Application of NRF to Nondestructive Assay of Nuclear Material

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Japan Atomic Energy Agency

Application of Compton Based Gamma Rays
FACET-II
Science Opportunities Workshops
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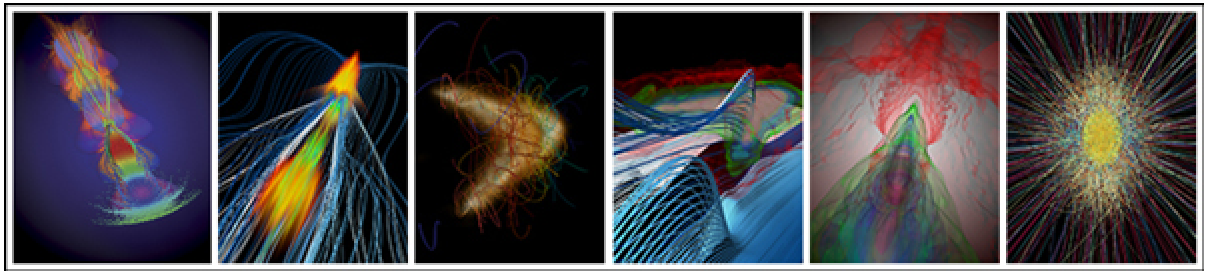

Collaborators

- Quantum Beam Science Center, JAEA
  - Laser Compton Scattered Gamma-ray Research Group
    T. Hayakawa, T. Shizuma, C.T. Angell,
    M. Sawamura, R. Nagai, N. Nishimori, M. Omer
  - Advanced Laser Development Group
    M. Mori
- Integrated Support Center for Nuclear Nonproliferation and Nuclear Security, JAEA
  - M. Seya, M. Koizumi
- KEK
  - H. Kawata, Y. Kobayashi and cERL team
  - J. Urakawa, H. Terunuma, A. Kosuge, T. Akagi
- Kyoto Univ.
  - H. Ohgaki
- Osaka Univ.
  - M. Fujiwara

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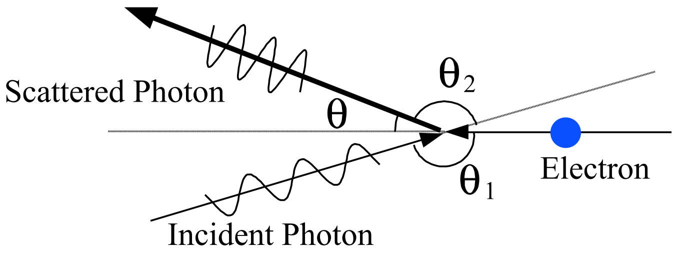
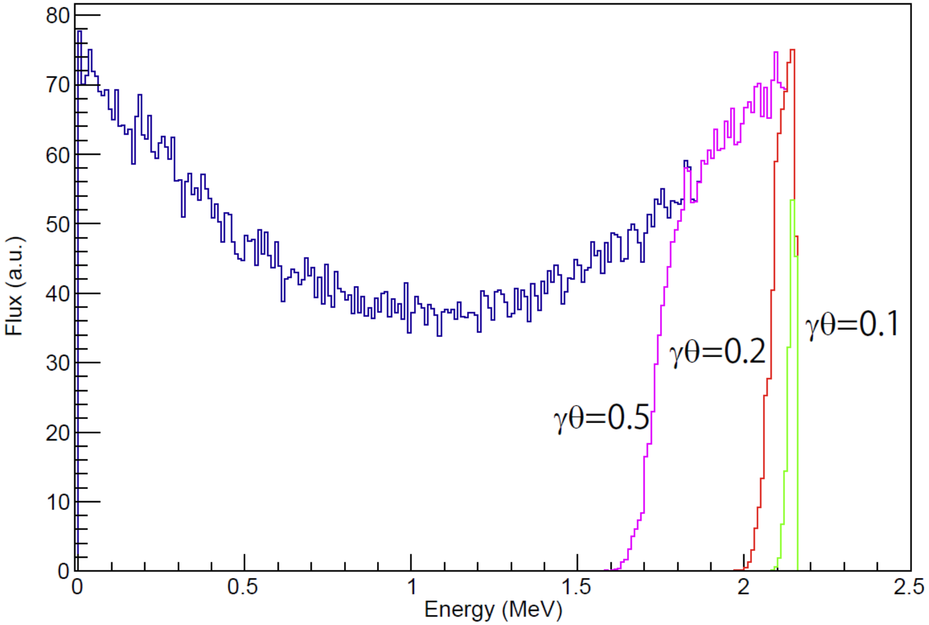
- ➢ A government (MEXT) subsidy for strengthening nuclear security (R. Hajima),
- ➢ Photon and Quantum Basic Research Coordinated Development Program from the MEXT (N. Terunuma)
- ➢ Funds for Integrated Promotion of Social System Reform and Research and Development (H. Ohgaki)

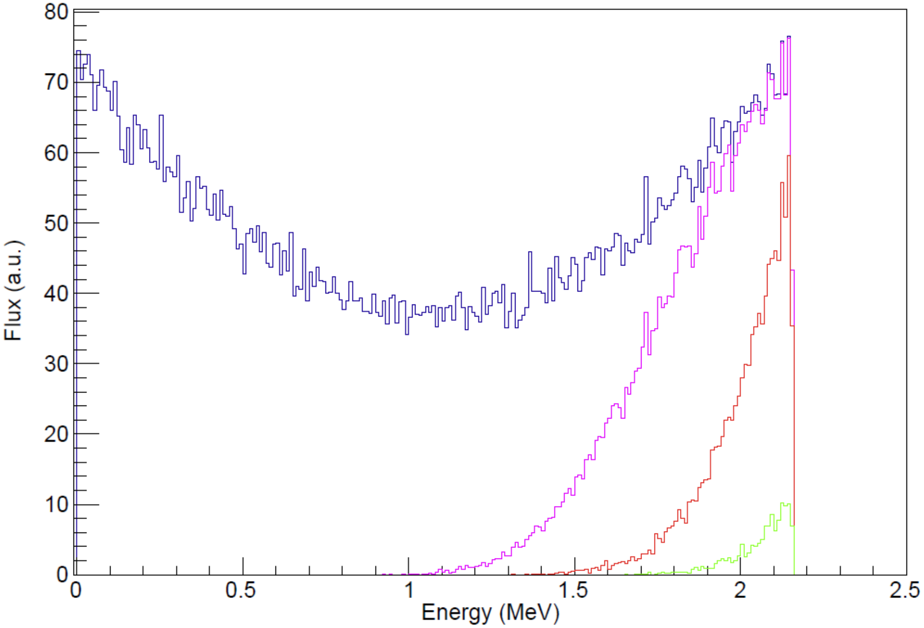
- principle of NRF-NDA
- Nuclear security
- Nuclear safeguards
  - Branch transition NRF
  - Cryo-NRF
  - Integral Resonance Transmission
  - Demo exp. for debris in a TMI-2 container
- LCS X-ray generation at Compact ERL

Laser Compton Scattering (LCS)



- ✓ Pencil like beam
- ✓ Energy Tunable & quasi-monochromatic
- ✓ Polarized (linear and circular)





electron beam
E = 350 MeV
ε_{nx} = 1 mm-mrad
σ_x = 30 μm
collision angle = 10 deg.

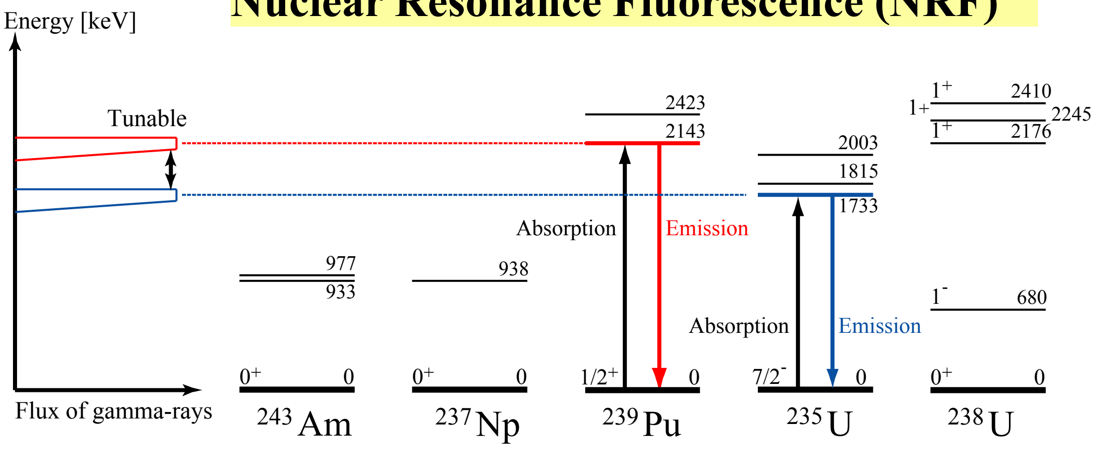
laser beam
λ = 1064 nm
σ_x = 30 μm

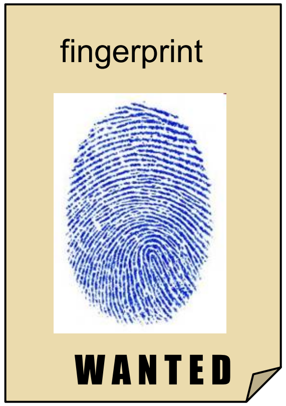
electron beam
E = 350 MeV
ε_{nx} = 5 mm-mrad
σ_x = 30 μm
collision angle = 10 deg.

laser beam
λ = 1064 nm
σ_x = 30 μm

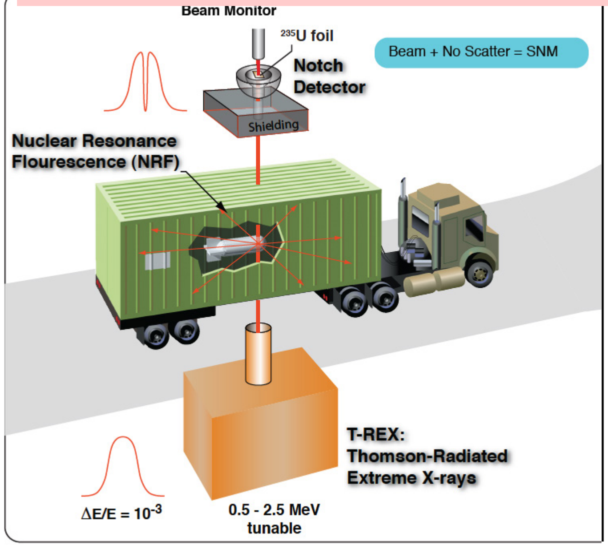
Nondestructive Detection & Measurement of Nuclear Material

Nuclear Resonance Fluorescence (NRF)



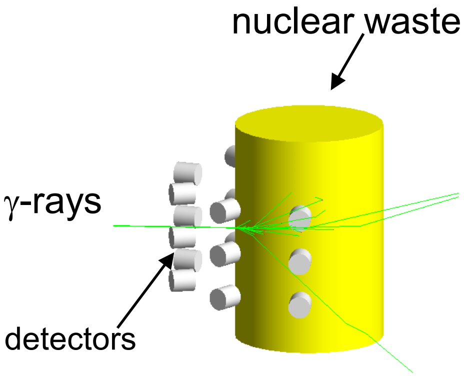


Detection of SNM in a cargo



SNM: special nuclear material

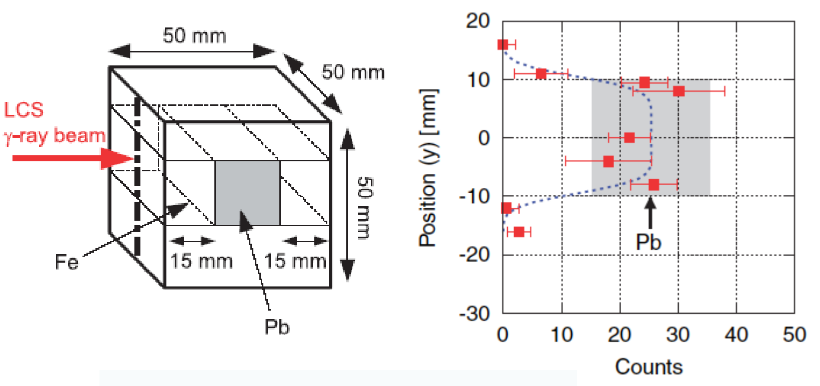
Management of nuclear material



detection and assay of isotopes
- -- U, Pu, and Minor Actinides
- -- alpha emitter
- -- difficult to measure by passive assay

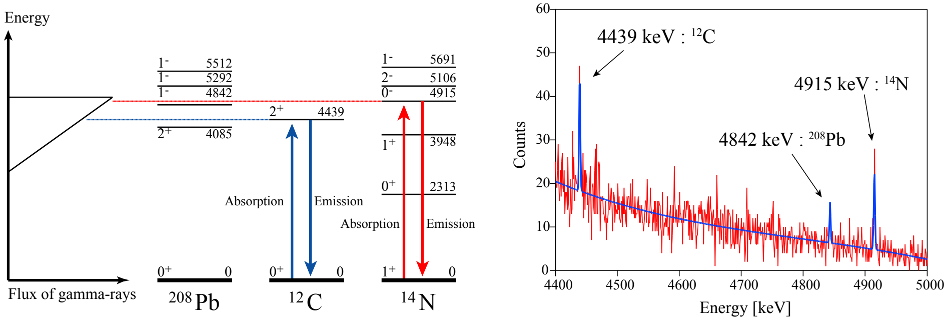
R. Hajima et al., J. Nucl. Sci. Tech. 45, 441 (2008)
J. Pruet et al., J. App. Phys. 99, 123102 (2006)

Experimental Demonstration – nondestructive detection of isotope



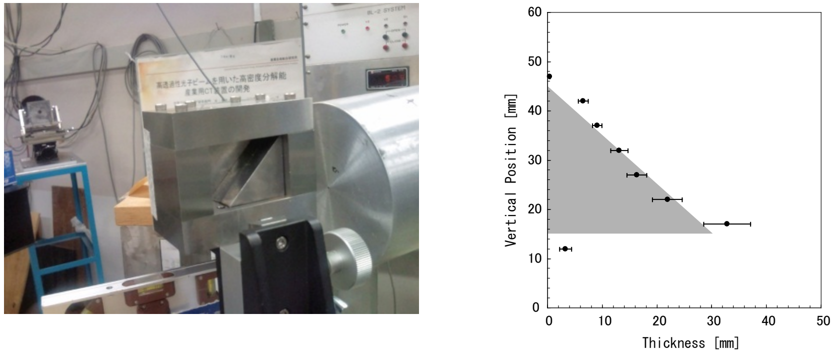
1-d isotope mapping

N. Kikuzawa et al., APEX 2, 036502 (2009).



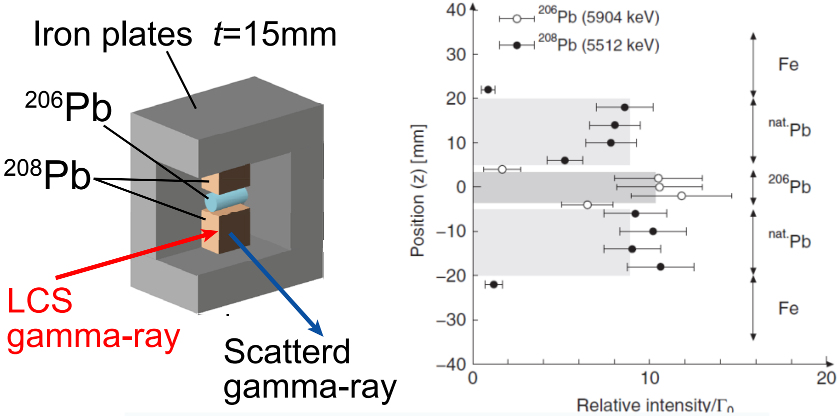
Detection of two isotopes

T. Hayakawa et al., RST 80, 045110 (2009).



2-d isotope mapping

H. Toyokawa et al., JJAP 50, 100209 (2011).



1-d mapping of two isotopes

T. Shizuma et al., RSI 83, 015103 (2011).

Nuclear Security

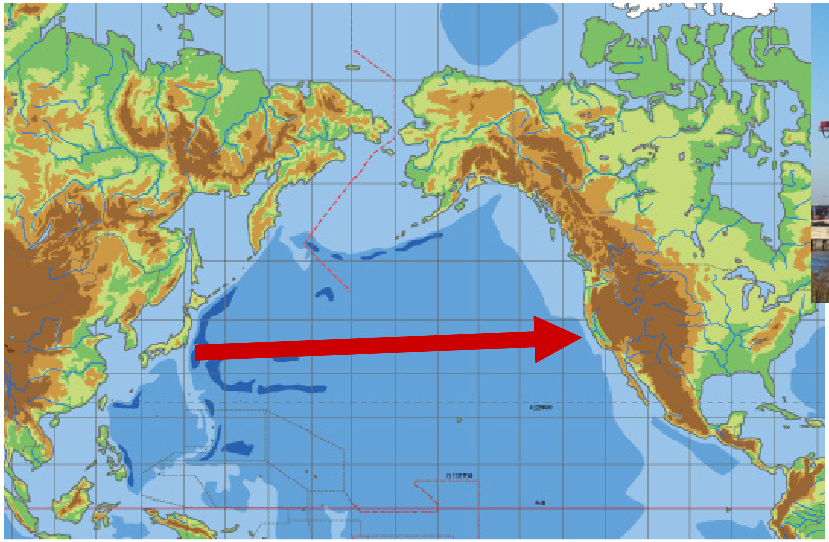
References:

H. Ohgaki et al., J. Korean Phys. Soc. 59, 3155 (2011).

I. Daito et al., J. Plasma Fusion Res. 88, 553 (2012)
R. Hajima et al., IPAC-2013

I. Daito et al., Proc. IEEE-NSS (2014)

Nuclear Security Application



From Yokohama
11,919 TEUs (2010. Jan.) ⟷ About 400 TEU / day

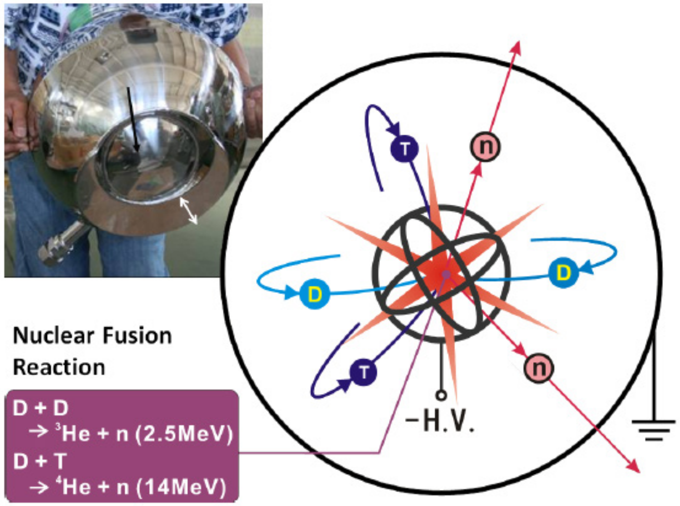
TEU= 20ft Container

Possible Terrorism = Nuclear Material Hidden in a Cargo

Neutron/gamma-ray hybrid system

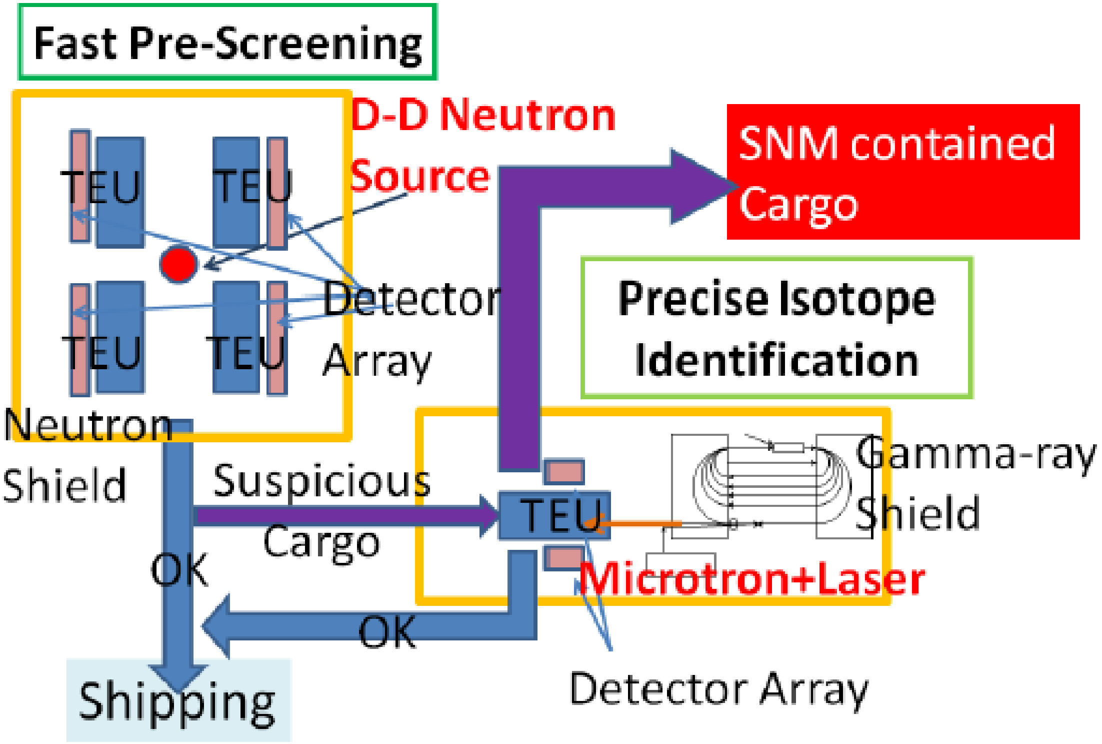
H. Ohgaki et al., J. Korean Phys. Soc. 59, 3155 (2011).

R&D Program (2010-2014) conducted by Kyoto U. and JAEA



Nuclear Fusion Reaction

D + D
→ ^{3}He + n (2.5MeV)
D + T
→ ^{4}He + n (14MeV)



Fast Pre-Screening

D-D Neutron Source

Detector Array

Neutron Shield

SNM contained Cargo

Precise Isotope Identification

Suspicious Cargo

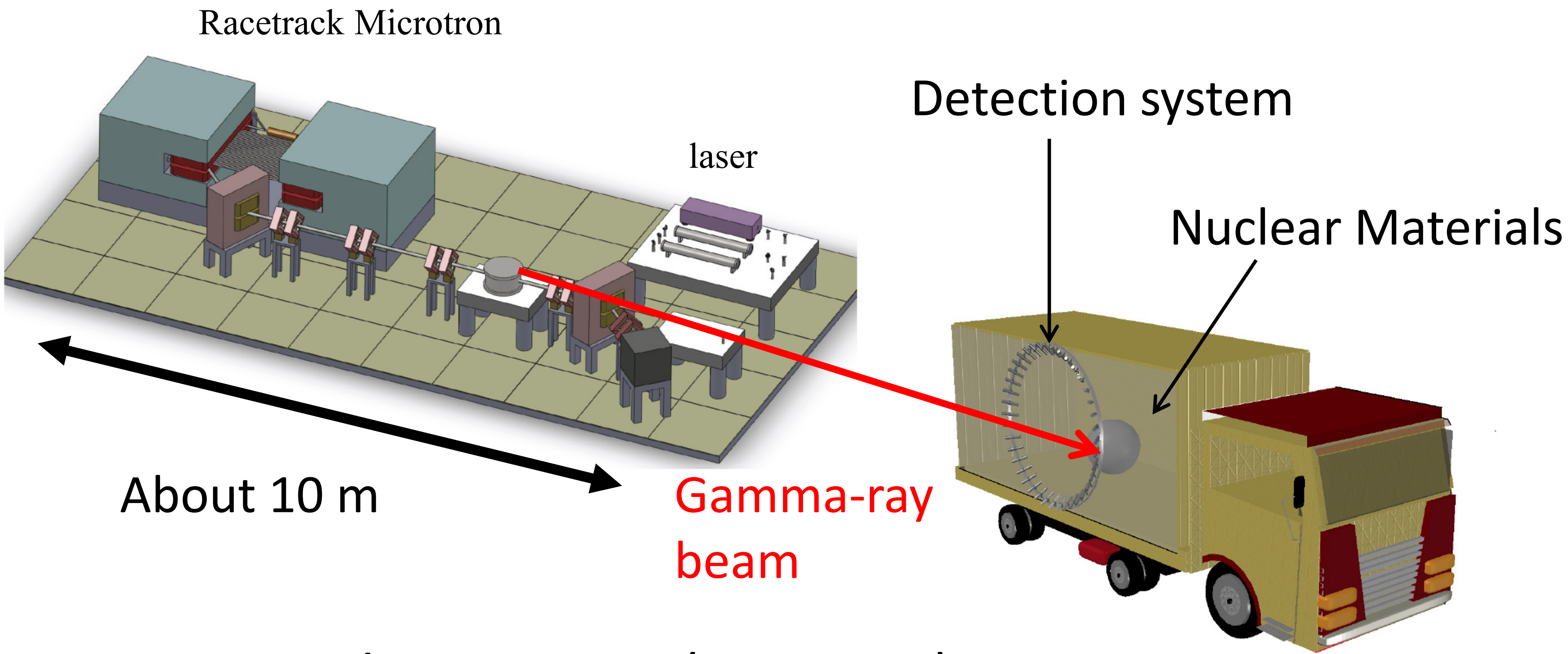
Gamma-ray Shield

Microtron+Laser

Detector Array

Shipping

Nuclear Material Detection by LCS γ-ray



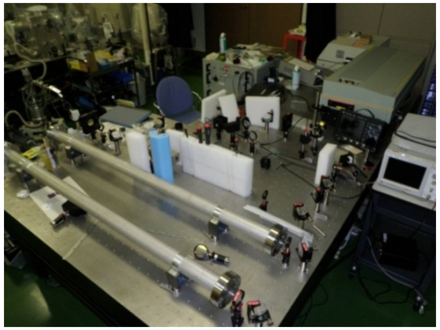
- Racetrack Microtron (250 MeV)
- Laser system (Nd:YAG 2 ω)

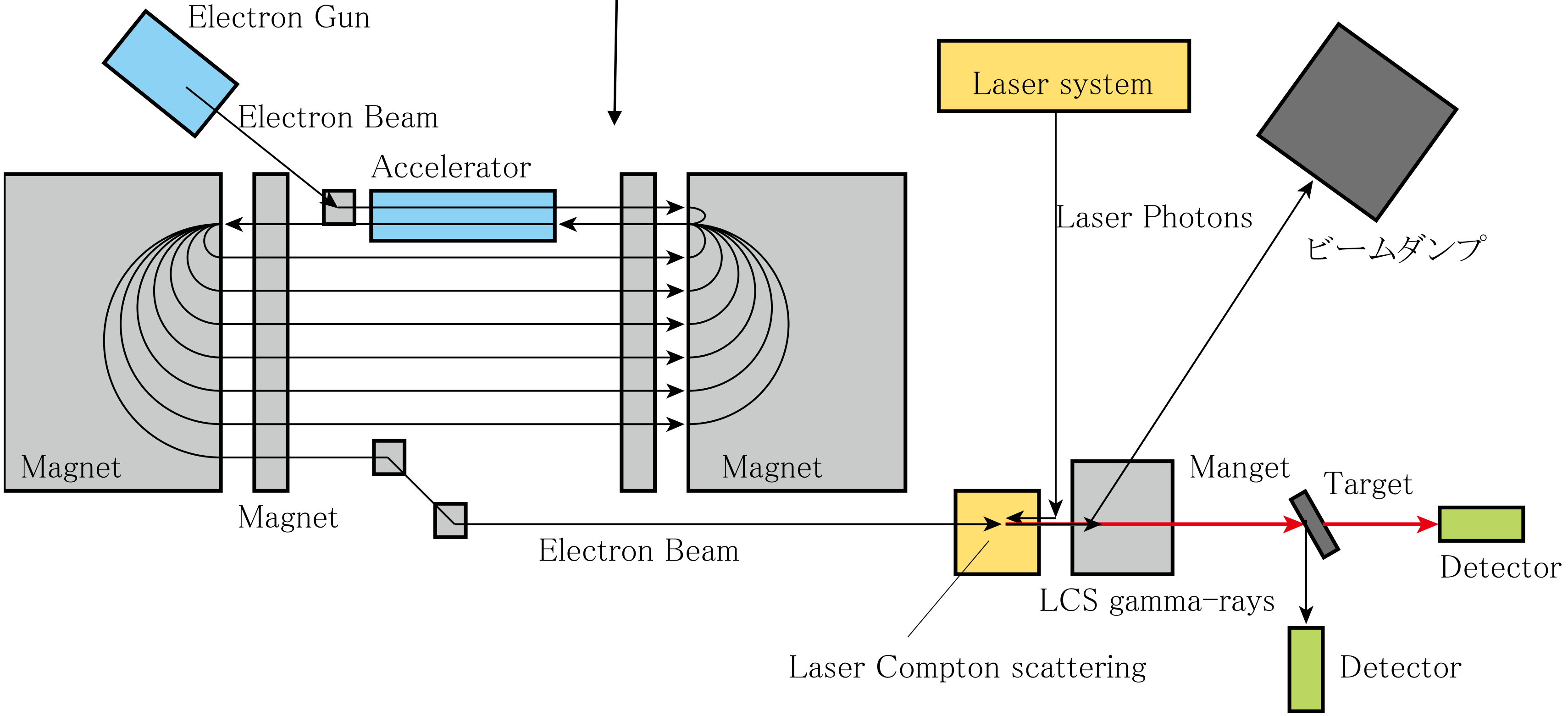
PoP experiment at JAEA

150 MeV Microtron
60 pC,
20ps (FWHM)

400mJ、200ps
（FWHM）

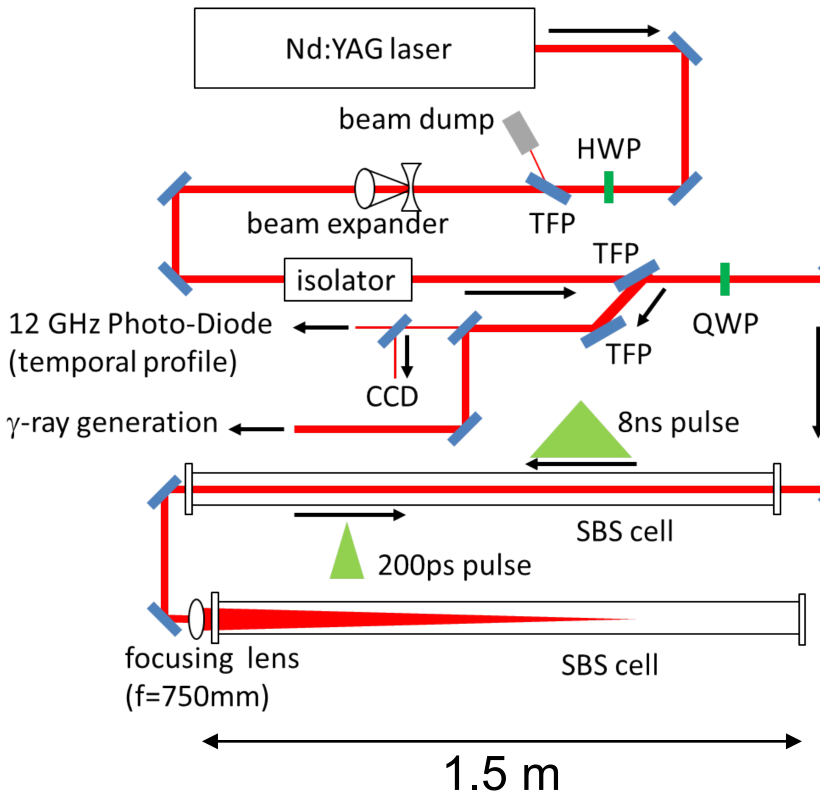
SBS Pulse compression

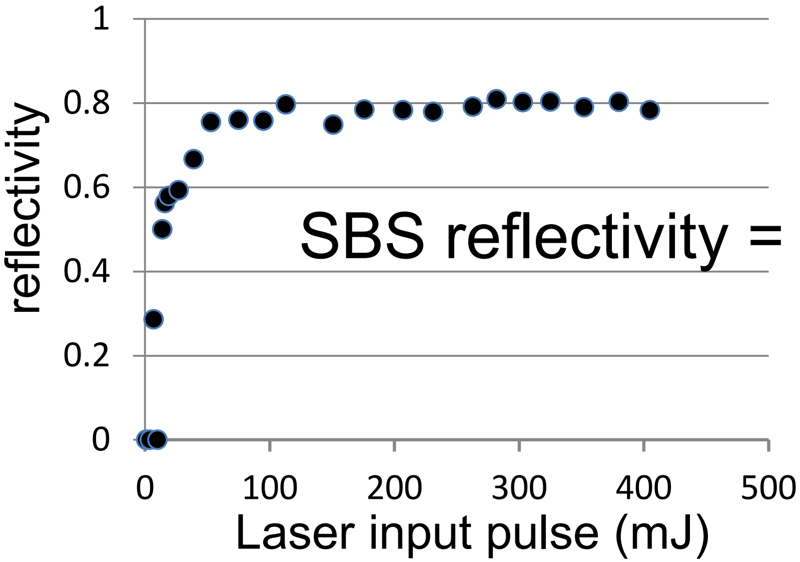




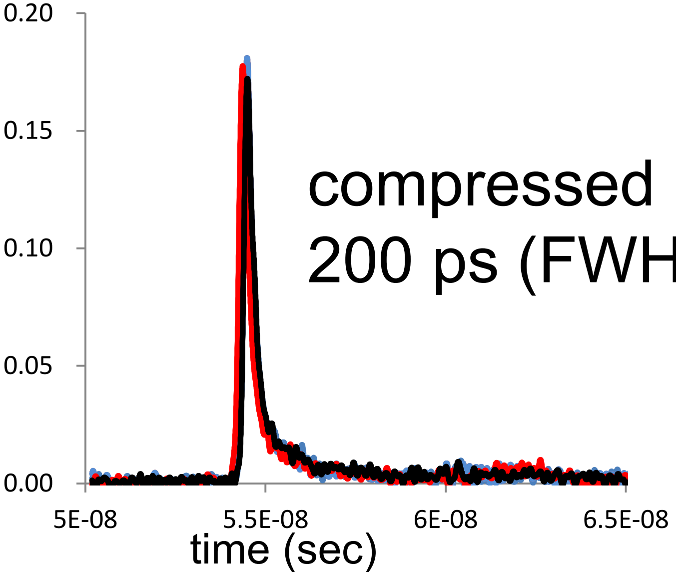
Laser pulse compression

I. Daito et al., J. Plasma Fusion Res. 88, 553 (2012)
R. Hajima et al., IPAC-2013





SBS reflectivity = 80%



compressed pulse
200 ps (FWHM)

SBS: Stimulated Brillouin Scattering

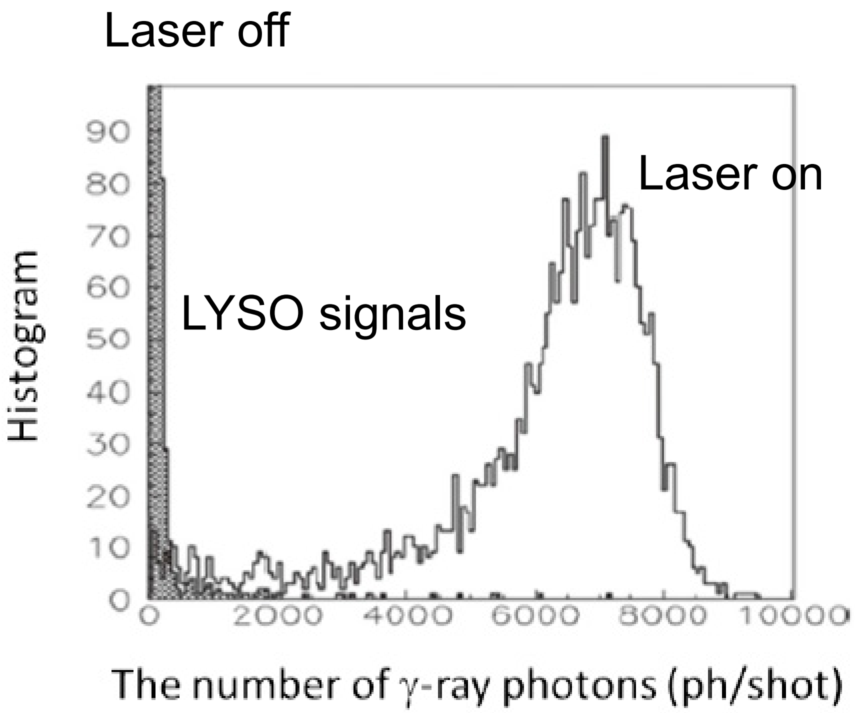
2x 1.5-m SBS cells with FC-40 Fluorinert

Evaluation of gamma-ray flux

Gamma-ray flux has been evaluated by two methods:

(1) Total energy measurement by LYSO

(2) Compton scattering from an Al plate measured by GSO



Microtron:
150 MeV, 45 pC, 20 ps (FWHM)

Laser:
1064 nm, 178 mJ, 8 ns → 200 ps
f=2.3 m, angle= 1.5 deg.

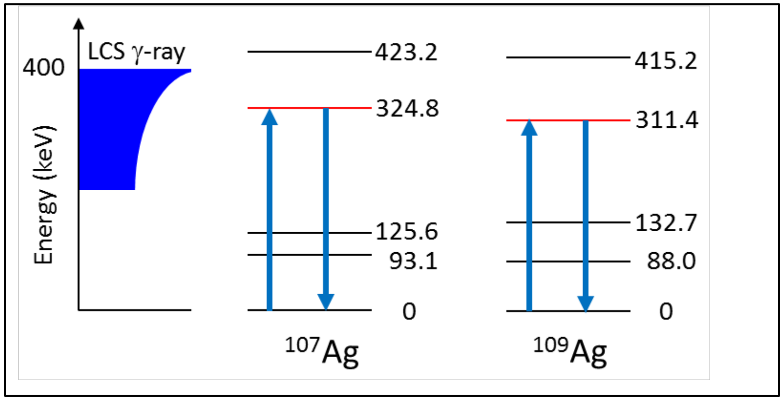
LYSO → $(6.3\pm1.5) \times 10^3$ /shot
GSO → $(5.58\pm0.24) \times 10^3$ /shot

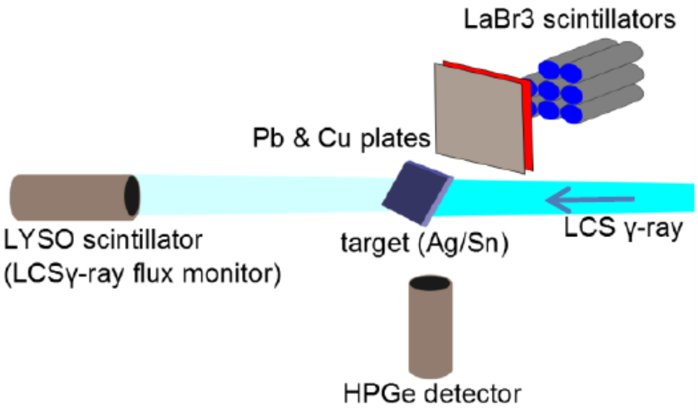
⇓

3.5×10^5/s @10Hz, 1J

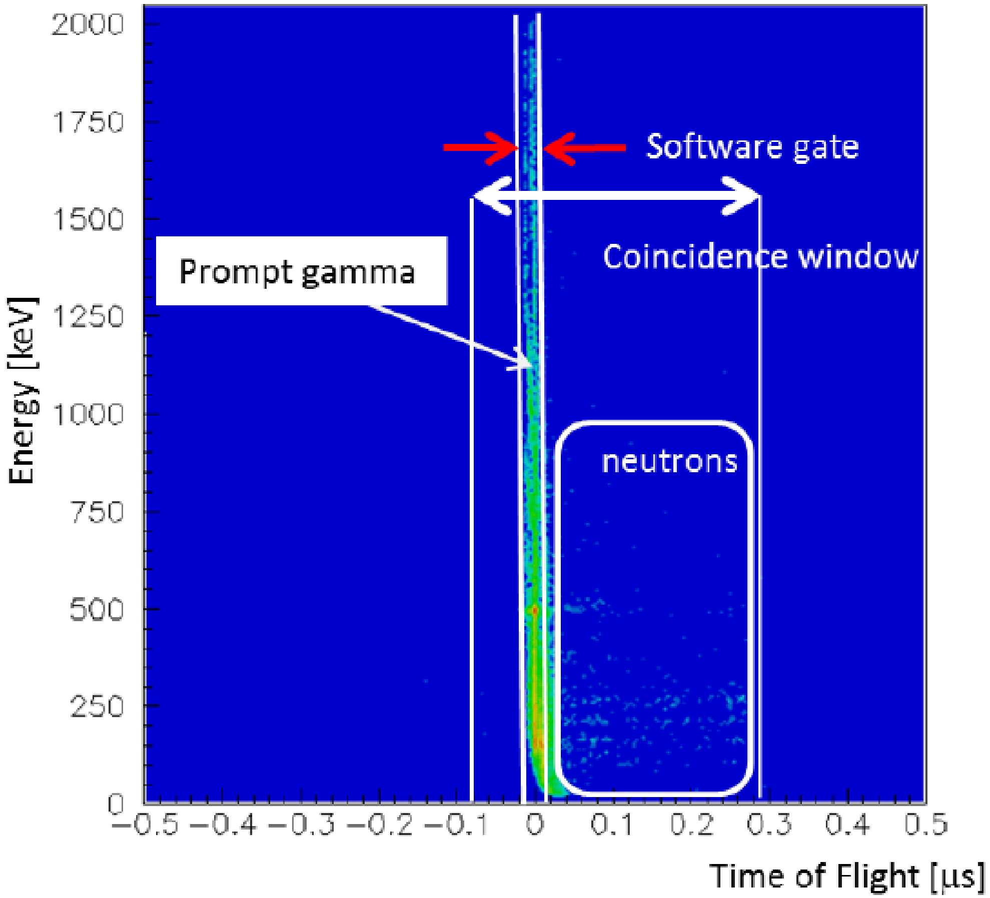
I. Daito et al., J. Plasma Fusion Res. 88, 553 (2012)
R. Hajima et al., IPAC-2013

PoP experiment: Detection of Ag





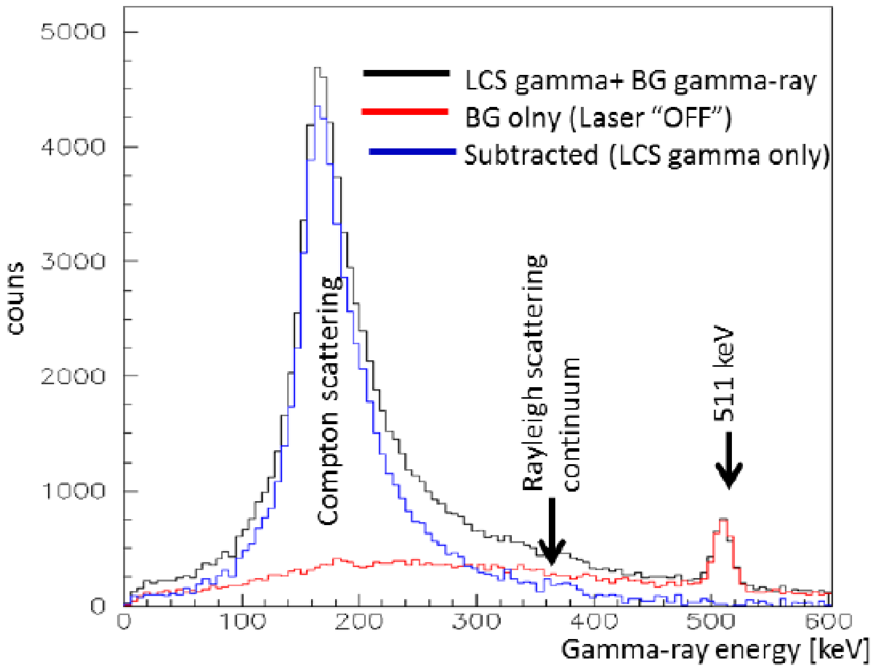
LaBr$_3$ signal



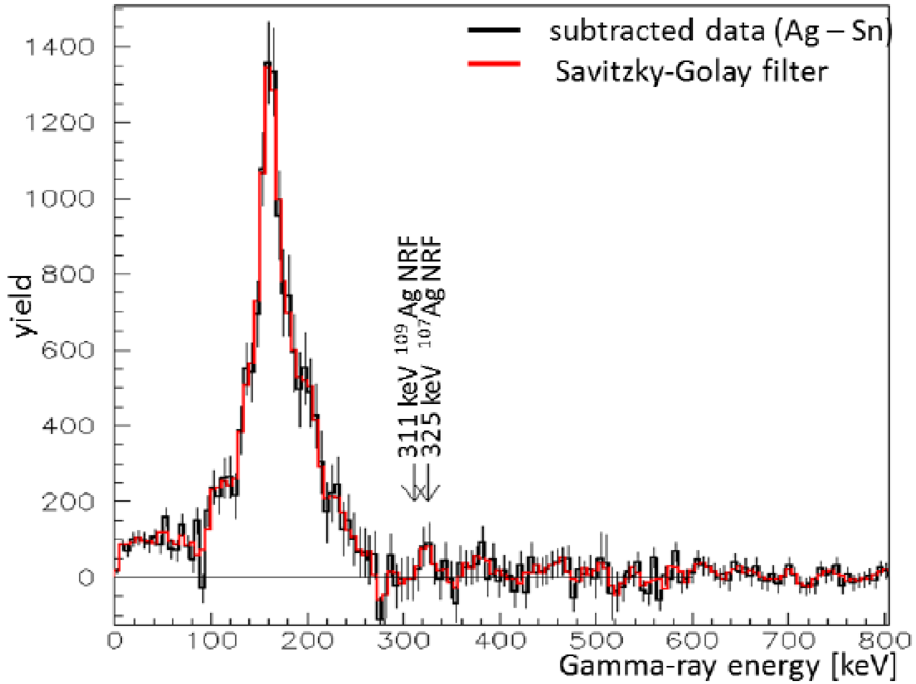
I. Daito et al., Proc. IEEE-NSS (2014)

PoP experiment: Detection of Ag

I. Daito et al., Proc. IEEE-NSS (2014)



LBr$_3$ signal (Ag target)



LBr$_3$ signal (Ag-Sn)

Peak area of Ag NRF signals, (2.2±1.1) x 10^2, is consistent with an expected signals, 1.1 x 10^2, from a Geant4 simulation.

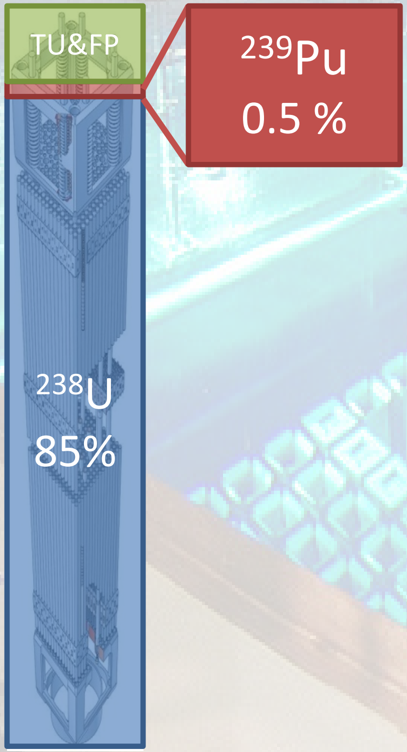
Nuclear Safeguards

References:

R. Hajima et al.,
Proposal of nondestructive radionuclide assay using
a high-flux gamma-ray source and nuclear resonance fluorescence,
J. Nucl. Sci. Tech. 45, 441 (2008).
T. Hayakawa et al.,
Nondestructive assay of plutonium and minor actinide in spent fuel
using nuclear resonance fluorescence with laser Compton scattering gamma-rays
NIM-A 621, 695 (2010).
T. Shizuma et al.,
Statistical uncertainties of nondestructive assay for spent nuclear fuel
by using nuclear resonance fluorescence
NIM-A 737, 170 (2014).

Non-Destructive Assay of ^{239}Pu

^{239}Pu Small Fraction of Spent Fuel

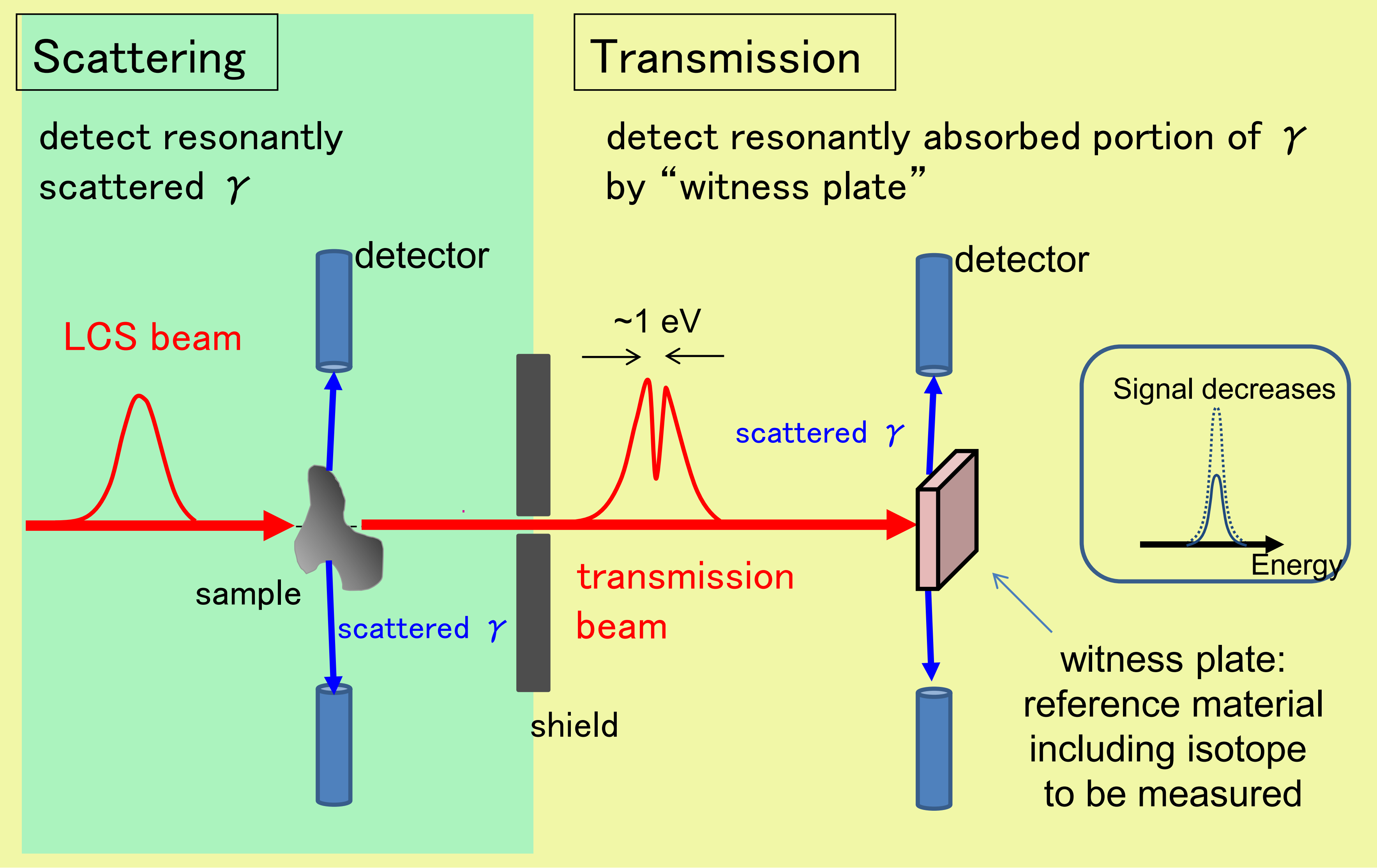


Challenge: Assay ^{239}Pu Non-destructively in spent nuclear fuel

Goal:

Measure ^{239}Pu mass to 1%

Two Measurement Methods



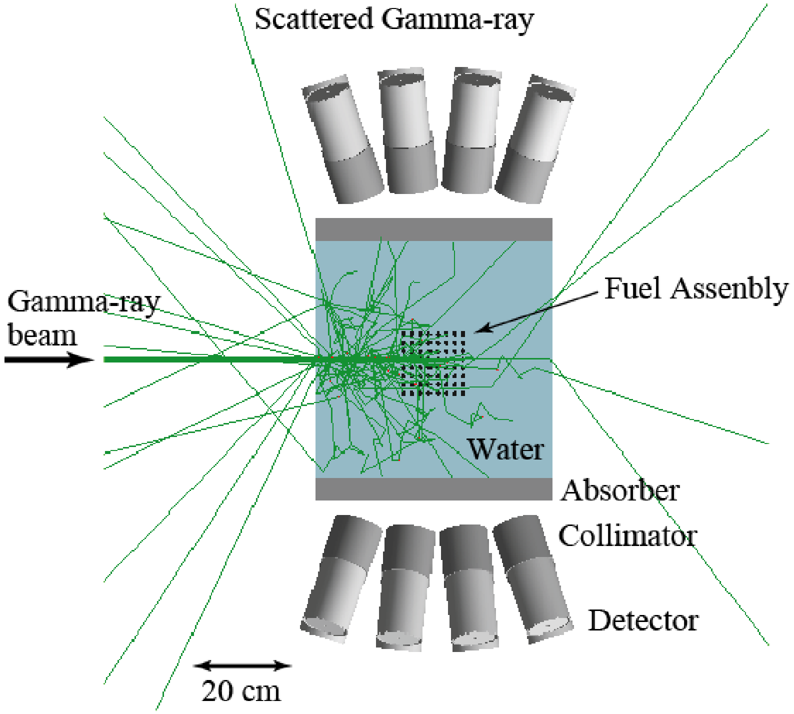
Scattering

detect resonantly scattered γ

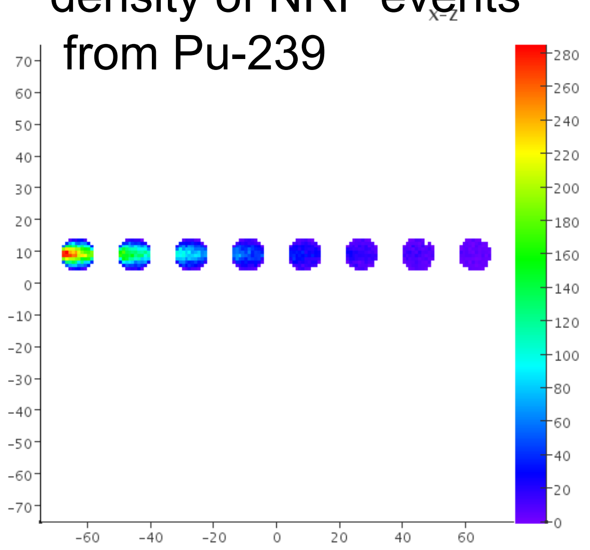
Transmission

detect resonantly absorbed portion of γ by “witness plate”

Spent Fuel NDA simulation



density of NRF events from Pu-239



T. Hayakawa, et al. NIMA (2010).

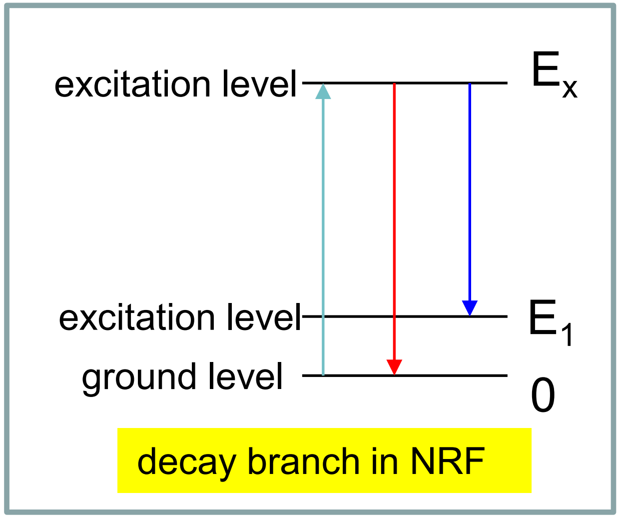
Spent fuel with 1% PuO_2

Statistical error of 2% with 4000-sec measurement

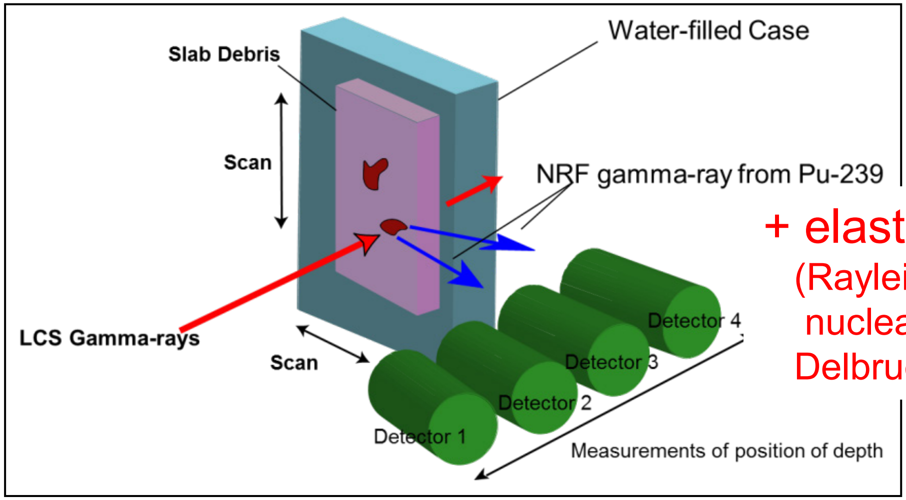
But the simulation does not include elastic scattering
– Rayleigh, nuclear Thomson and Delbruck scattering

Elastic scattering: ~0.5 mb x 1 keV bin = ~5 eV b ⟺ NRF: 10-50 eV b

Detection of decay branch in NRF



decay branch in NRF



+ elastic scattering
(Rayleigh,
nuclear Thomson,
Delbruck scattering)

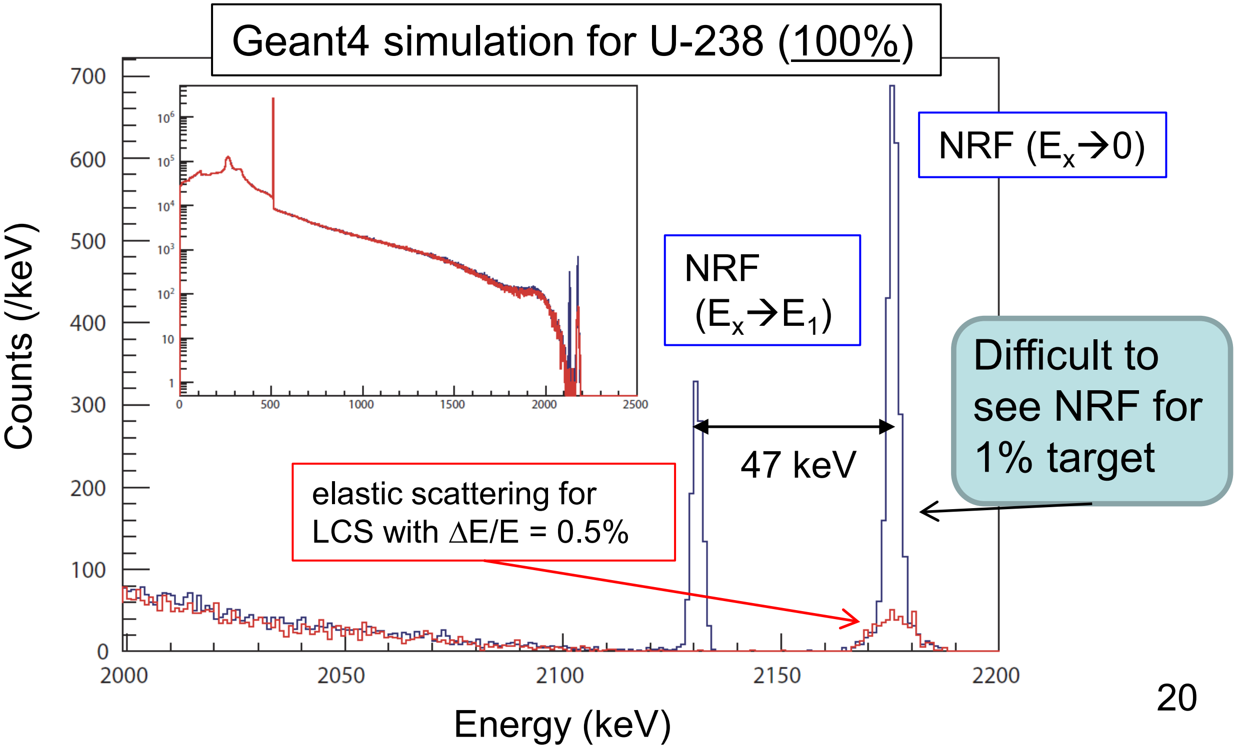
gamma-ray bandwidth

existing LCS : ΔE/E ~ 3-5%

future LCS : ΔE/E < 0.5%

decay branch can separate NRF from elastic scattering

Geant4 simulation for U-238 (100%)



NRF (E_x→0)

NRF
(E_x→E_1)

47 keV

elastic scattering for
LCS with ΔE/E = 0.5%

Difficult to see NRF for 1% target

T. Shizuma et al., Nucl. Instr. Meth. A 737, 170 (2014)

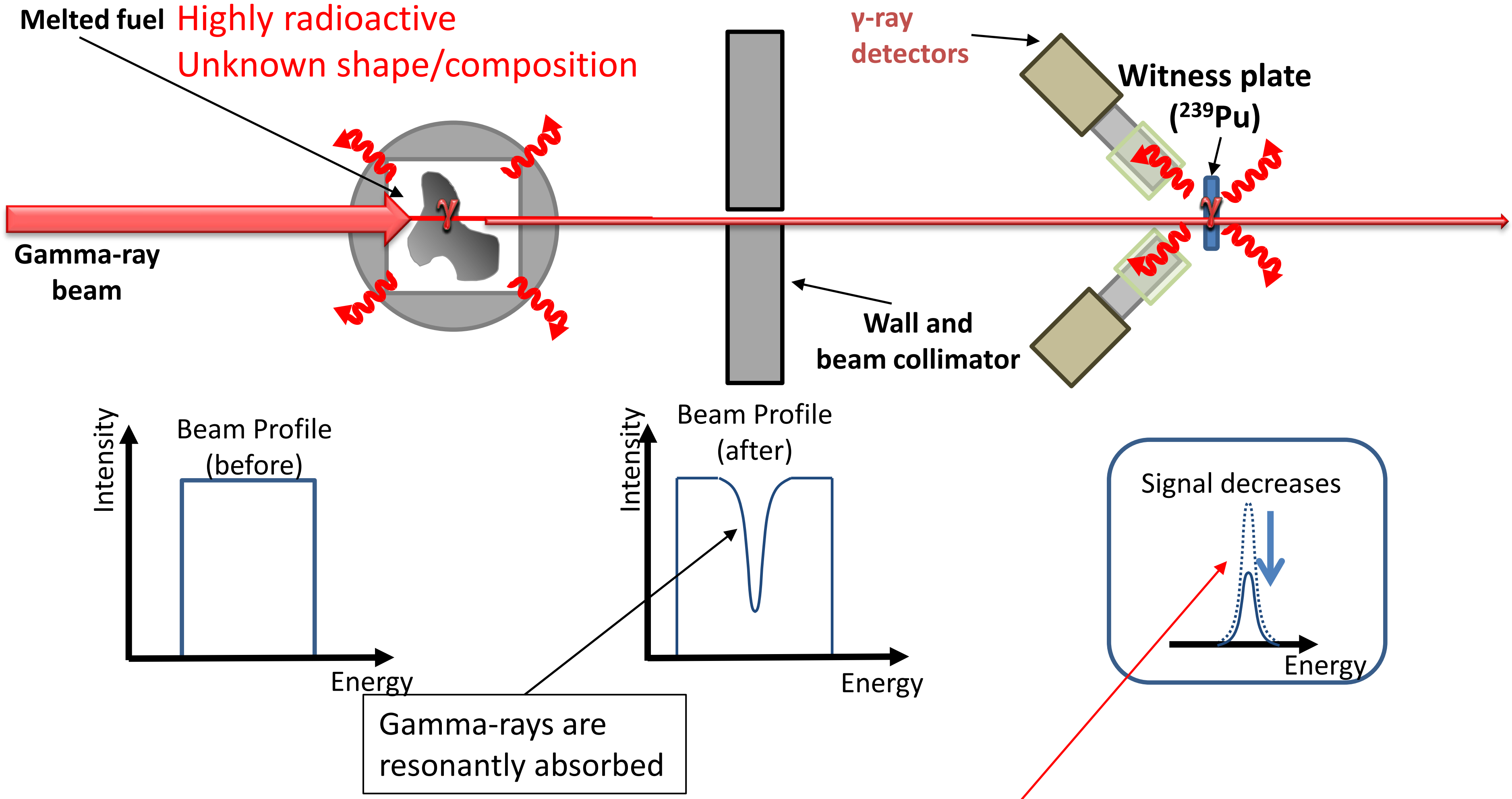
Cryo NRF

References:

C.T. Angell et al., “Temperature Effects on Non-destructive Assay using Integral Resonance Transmission”
Annual Meeting of Atomic Energy Society of Japan, 2013.

C.T. Angell, “Improving Efficacy and Enabling in situ Thermometry Using Cryogenic Transmission Nuclear Resonance Fluorescence”, Submitted to NIM-B

Transmission NRF (Single Resonance)



Melted fuel Highly radioactive

Unknown shape/composition

γ-ray detectors

Witness plate (^{239}Pu)

Gamma-ray beam

Wall and beam collimator

Beam Profile (before)

Beam Profile (after)

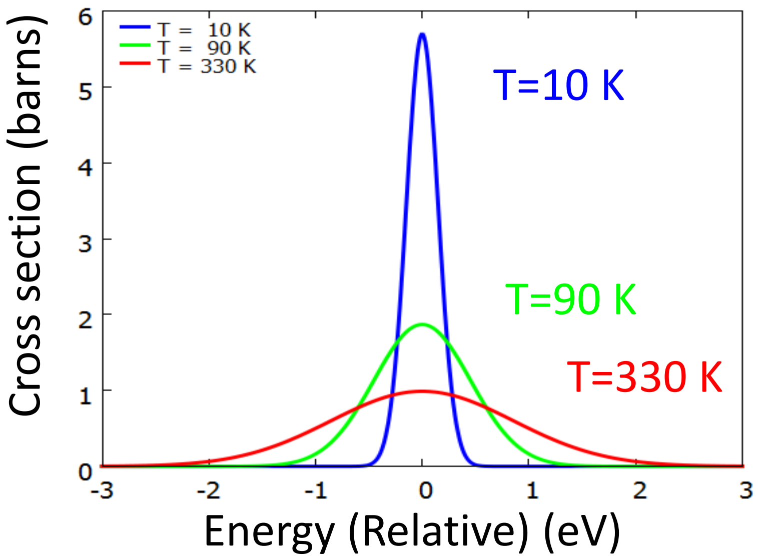
Signal decreases

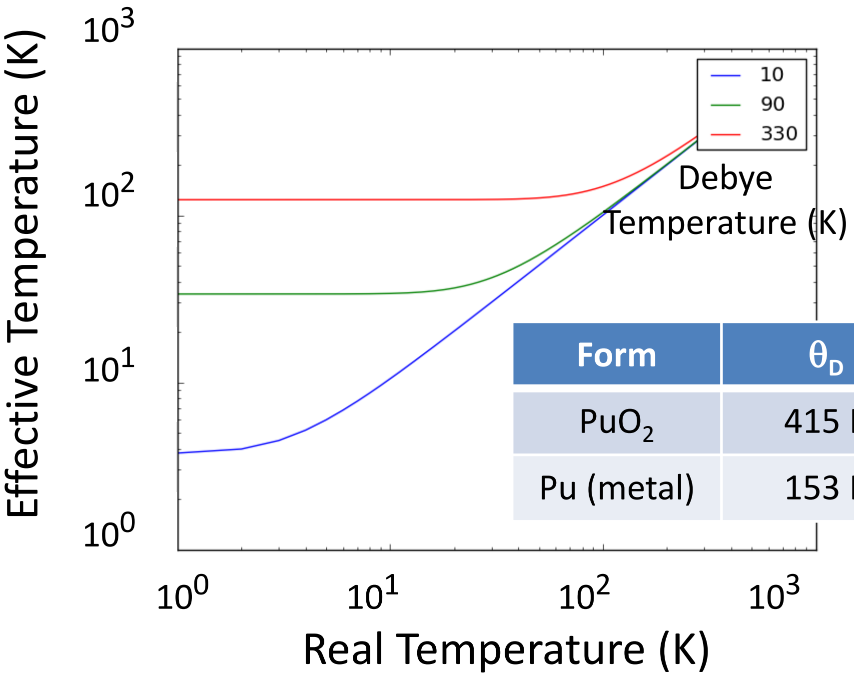
Gamma-rays are resonantly absorbed

Decrease in signal rate proportional to upstream ^{239}Pu mass.

Effect of Temperature on Resonance Width – General

- Lower temperature reduces the resonance width
  - Doesn’t change area
- Debye Temperature, θ_D, – “temperature” of lattice vibrations
- Asymptotic limit to how much you can narrow a resonance
- Debye Temperature depends on chemical form of material

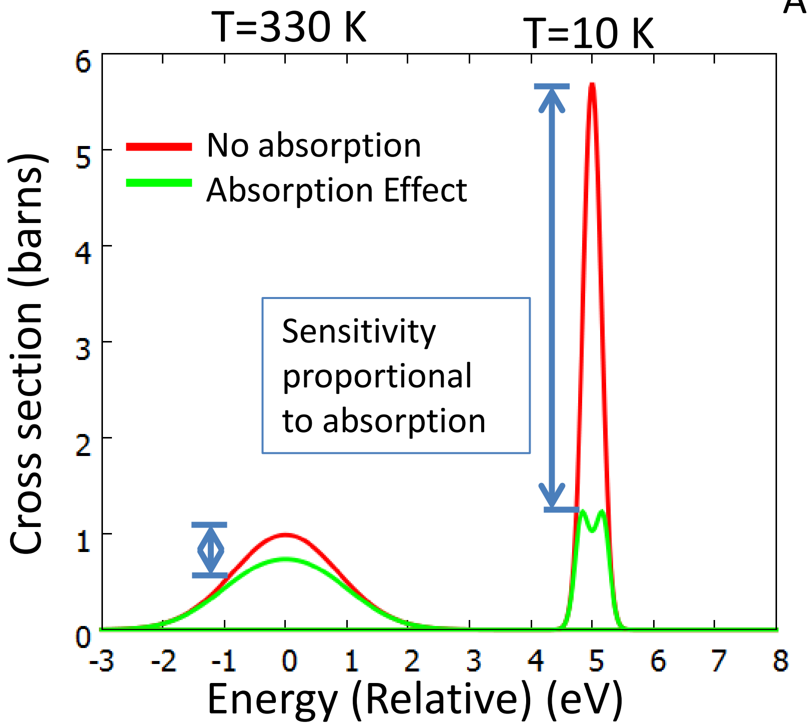




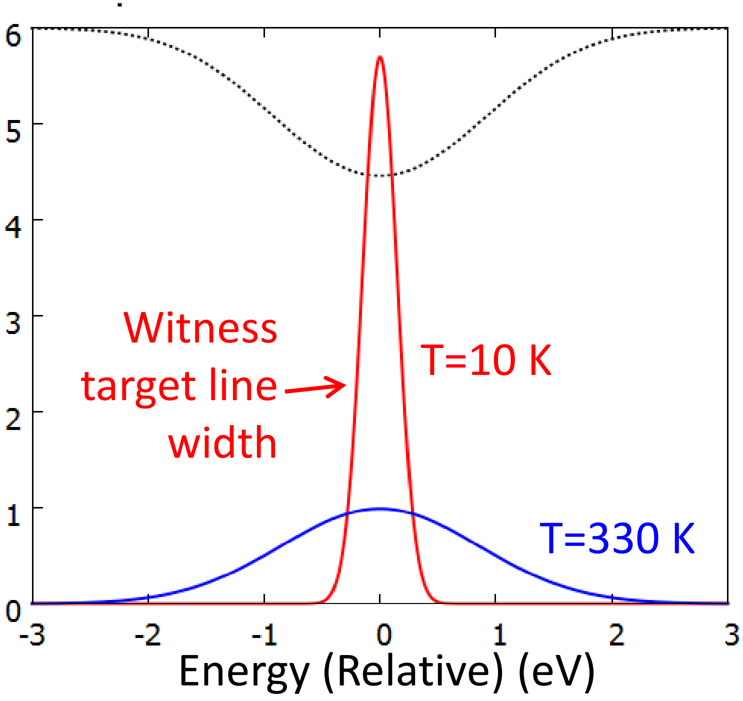
Form	θ_D
PuO_2	415 K
Pu (metal)	153 K

Effect of Temperature on Absorption Measurement

- For absorption must consider integral of absorbed flux profile X line shape.



Attenuated Flux from T=330 K Resonance in melted fuel



Reducing only the Witness target temperature can improve sensitivity

Cryo-NRF applications

- Reduction of measurement time in NRF-NDA
  - 40% reduction with a witness target at 77 K
  - 80% reduction with absorption and witness targets at 77 K
- In situ thermometry
  - Transmission NRF with two witness target temperatures, 77 K and 1000 K, for example
  - Unknown temperature of absorption target can be identified
  - Possible applications: internal combustion engine, chemical reactor, plasma ions ….

Integral Resonance Transmission Method

References:

C.T. Angell et al.,
"Demonstrating the Integral Resonance Transmission Method: Conceptual and Experimental Studies",
Proc. Annual Meeting of INMM (2015).

C.T. Angell, R. Hajima and B.J. Quiter.
"Non-destructive assay of spent nuclear fuel using the integral resonance transmission technique"
Submitted to Annals of Nuclear Energy

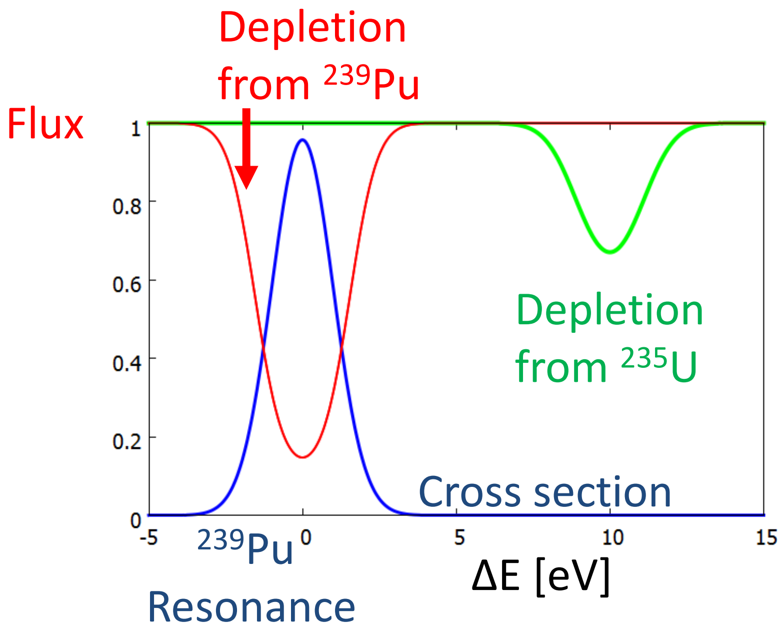
C.T. Angell et al.,
"Branching and fragmentation of dipole strength in 181Ta"
To be submitted

Integral resonance transmission (IRT)

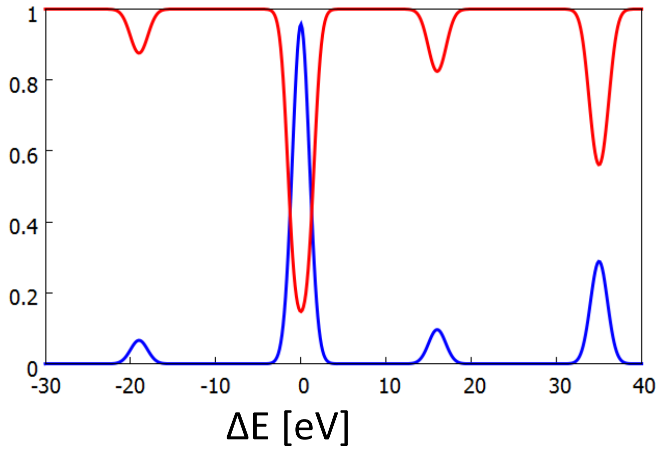
- Uses multiple resonances :

  Retains Isotopic Sensitivity

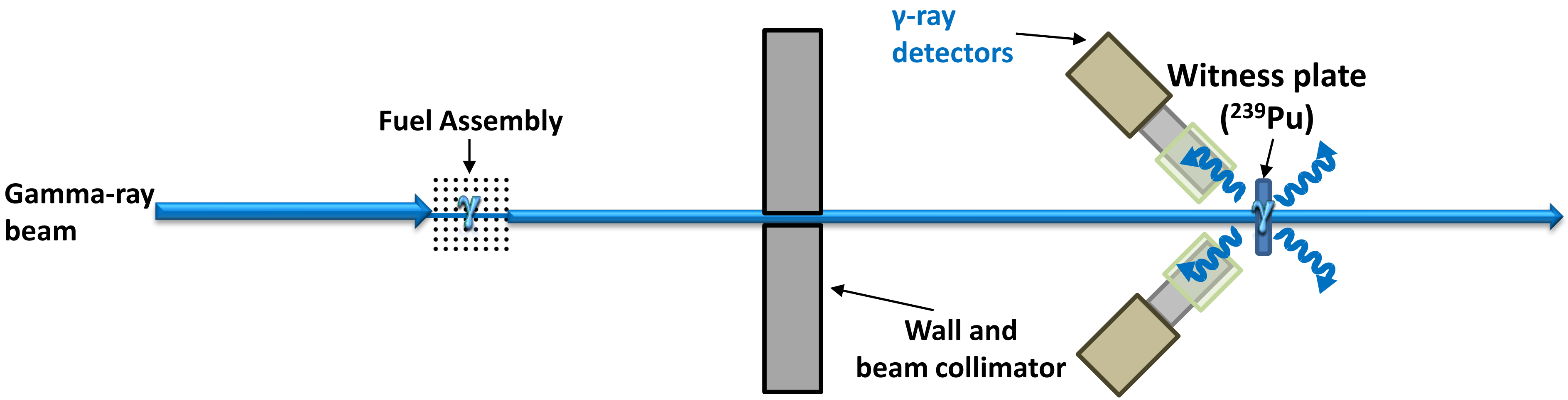
  - Each resonance carries information on absorption (i.e. mass)
- Requires monoenergetic γ-ray beam
  - Removes unrelated beam background from down scattering related with broad spectrum γ -ray sources
- Enables the use of scintillator detectors
  - Overcomes final problem of count rate at witness station
  - Count rate limit 2 orders of magnitude higher than HPGe



Depletion from multiple resonances in ^{239}Pu



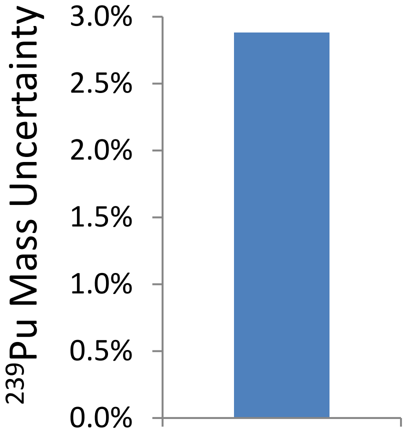
Performance estimate



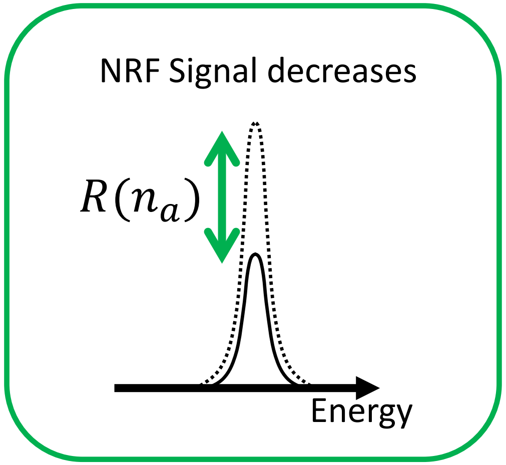
Assumption	
Measurement Time	8 hours
Areal density -BWR	64 g/cm^2
Flux	10^7 γ/(s eV)
^{239}Pu [% mass]	1%
^{238}U [% mass]	99%
Witness Plate	1 cm
^{239}Pu I_{cs}	13.5 eV b

Beam parameters are from estimated performance of the ERL.

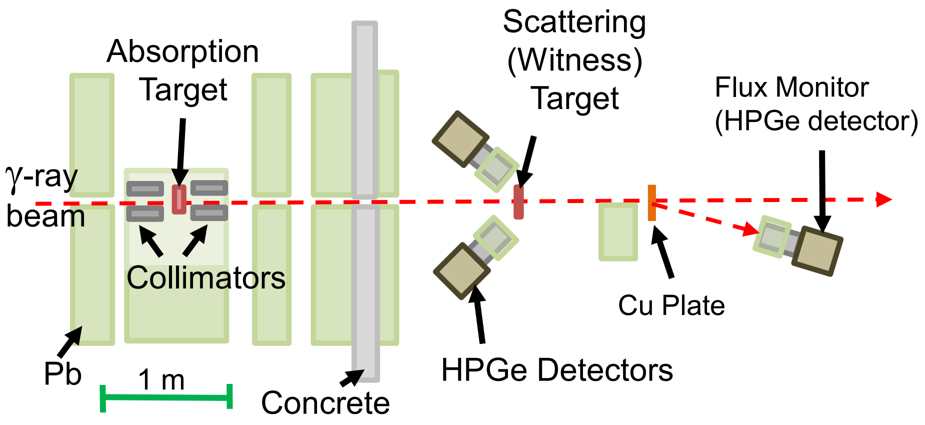
Results

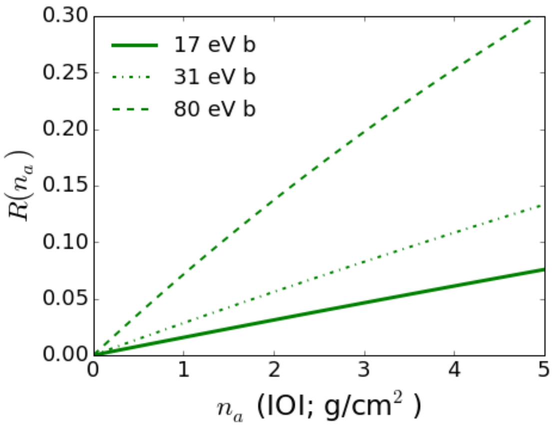


Transmission NRF experiment overview



- Transmission NRF measurements done to
  - A. Validate IRT technique (^{181}Ta, ^{239}Pu)
  - B. Demonstrate assay feasibility for a TMI-2 canister (^{27}Al)
- All transmission NRF measurements are similar
  - A. Two measurements: with and with/out absorption target
  - B. Decrease in NRF scattering from witness target proportional to absorbing isotope mass
- All measurements used an identical setup:



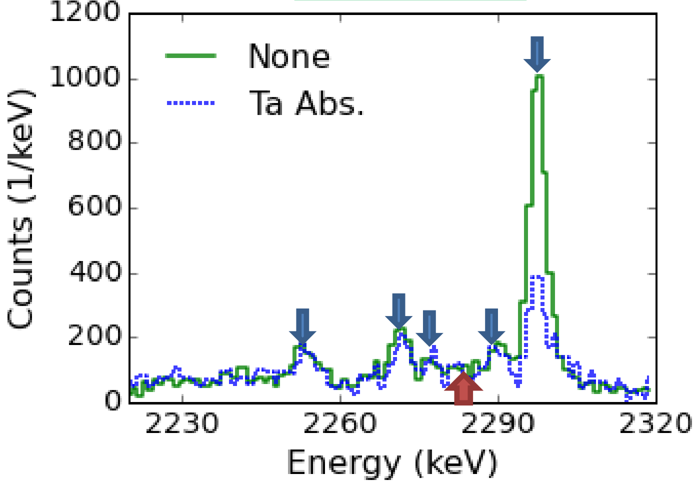


IRT technique demonstrated with ^{181}Ta

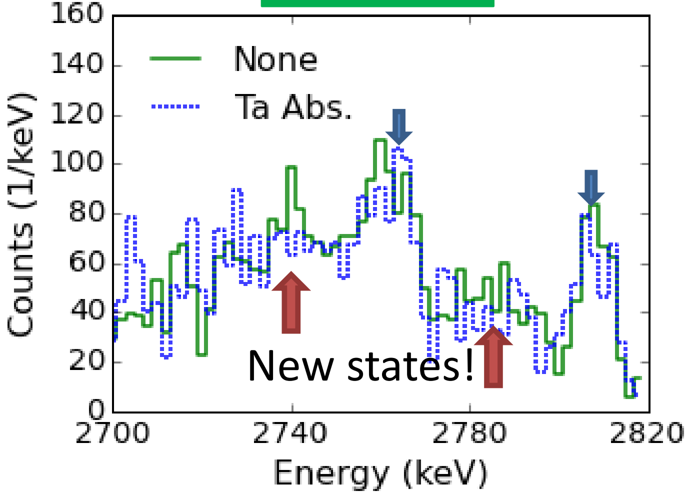
- 2.27 MeV: previously reported states all confirmed, and resonance absorption observed.
- 2.75 MeV: absorption from unresolved states clearly seen, demonstrating the IRT method.
  - All previously reported states confirmed and resonance absorption observed.

Energy (Mev)	Absorption Target	$R(n_a)$
2.27	Ta (2 cm)	0.3
2.75	Ta (3 cm)	0.1

2.27 MeV



2.75 MeV



Demonstration for TMI-2 containers

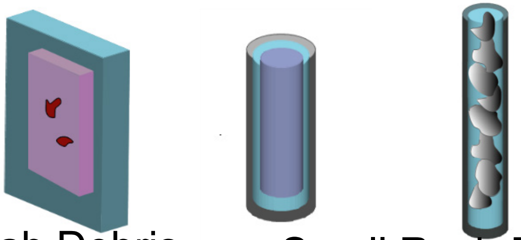
Reference:

C.T. Angell et al.,
“Demonstration of a transmission nuclear resonance fluorescence measurement for a realistic radioactive waste canister scenario”
NIM-B 347, 11 (2015)

LCS γ-ray for Fukushima

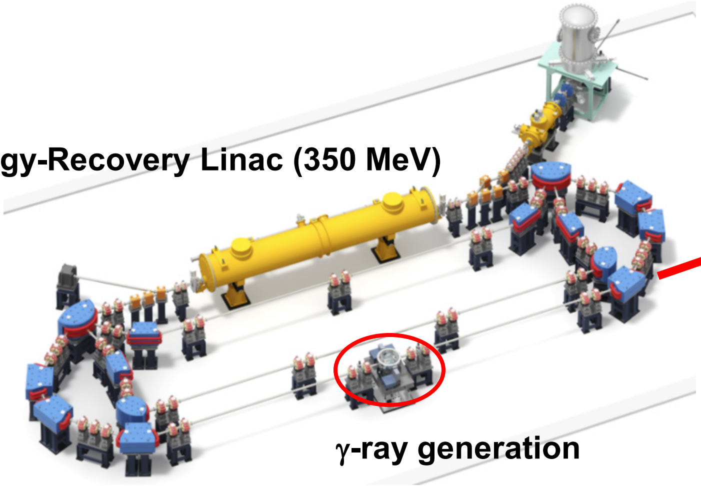
Measurement of Pu in the melted fuel → necessary for nuclear nonproliferation!

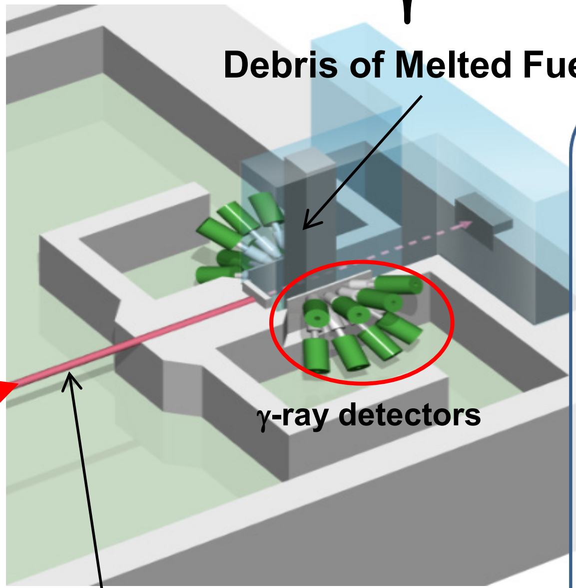
removal of debris
from the core ~2022



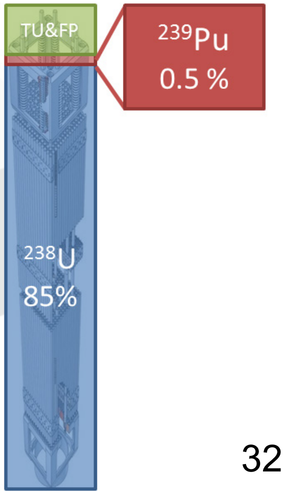
Debris of Melted Fuel

Energy-Recovery Linac (350 MeV)





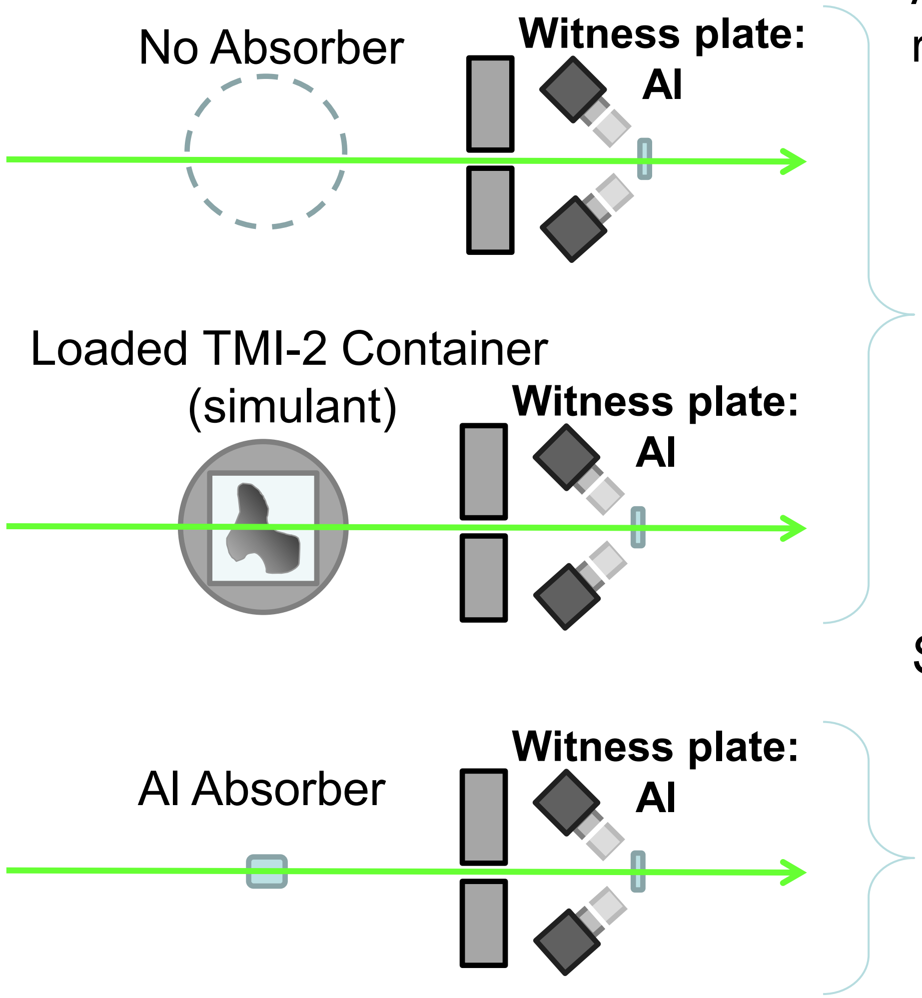
^{239}Pu Small Fraction
of Spent Fuel



Demonstration for Debris in a TMI-2 container

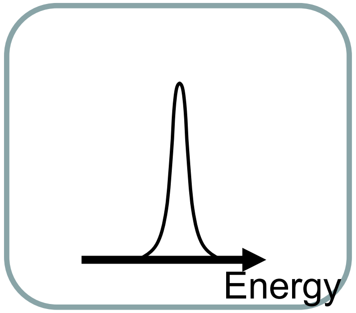
Experiment at Duke/HIγS
(LCS γ facility)

(TMI: Three Mile Island)



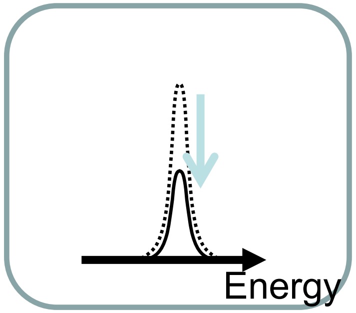
Al witness target chosen as it has strong resonance at similar energies to ^{239}Pu

No change expected!



Since witness target is Al, no absorption expected from simulant container

Signal decrease expected



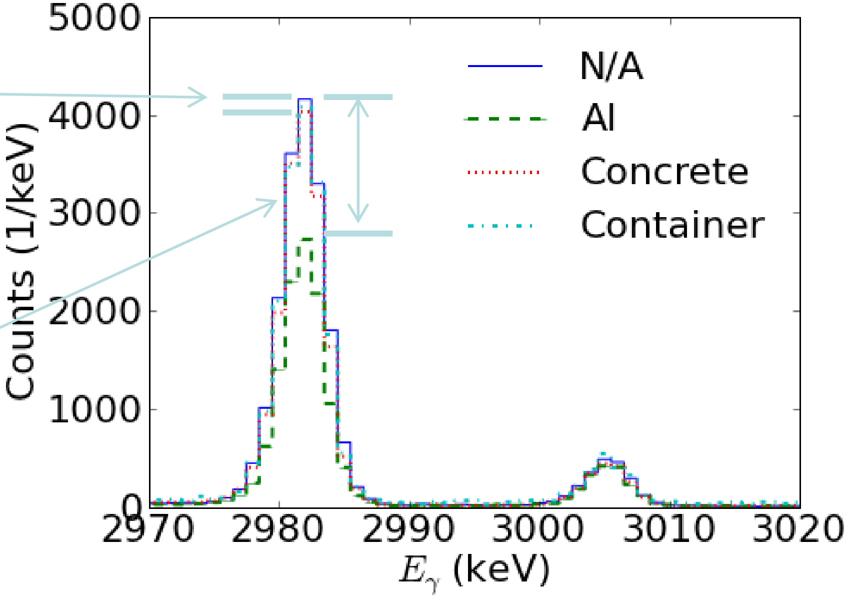
Using Al absorber can verify that experiment was done correctly.

Demonstration for Debris in a TMI-2 container

Verified NRF transmission feasible for TMI-2 container!

Small difference with concrete and container verified – concrete has small amount of Al

Large difference with Al absorber verified



NRF peaks normalized to flux

Absorber	Expected	Measured
Concrete	0.96±0.01	0.95±0.02
Container	0.96±0.01	0.97±0.03
Al	0.66±0.01	0.65±0.02

C.T. Angell et al., Nucl. Instr. Meth. B 347, 11 (2015)

Analytical study shows 3.7h – 22h measurement for ^{239}Pu in melted fuel with 3% accuracy by using a future ERL-LCS

C.T. Angell et al., to be published

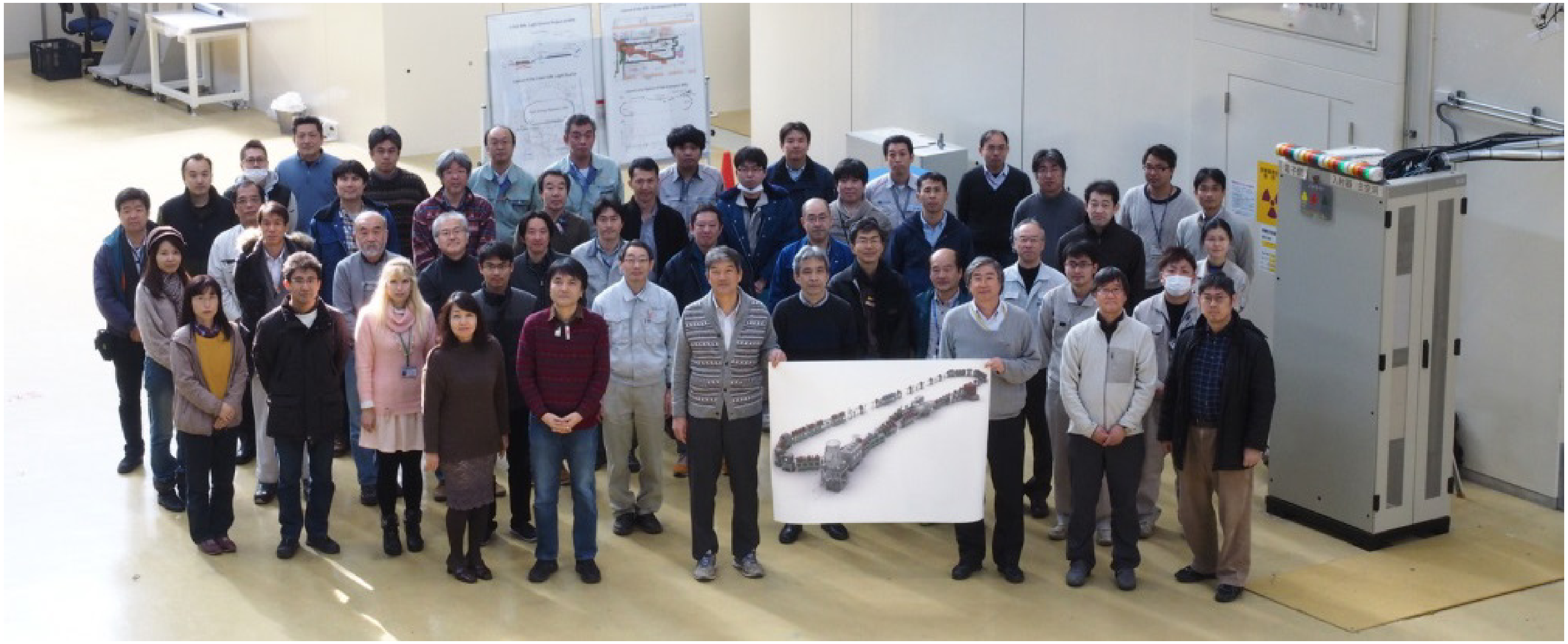
Experiment at Compact ERL

References:

T. Akagi et al., Proc. IPAC-2014, p.2072
A. Kosuge et al., Proc. IPAC-2015, TUPWA-66

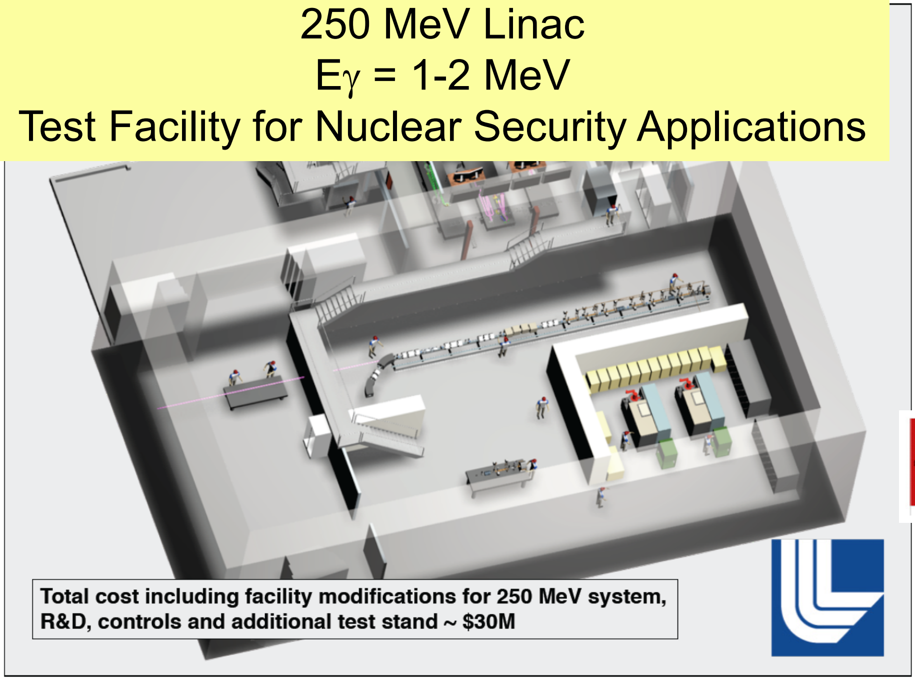
R. Nagai et al., Proc. IPAC-2015, TUPJE002

S. Sakanaka et al., Proc. IPAC-2015, TUBC1



New generation of LCS Gamma-ray Sources

T-REX @ Lawrence Livermore Natl. Lab.
250 MeV Linac
Eγ = 1-2 MeV
Test Facility for Nuclear Security Applications



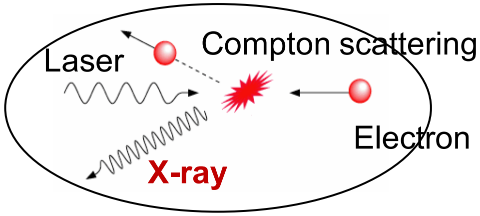
Total cost including facility modifications for 250 MeV system, R&D, controls and additional test stand ~ $30M

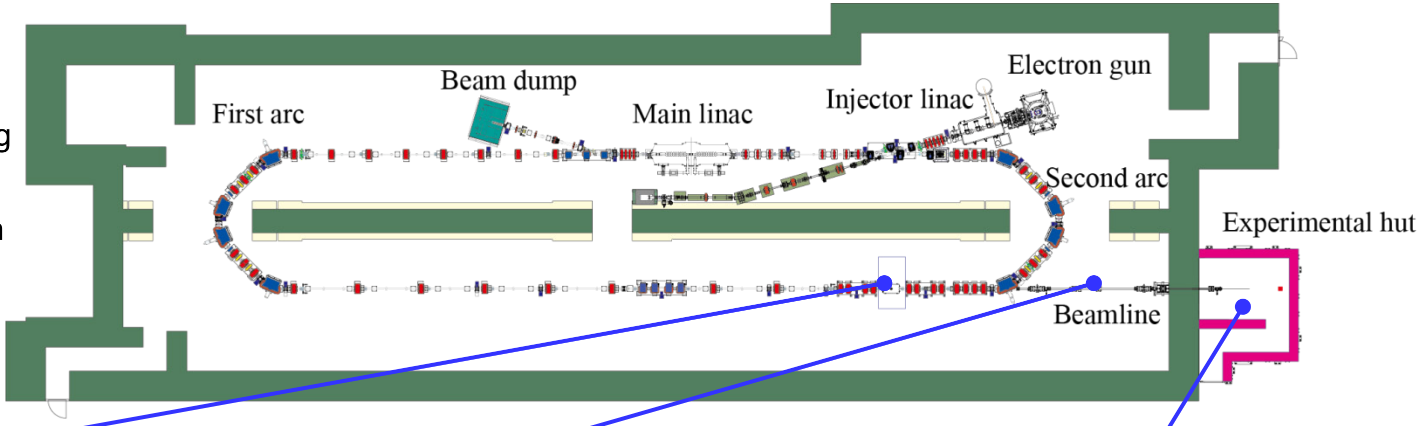
ELI-NP : Complex of PW lasers and LCS

ERL-based LCS gamma-ray @ KEK-JAEA
Test Facility for Nuclear Material Safeguards Applications

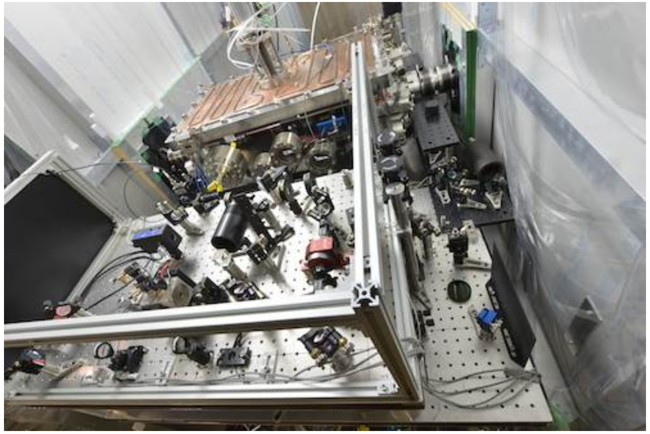
LCS Experiment at Compact ERL

Demonstration of technologies relevant to future ERL-based LCS sources





Laser enhancement cavity and 45W laser

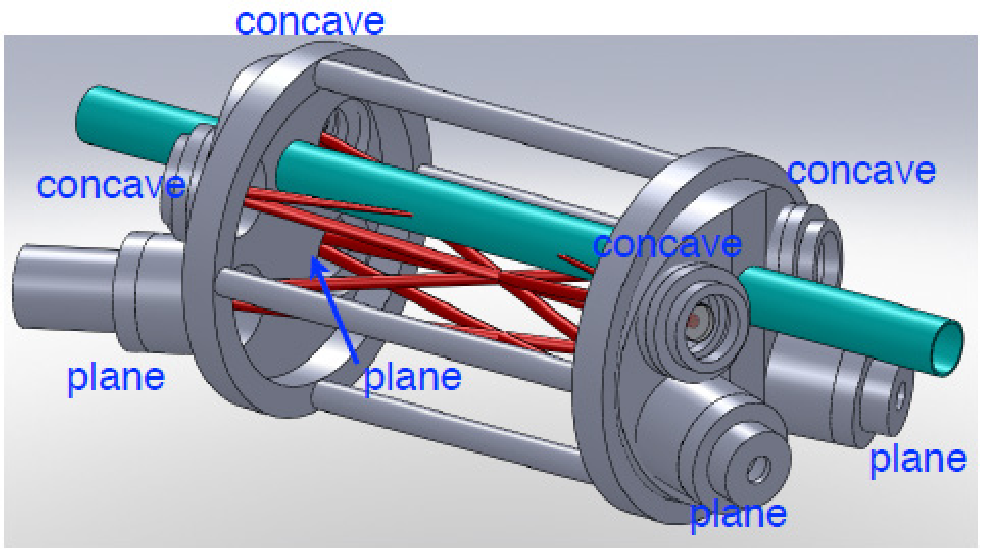


Beam line

Experimental hut

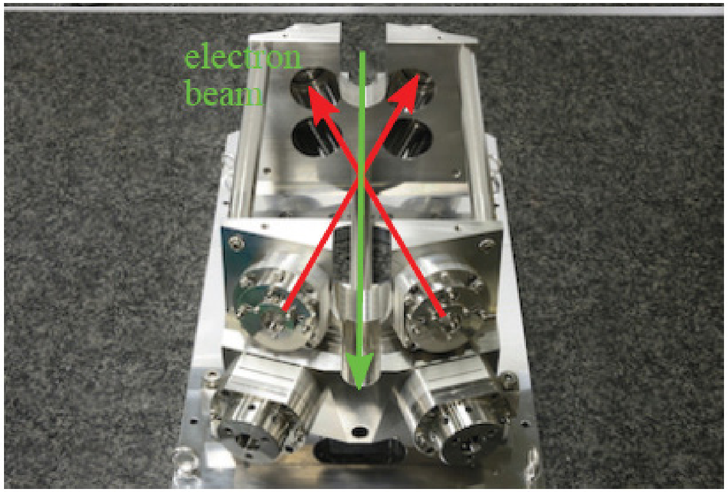
Work supported by:
A government (MEXT) subsidy for strengthening nuclear security (R. Hajima, JAEA)
Photon and Quantum Basic Research Coordinated Development Program from the MEXT (N. Terunuma, KEK)

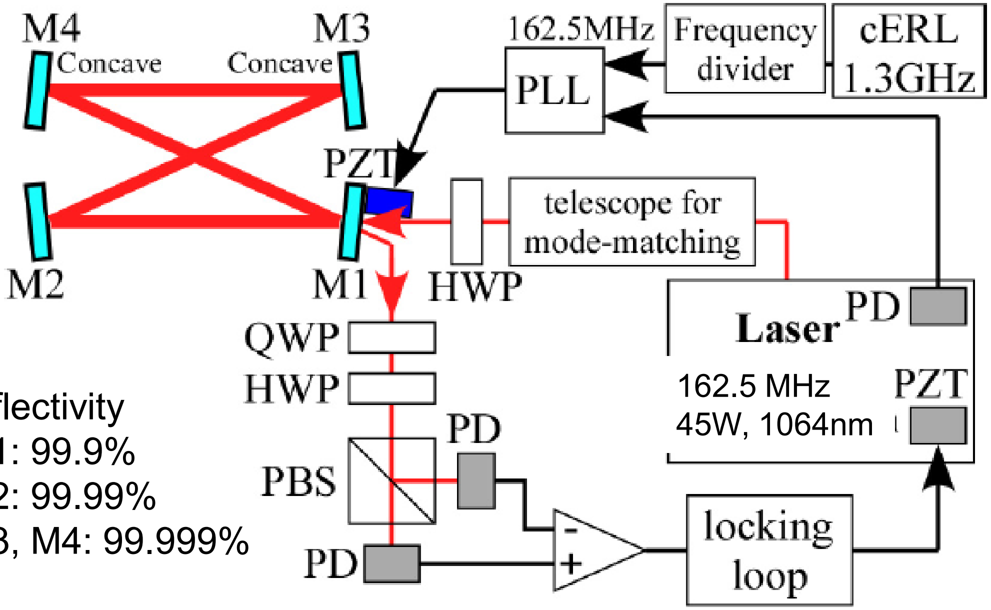
Laser Enhancement Cavity



Developed by T. Akagi (KEK)

T. Akagi et al., Proc. IPAC-2014, p.2072
A. Kosuge et al., Proc. IPAC-2015, TUPWA-66





reflectivity
M1: 99.9%
M2: 99.99%
M3, M4: 99.999%

Spot size: σ=30μm

Can store two beams independently

⇩

Fast polarization switch at 325 MHz
or
Double the laser power at LCS

(Single laser for the first experiment)

Beam Optics for the LCS

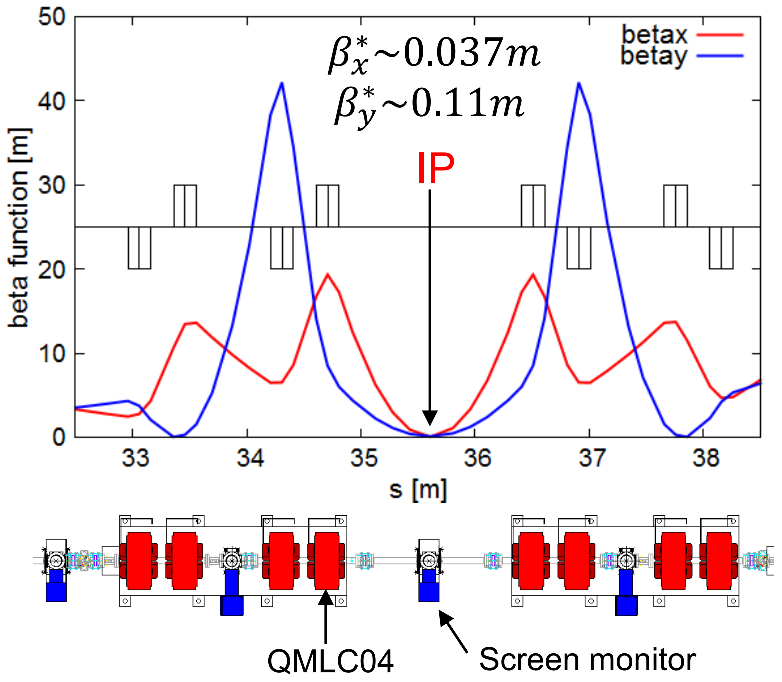
- Low-beta insertion for small beam sizes at IP
- Transport beams to the dump with small beam losses

Beam optics was established

IP: interaction point

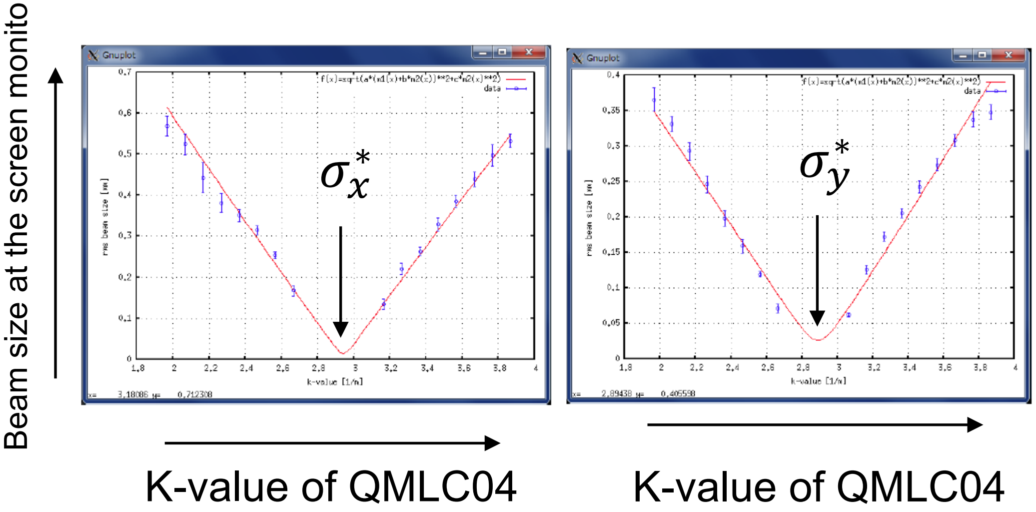
Design optics (example: “70% middle” optics)

$\sigma_x^* = 21\ \mu\text{m},\ \sigma_y^* = 33\ \mu\text{m}$ at IP



Beam sizes at IP were estimated from Q-scan data

$\sigma_x^* \sim 13\ \mu\text{m},\ \sigma_y^* \sim 25\ \mu\text{m}$ (example)



$\sigma_x^*, \sigma_y^* <$ (resolution of the screen monitor)

Bunch charge: 0.5 pC/bunch,
Normalized emittances: $(\varepsilon_{nx},\varepsilon_{ny})$=(0.47, 0.39) mm·mrad

S. Sakanaka et al., Proc. IPAC-2015, TUBC1

X-ray Produced by LCS

Parameters of electron beams:

Energy [MeV]	20
Bunch charge [pC]	0.36
Bunch length [ps, rms]	2
Spot size [μm, rms]	30
Emittance [mm mrad, rms]	0.4
Repetition Rate [MHz]	162.5
Beam current [μA]	58

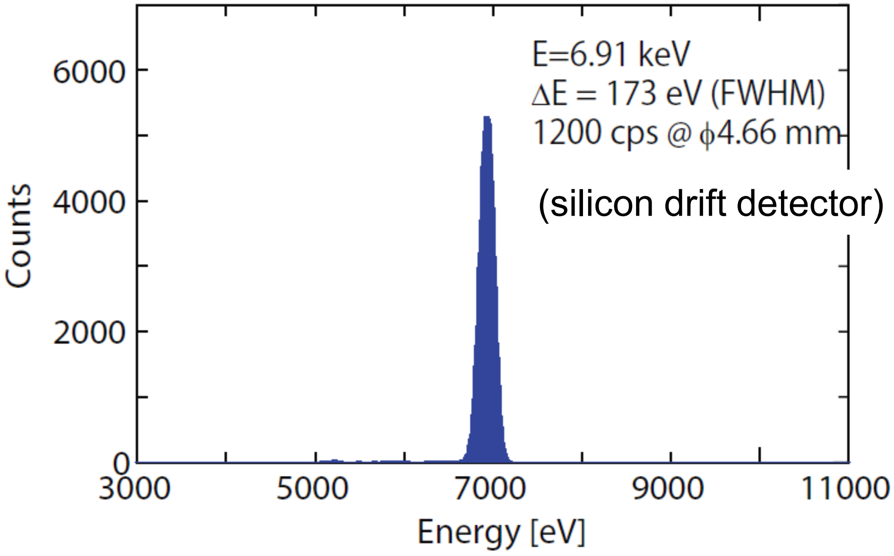
Parameters of laser (enhanced by cavity):

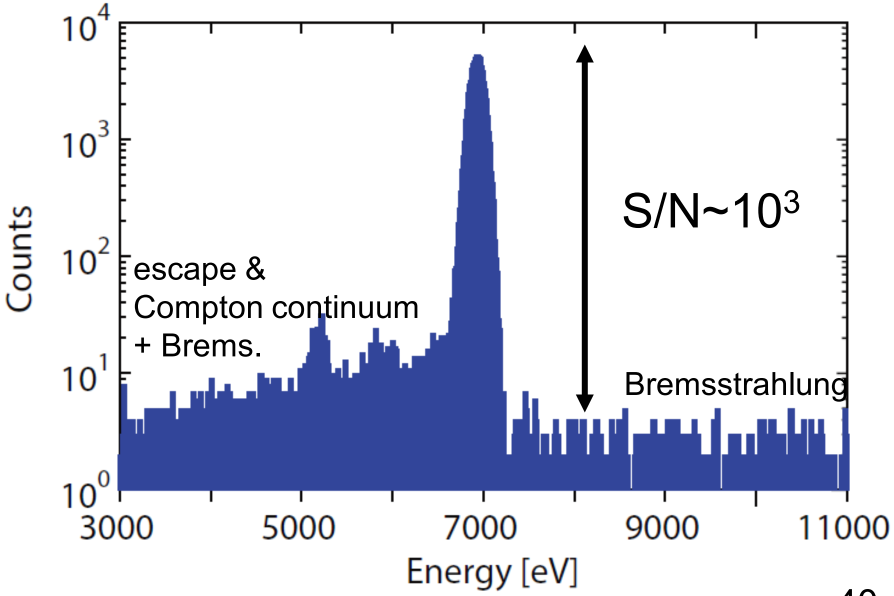
Center wavelength [nm]	1064
Pulse energy [μJ]	64
Pulse length [ps, rms]	5.65
Spot size [μm, rms]	30
Collision angle [deg]	18
Repetition rate [MHz]	162.5
Intracavity power [kW]	10

Results:

Photon energy = 6.9 keV
Detector count rate = 1200 cps @ϕ4.66mm (*)
Source flux = 4.3 x 10^7 ph/s (**)

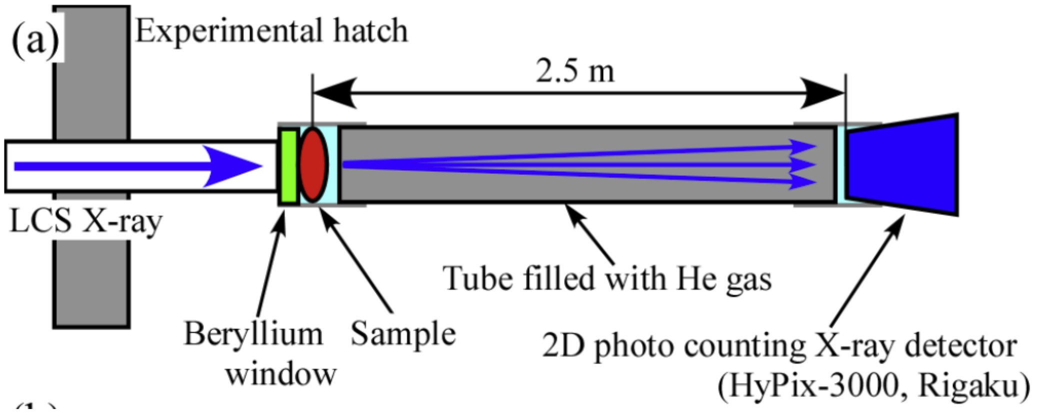
(*) Detector collecting angle is 4.66mm/16.6m = 0.281 mrad
(**) CAIN/EGS simulations with the detector count rate



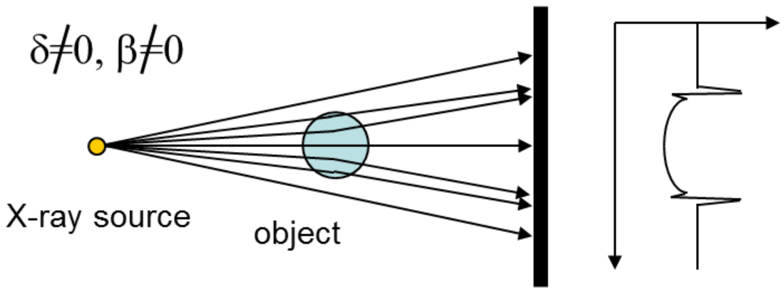


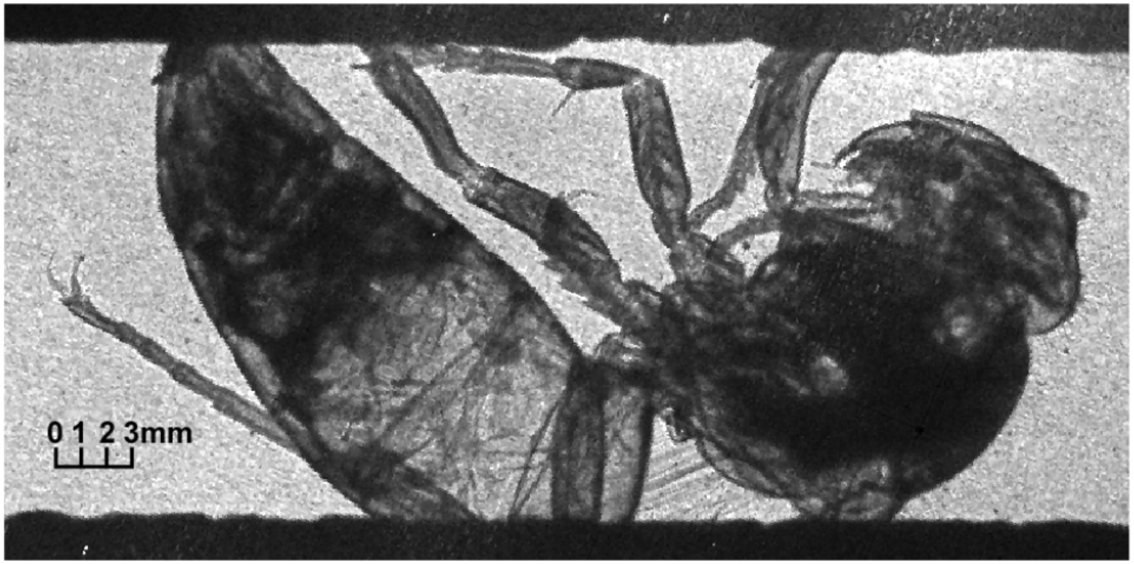
R. Nagai et al., Proc. IPAC-2015, TUPJE002

X-ray imaging with a LCS beam



phase contrast imaging

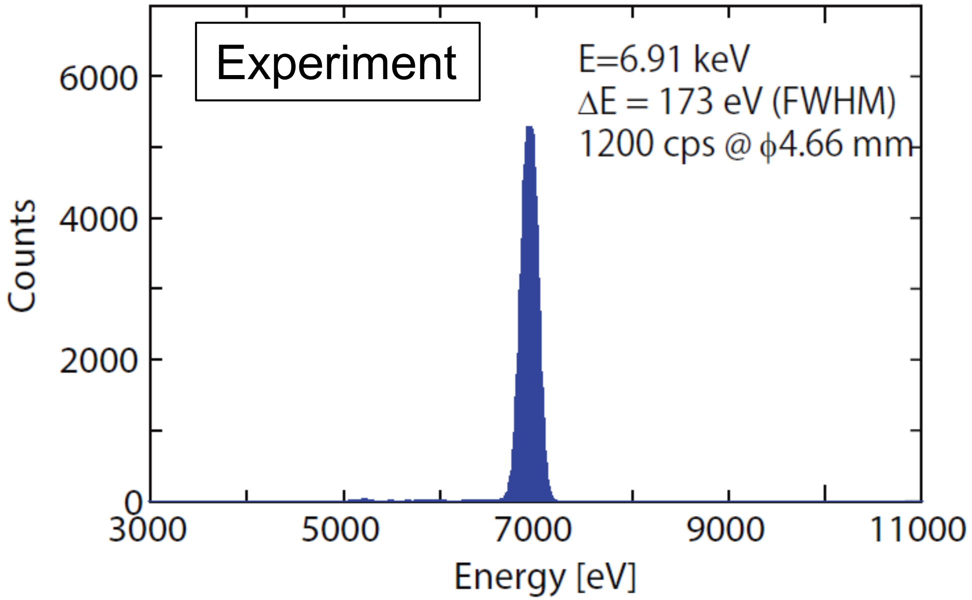


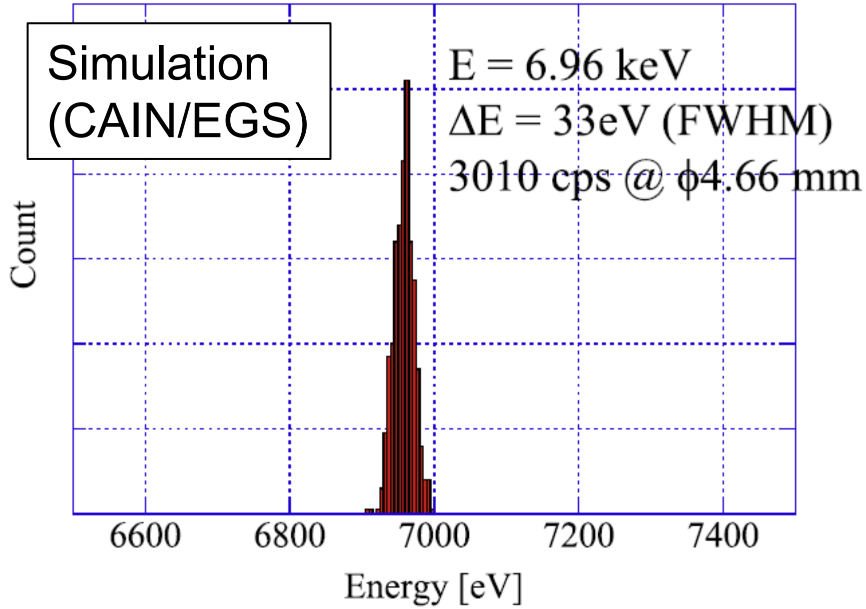


An X-ray image of a hornet taken with LCS-produced X-ray.
Detector: HyPix-3000 from RIGAKU. Detector was apart from the sample by approx. 2.5 m.

A. Kosuge et al., Proc. IPAC-2015, TUPWA066

Comparison with a simulation





Flux

consistent within a factor of 2.5

Bandwidth

detector resolution = 153eV@5.9keV (Fe-55)

Assuming quadratic nature for convolution of width, the energy width of the LCS photon beam is estimated to be

$$\sqrt{173^2 - 153^2} = 81\ \mathrm{eV}$$

We consider the detector resolution is not enough. We plant to make another experiment with a crystal monochromator.

Summary

- There are strong demands for nuclear security and safeguards technologies in the world
- NRF is one of the promising processes to realize non-destructive detection/measurement of nuclear material
- R&D’s on NRF-NDA have been carried out and still continue. We still have a large room to improve the system.
- High-flux gamma source is a key facility for
  - R&D’s of measurement methods
  - acquisition of missing nuclear data (minor actinides, especially)
  - exploring other NRF applications, in situ thermometry for example