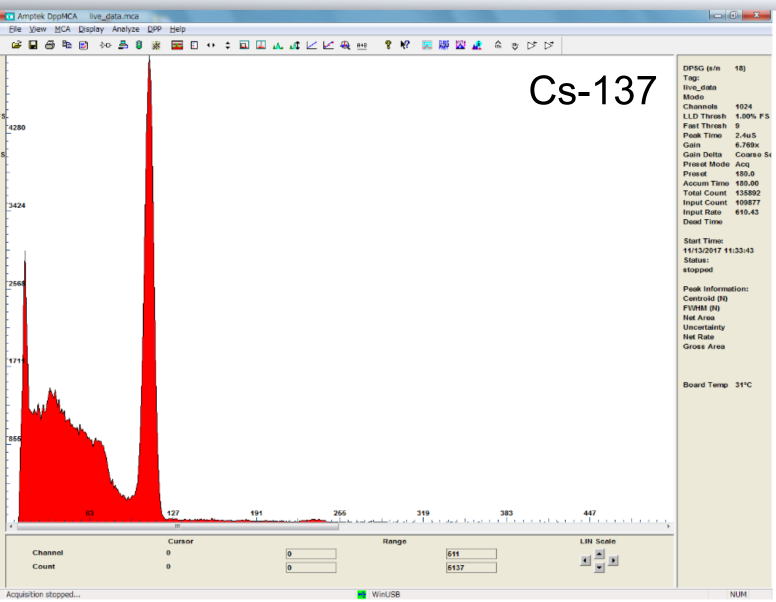
Energy calibration and energy resolution of NaI scintillation detectors

Examples of pulse height spectra (Both examples measured in 3 minutes)



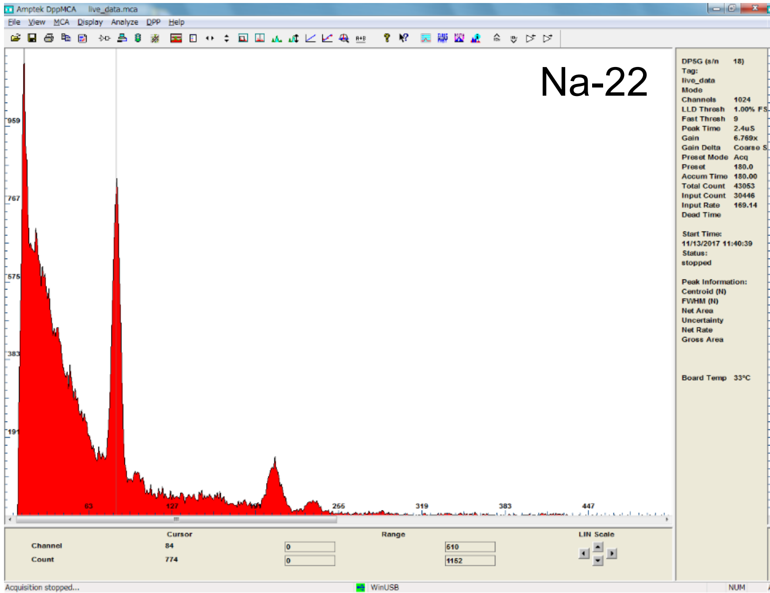
```
DP5G (s/n      18)
Tag:
live_data
Mode
Channels       1024
LLD Thresh     1.00% FS
Fast Thresh    9
Peak Time      2.4uS
Gain           6.769x
Gain Delta     Coarse S
Preset Mode    Acq
Preset         180.0
Accum Time     180.00
Total Count    26344
Input Count    16173
Input Rate     89.85
Dead Time

Start Time:
11/13/2017 11:22:05
Status:
stopped

Peak Information:
Centroid (N)
FWHM (N)
Net Area
Uncertainty
Net Rate
Gross Area

Board Temp     30°C
```

```
         Cursor        Range
Channel  0        0    511
Count    0        0    799
```



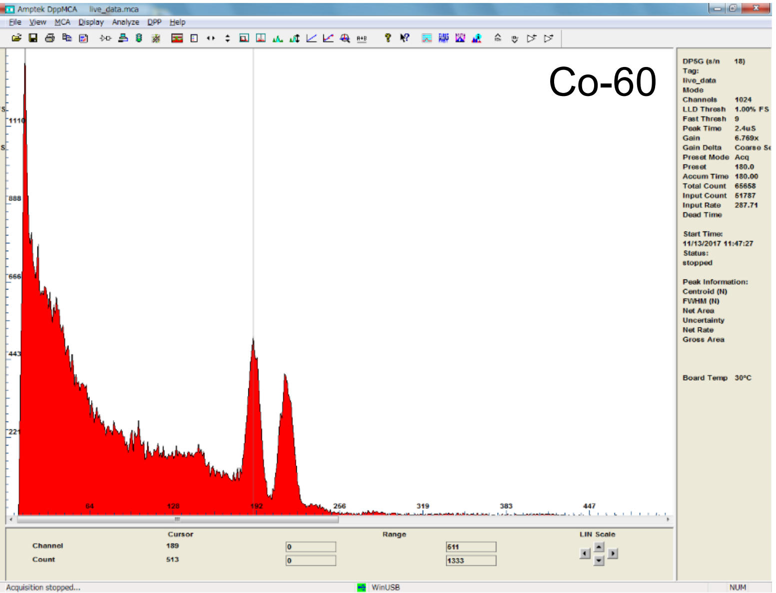
```
DP5G (s/n      18)
Tag:
live_data
Mode
Channels       1024
LLD Thresh     1.00% FS
Fast Thresh    9
Peak Time      2.4uS
Gain           6.769x
Gain Delta     Coarse Se
Preset Mode    Acq
Preset         180.0
Accum Time     180.00
Total Count    135892
Input Count    109877
Input Rate     610.43
Dead Time

Start Time:
11/13/2017 11:33:43
Status:
stopped

Peak Information:
Centroid (N)
FWHM (N)
Net Area
Uncertainty
Net Rate
Gross Area

Board Temp     31°C
```

```
         Cursor        Range
Channel  0        0    511
Count    0        0    5137
```



```
DP5G (s/n      18)
Tag:
live_data
Mode
Channels       1024
LLD Thresh     1.00% FS
Fast Thresh    9
Peak Time      2.4uS
Gain           6.769x
Gain Delta     Coarse S
Preset Mode    Acq
Preset         180.0
Accum Time     180.00
Total Count    43053
Input Count    30446
Input Rate     169.14
Dead Time

Start Time:
11/13/2017 11:40:39
Status:
stopped

Peak Information:
Centroid (N)
FWHM (N)
Net Area
Uncertainty
Net Rate
Gross Area

Board Temp     33°C
```

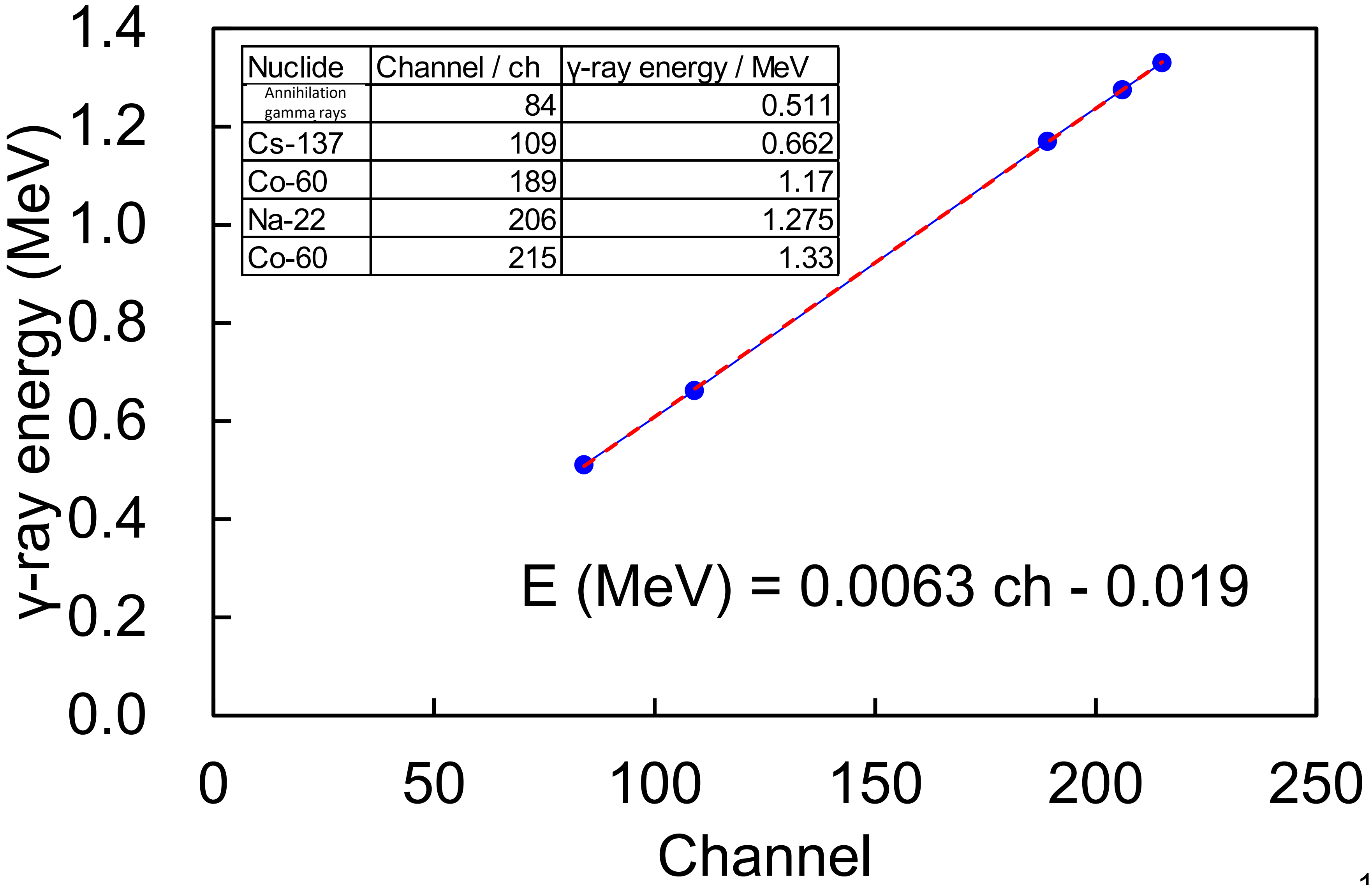
```
         Cursor        Range
Channel  84       0    510
Count    774      0    1152
```



```
DP5G (s/n      18)
Tag:
live_data
Mode
Channels       1024
LLD Thresh     1.00% FS
Fast Thresh    9
Peak Time      2.4uS
Gain           6.769x
Gain Delta     Coarse Se
Preset Mode    Acq
Preset         180.0
Accum Time     180.00
Total Count    65658
Input Count    51787
Input Rate     287.71
Dead Time

Start Time:
11/13/2017 11:47:27
Status:
stopped

Peak Information:
Centroid (N)
FWHM (N)
Net Area
Uncertainty
Net Rate
Gross Area

Board Temp     30°C
```

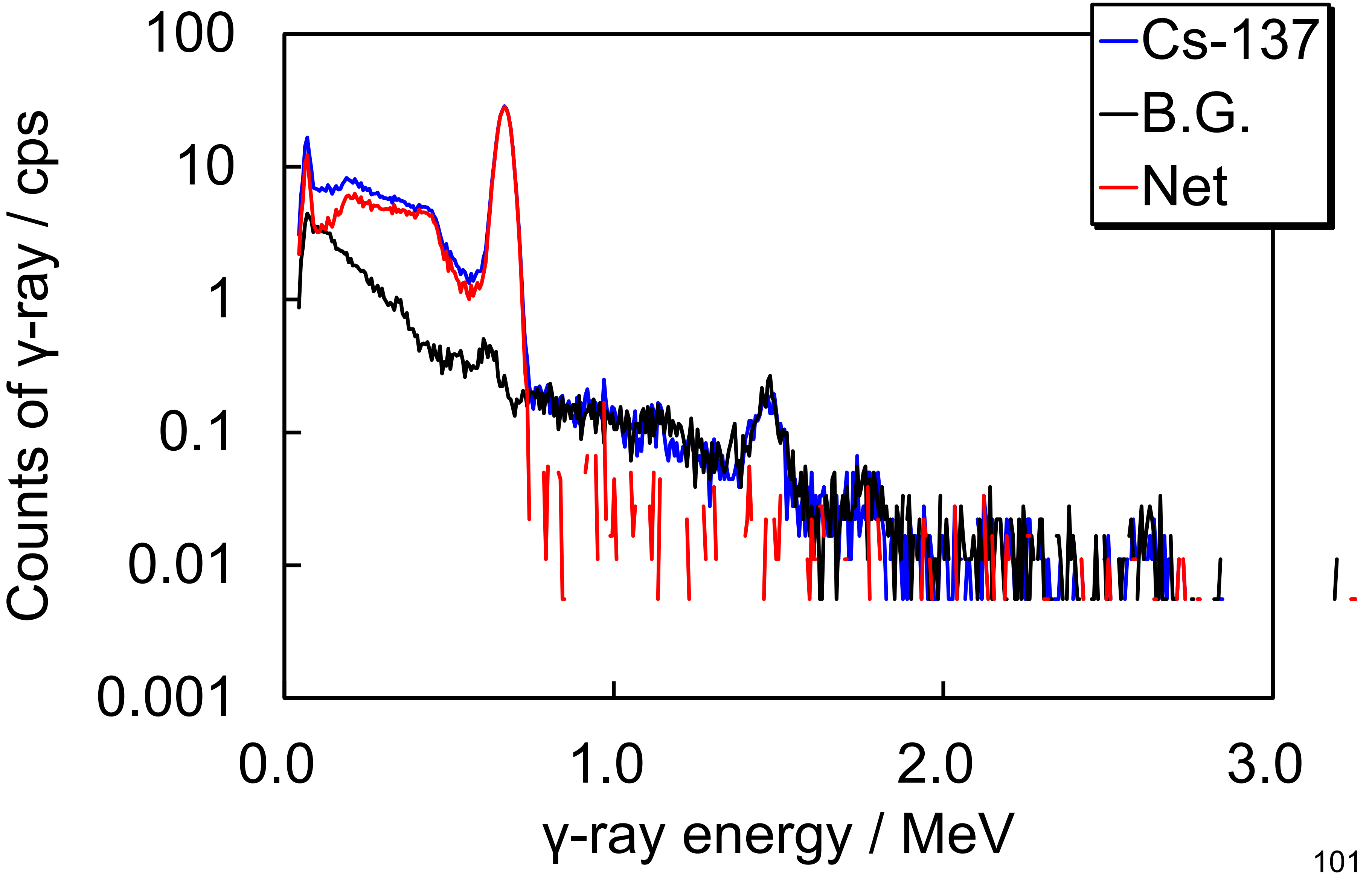
```
         Cursor        Range
Channel  189      0    511
Count    513      0    1333
```

Calibration curves for pulse height channel and γ-ray energy (to which we also add natural K-40 (1.46 MeV))

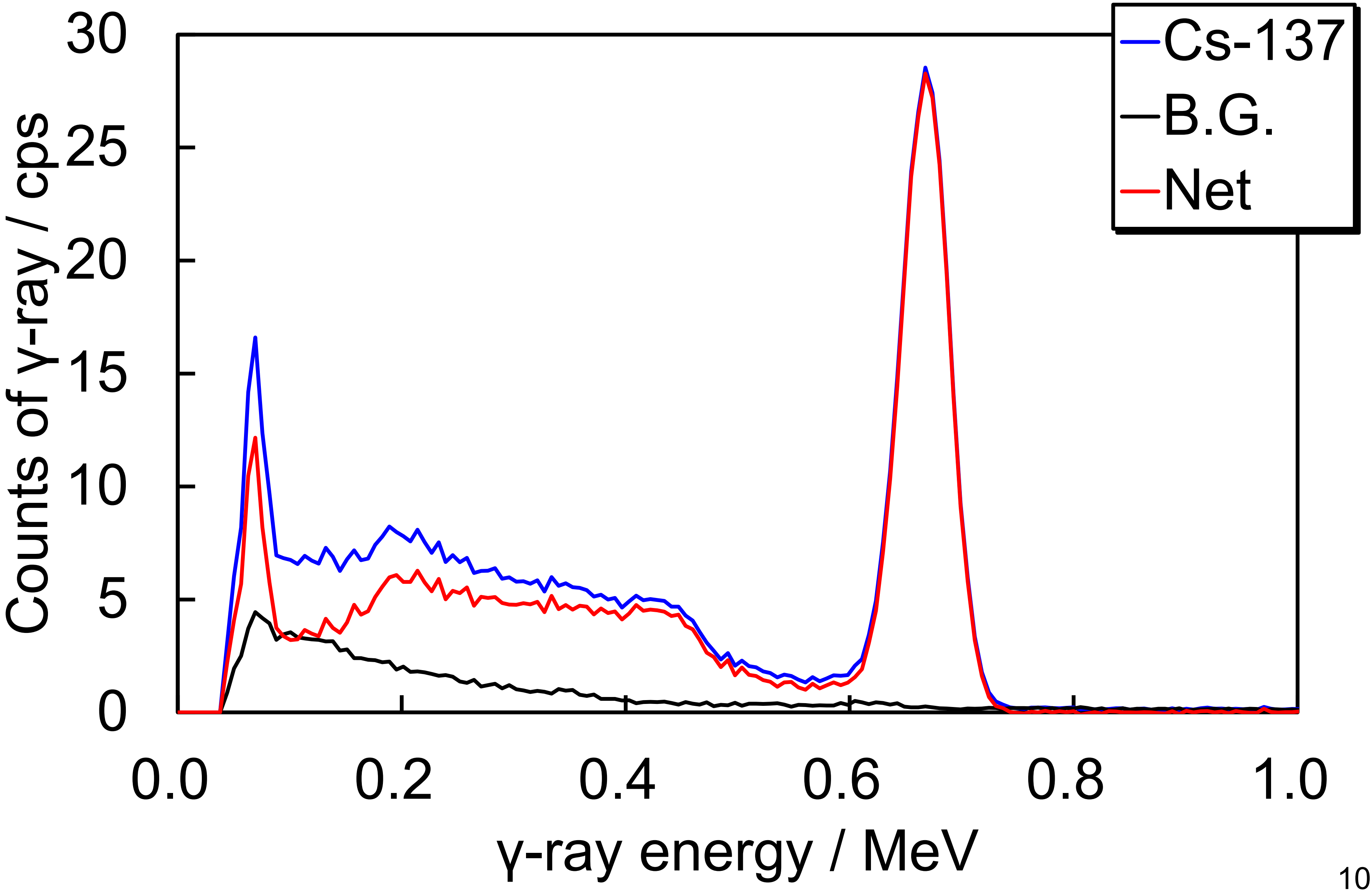


Nuclide	Channel / ch	γ-ray energy / MeV
Annihilation gamma rays	84	0.511
Cs-137	109	0.662
Co-60	189	1.17
Na-22	206	1.275
Co-60	215	1.33

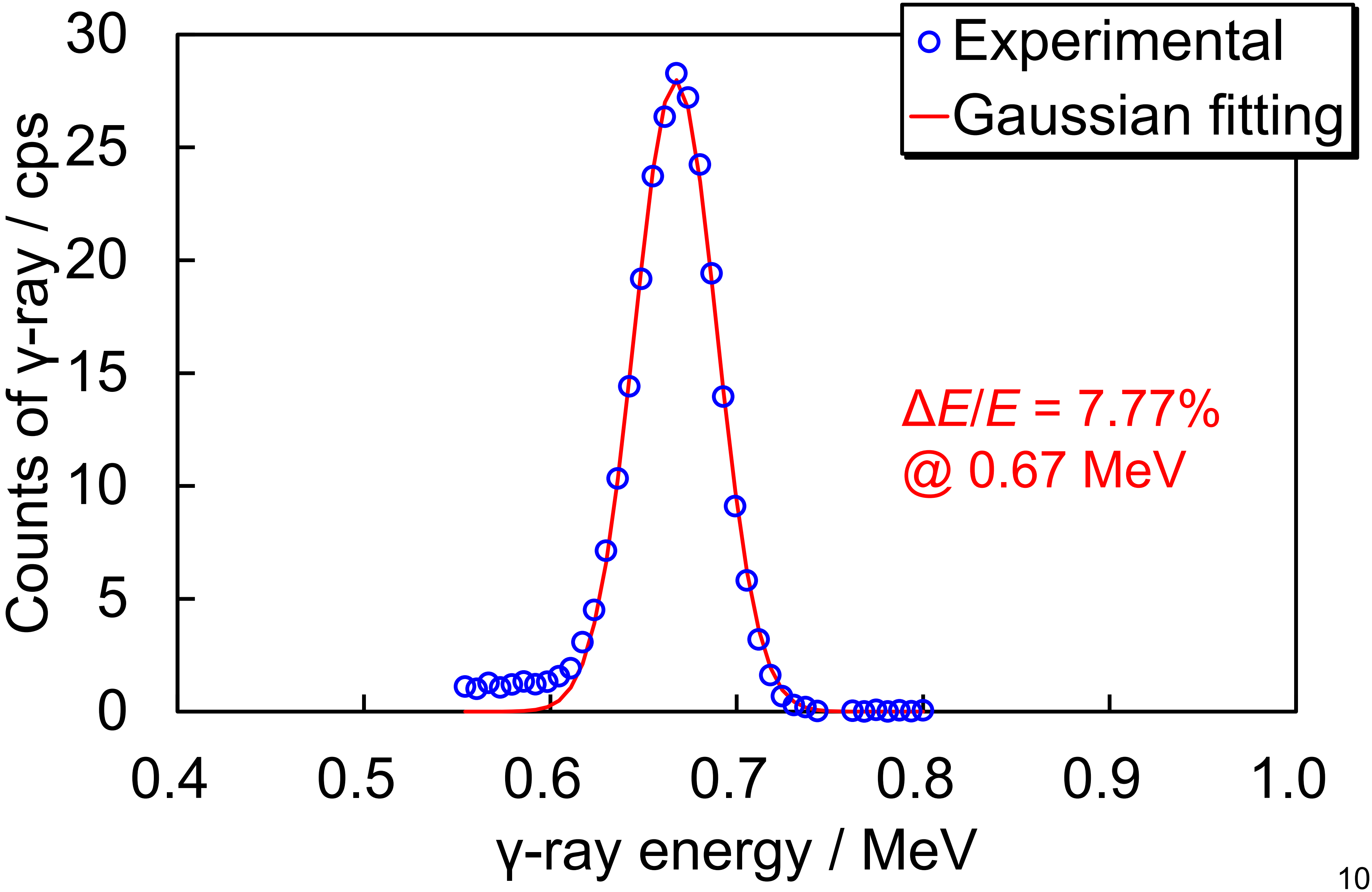
γ-ray spectrum of Cs-137



γ-ray spectrum of Cs-137 (Display switched)



Evaluation of energy resolution (Fitting using Excel solver)



Backgrounds and how to deal with them

Identification and Qualitative Analysis

Element (nuclide) analysis

Measurement of γ-ray spectra of metal samples and nuclide identification

- Measure the γ-ray spectrum of the mystery metal sample using MCA and read the peak center channel of all absorption peaks.
- Estimate the γ-ray energy of a metal sample using an energy calibration curve.
- As a result, which of the following radionuclides is considered to be induced in the metal?

^{64}Cu	0.51 MeV
^{65}Zn	1.11 MeV
^{195m}Pt	0.13 MeV
^{198}Au	0.41 MeV
^{203}Hg	0.28 MeV

Quantitative analysis

Estimation of neutron flux ⇔ Quantitative determination of elements

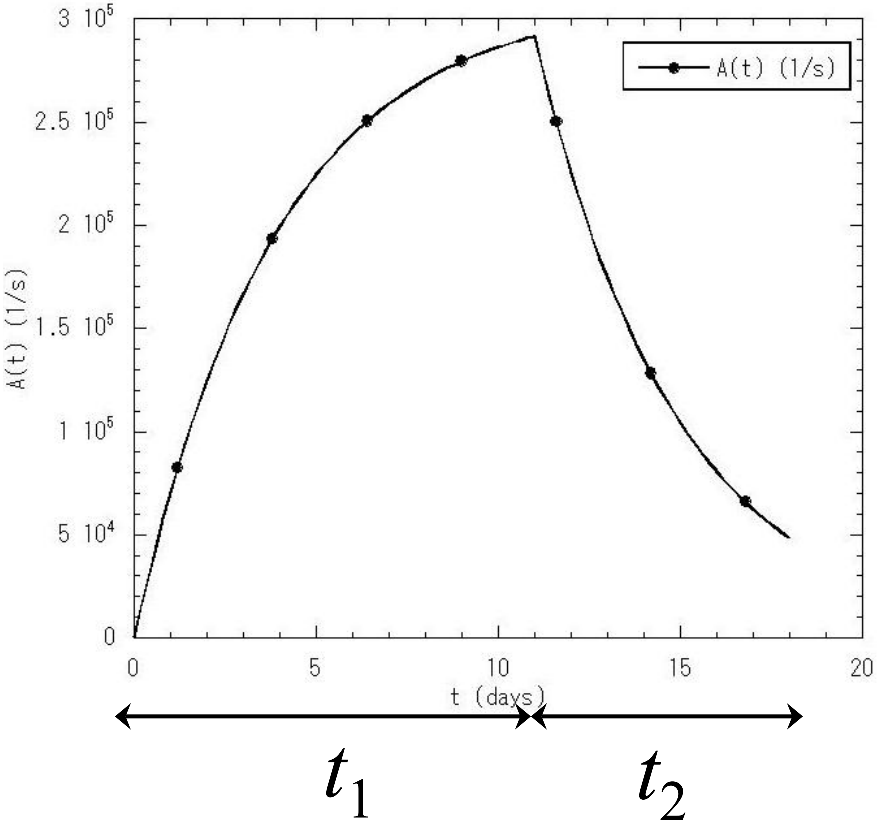
Radioactivity in a sample induced by neutron irradiation

- Average neutron flux in neutron moderator：

  φ （$1/\mathrm{cm}^2/\mathrm{s}$)）
- Number of nuclei to be activated in the sample placed in the neutron moderator：N
- Effective activation cross section of its nuclei：σ（cm^2)
- Decay constants of radioactive nuclei：λ （1/s)
- Radioactivity induced in the sample when only time t_1（s） has elapsed since the start of irradiation.：

  $\varphi N\sigma$（1-exp（-λt_1)） （1/s)

Example of calculation of time variation of radioactivity of a sample



$$A(t_1) = \varphi N \sigma (1 - \exp(-\lambda t_1))$$

$$A(t_2) = A(t_1) \exp(-\lambda t_2)$$

Figure for a sample containing 0.01 g of 197Au irradiated for 11 days with a neutron flux of φ = **10^8（$1/\mathrm{cm}^2/\mathrm{s}$）**

Let's estimate the neutron flux φ

$$A(t_1) = \varphi N \sigma (1 - \exp(-\lambda t_1))$$
$$A(t_2) = A(t_1) \exp(-\lambda t_2)$$

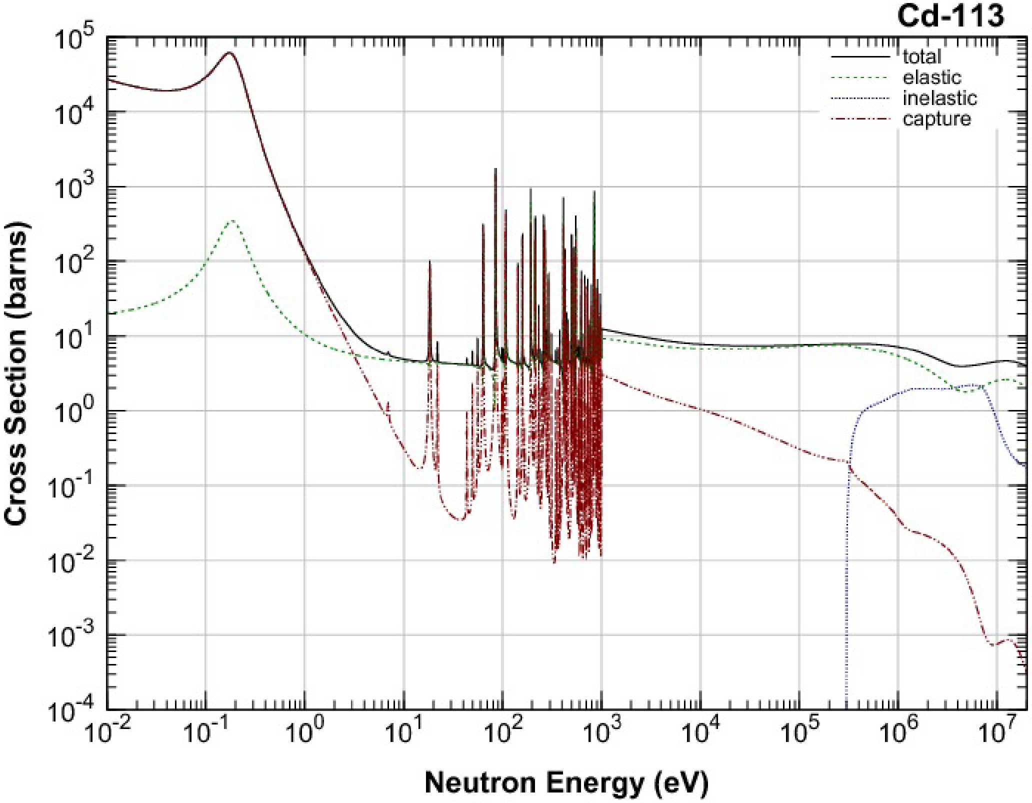
- An irradiated gold foil weights 0.05 g. Then what is the number of nuclei, *N* ?
- Effective activation cross section of ^{197}Au :

$$\sigma_{\text{thermal}} = 9.865 \times 10^{-23} (\text{cm}^2)$$
$$\sigma_{\text{resonance}} = 1.571 \times 10^{-21} (\text{cm}^2)$$

- ^{198}Au decay constant : $\lambda = 2.97 \times 10^{-6} (1/\text{s})$
- t_1: s
- t_2: s

Estimation of thermal and epithermal neutron fluxes using Cd filter

- If a Cd filter is attached to the gold foil, the neutron flux can be estimated for higher energy neutrons than thermal neutrons.
- Subtracting this neutron flux from the flux without Cd, the thermal neutron flux can be derived.



Necessary precautions for accurate radioactivity measurements

- **γ-ray detection efficiency (with γ-ray energy dependence)**
  - Depends on the cross section of the photoelectric effect of the scintillator
  - Almost independent of the energy of the secondary electrons. (γ-ray scintillators are often heavy elements, so the electrons stop almost entirely within the scintillator.)
  - Absorption by window material and self-shielding of the sample must be considered.
- **Geometric detection efficiency**
- **γ-ray emission rate from the target nuclide (γ-rays are not always emitted per decay (transition))**
- **Counting loss**

If the neutron flux φ is known, the N of an identified element can be estimated（Elemental determination）

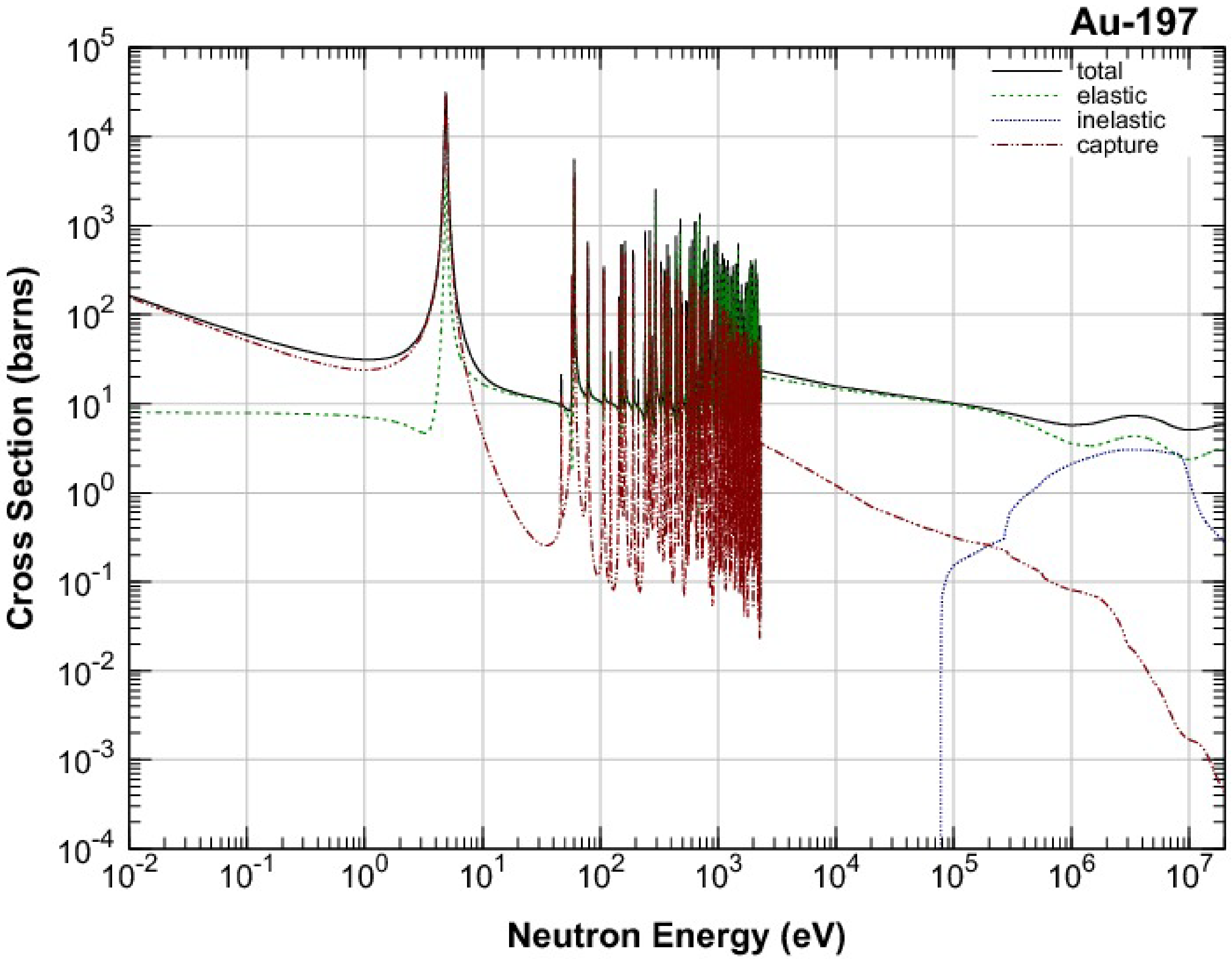
$$A(t_1) = \varphi N \sigma (1 - \exp(-\lambda t_1))$$

$$A(t_2) = A(t_1) \exp(-\lambda t_2)$$

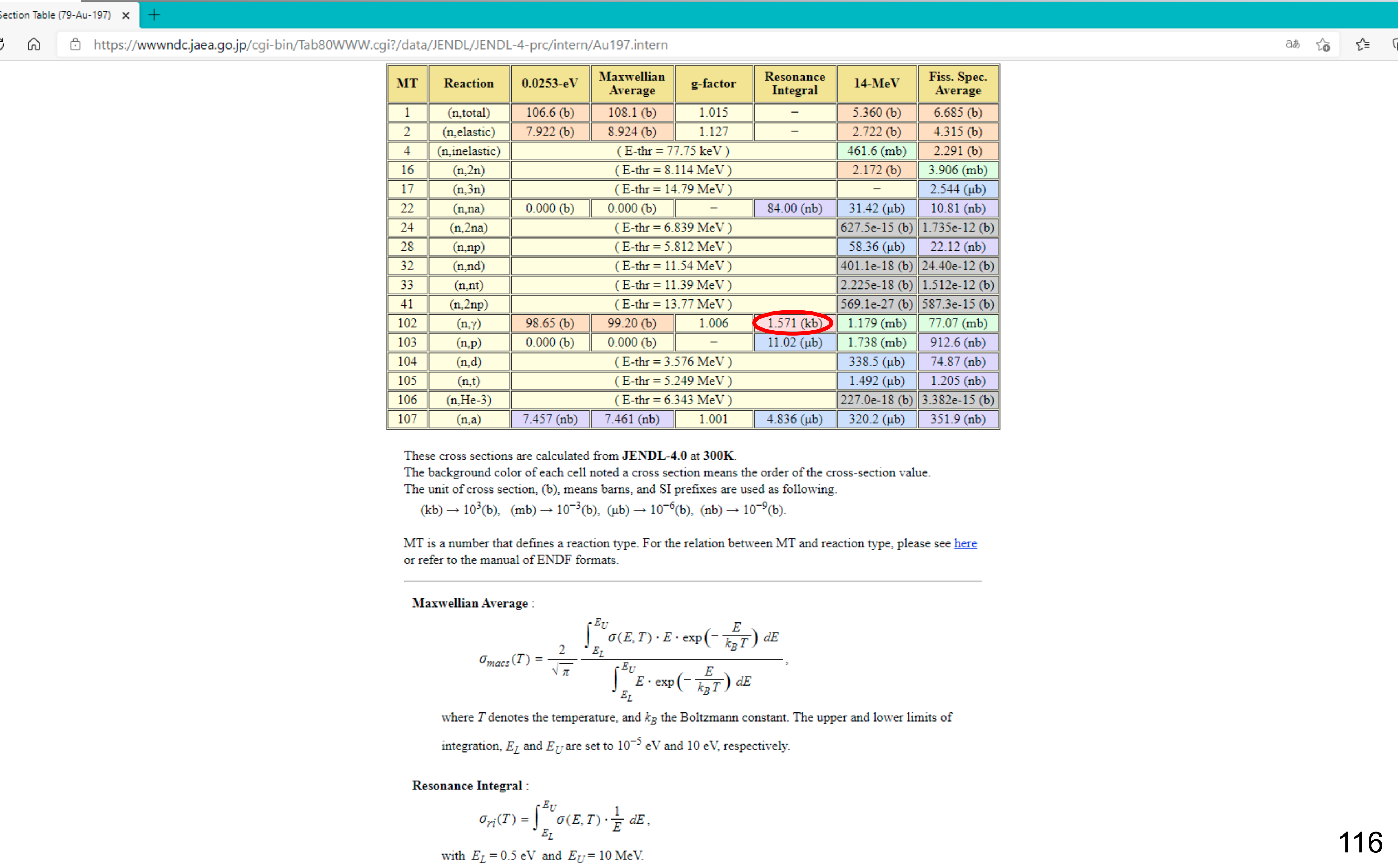
- **Since they have already been identified, σ and λ are known.**

Reference information on quantitative analysis

Cross-section of Au-197 (1) (JENDL-4.0)



Cross section of Au-197 (2) (JENDL-4.0)



https://wwwndc.jaea.go.jp/cgi-bin/Tab80WWW.cgi?/data/JENDL/JENDL-4-prc/intern/Au197.intern

MT	Reaction	0.0253-eV	Maxwellian Average	g-factor	Resonance Integral	14-MeV	Fiss. Spec. Average
1	(n,total)	106.6 (b)	108.1 (b)	1.015	–	5.360 (b)	6.685 (b)
2	(n,elastic)	7.922 (b)	8.924 (b)	1.127	–	2.722 (b)	4.315 (b)
4	(n,inelastic)	(E-thr = 77.75 keV)				461.6 (mb)	2.291 (b)
16	(n,2n)	(E-thr = 8.114 MeV)				2.172 (b)	3.906 (mb)
17	(n,3n)	(E-thr = 14.79 MeV)				–	2.544 (μb)
22	(n,na)	0.000 (b)	0.000 (b)	–	84.00 (nb)	31.42 (μb)	10.81 (nb)
24	(n,2na)	(E-thr = 6.839 MeV)				627.5e-15 (b)	1.735e-12 (b)
28	(n,np)	(E-thr = 5.812 MeV)				58.36 (μb)	22.12 (nb)
32	(n,nd)	(E-thr = 11.54 MeV)				401.1e-18 (b)	24.40e-12 (b)
33	(n,nt)	(E-thr = 11.39 MeV)				2.225e-18 (b)	1.512e-12 (b)
41	(n,2np)	(E-thr = 13.77 MeV)				569.1e-27 (b)	587.3e-15 (b)
102	(n,γ)	98.65 (b)	99.20 (b)	1.006	1.571 (kb)	1.179 (mb)	77.07 (mb)
103	(n,p)	0.000 (b)	0.000 (b)	–	11.02 (μb)	1.738 (mb)	912.6 (nb)
104	(n,d)	(E-thr = 3.576 MeV)				338.5 (μb)	74.87 (nb)
105	(n,t)	(E-thr = 5.249 MeV)				1.492 (μb)	1.205 (nb)
106	(n,He-3)	(E-thr = 6.343 MeV)				227.0e-18 (b)	3.382e-15 (b)
107	(n,a)	7.457 (nb)	7.461 (nb)	1.001	4.836 (μb)	320.2 (μb)	351.9 (nb)

These cross sections are calculated from **JENDL-4.0** at **300K**.
The background color of each cell noted a cross section means the order of the cross-section value.
The unit of cross section, (b), means barns, and SI prefixes are used as following.

(kb) → 10^3(b), (mb) → 10^{-3}(b), (μb) → 10^{-6}(b), (nb) → 10^{-9}(b).

MT is a number that defines a reaction type. For the relation between MT and reaction type, please see here or refer to the manual of ENDF formats.

Maxwellian Average :

$$\sigma_{macs}(T) = \frac{2}{\sqrt{\pi}} \frac{\displaystyle\int_{E_L}^{E_U} \sigma(E,T) \cdot E \cdot \exp\left(-\frac{E}{k_B T}\right) dE}{\displaystyle\int_{E_L}^{E_U} E \cdot \exp\left(-\frac{E}{k_B T}\right) dE},$$

where T denotes the temperature, and k_B the Boltzmann constant. The upper and lower limits of integration, E_L and E_U are set to 10^{-5} eV and 10 eV, respectively.

Resonance Integral :

$$\sigma_{ri}(T) = \int_{E_L}^{E_U} \sigma(E,T) \cdot \frac{1}{E}\, dE,$$

with $E_L = 0.5$ eV and $E_U = 10$ MeV.

Why $\varphi N\sigma(1-\exp(-\lambda t))$?

$$\frac{dA(t)}{dt} = -\lambda A(t) + \lambda\varphi N\sigma$$

Initial condition is $A(0) = 0$

- First term on the right-hand side：Decay of radioactivity per second
- Second term on the right-hand side：Increase in radioactivity per second due to radiation capture
- $\varphi N\sigma$: Saturated radioactivity

Decay of radioactivity

- **Radioactivity of the sample at any time after the end of irradiation :$A(0)$（1/s）**
- **The radioactivity of the sample at the time t_2（s） elapsed from that point. :$A(0)\exp$（$-\lambda t_2$）（1/s）**

$$\frac{dA(t)}{dt} = -\lambda A(t)$$

Initial condition is A(0) at t = 0