

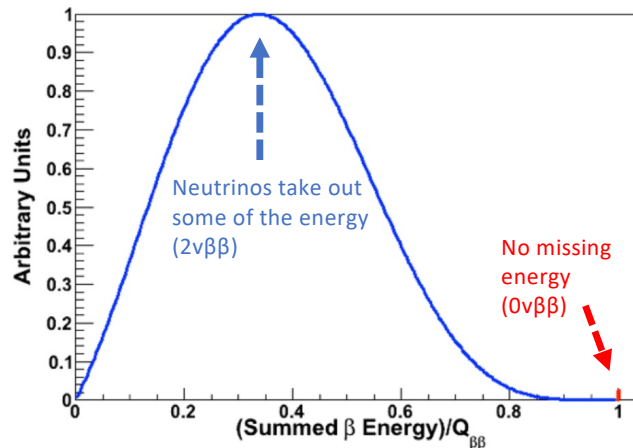
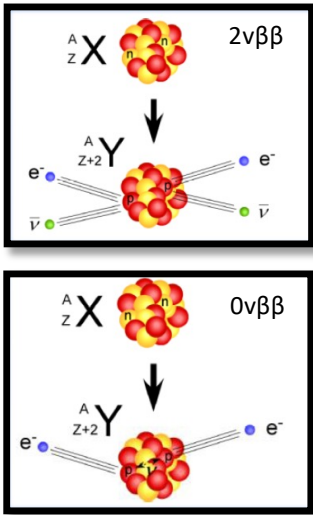


Laser isotope separation of ^{48}Ca for the CANDLES experiment

Kenji Mishima
RCNP, The Univ. of Osaka

mishima@rcnp.osaka-u.ac.jp

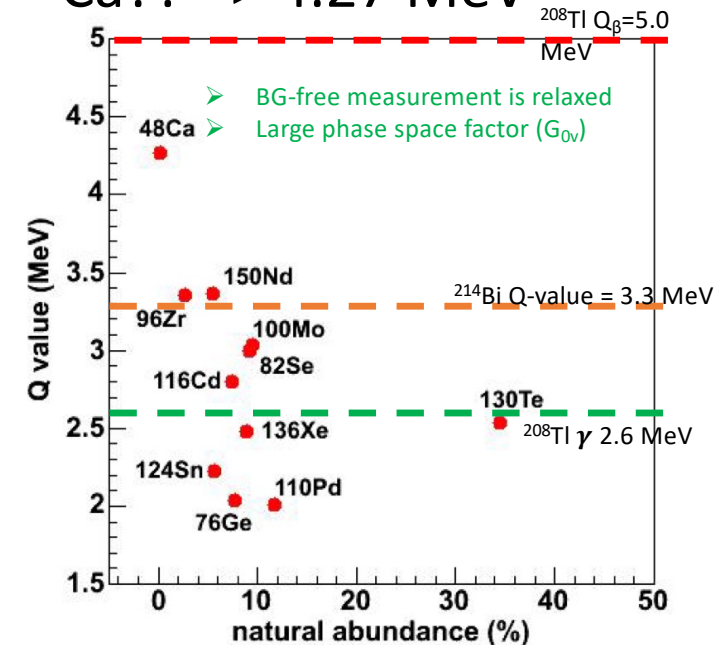
Calcium fluoride for studies of Neutrino and Dark matters by Low Energy Spectrometer (CANDLES)



Requirement for $0\nu\beta\beta$

- BG free measurement
- High energy resolution
- A large amount of double beta decay nuclide

Why $^{48}\text{Ca}?? \Rightarrow 4.27 \text{ MeV}$



$$\langle m_{\beta\beta} \rangle^2 \propto \frac{1}{T_{1/2}^{0\nu\beta\beta} G_{0\nu} |M_{0\nu}|^2}$$

$\langle m_{\beta\beta} \rangle$: Majorana neutrino mass
 $T_{1/2}^{0\nu\beta\beta}$: Half-life
 $G_{0\nu}$: Phase space factor
 $M_{0\nu}$: Nuclear matrix element

Majorana particle

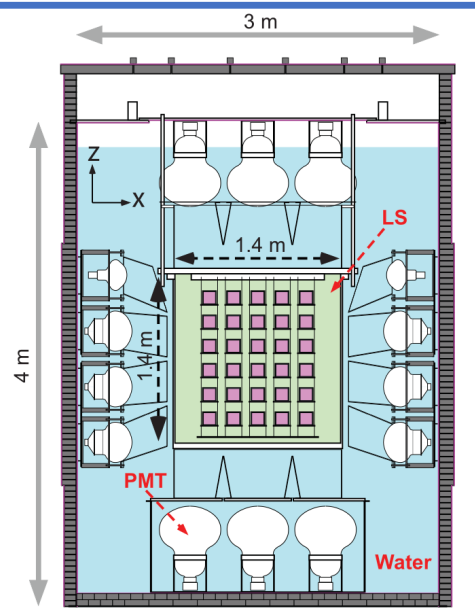
- Violates the Lepton number conservation law
- Particle $\leftrightarrow$ Anti-particle
- Matter dominated universe
- Absolute neutrino mass

<https://www-conf.slac.stanford.edu/ssi/2010/Hall080610.pdf>

Calcium fluoride for studies of Neutrino and Dark matters by Low Energy Spectrometer (CANDLES)



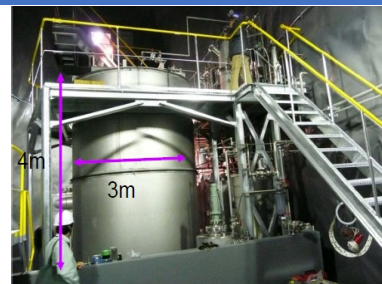
- Kamioka mine for zinc and lead
- Kamioka Lab for Underground Sciences
 - 1000 m.w.e. depth
 - 400 km away from Osaka



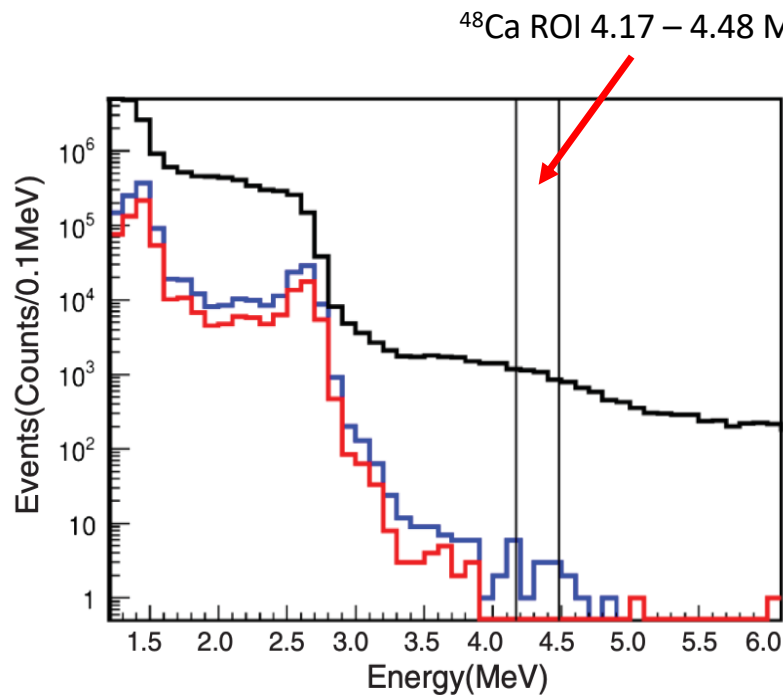
S. Ajimura, et al. *Physical Review D* 103.9 (2021): 092008.

- Liquid scintillator (LS)
 - 4π active shield(2 m³)
- 62 Large photomultiplier tube
- Shielding system
 - Pb : 10-12cm
 - B₄C sheet : 5mm

CANDLES III – 305 kg **CaF₂ Crystals** (3.2 kg × 96 pieces) ~ **0.35 kg ⁴⁸Ca**



Calcium fluoride for studies of Neutrino and Dark matters by Low Energy Spectrometer (CANDLES)

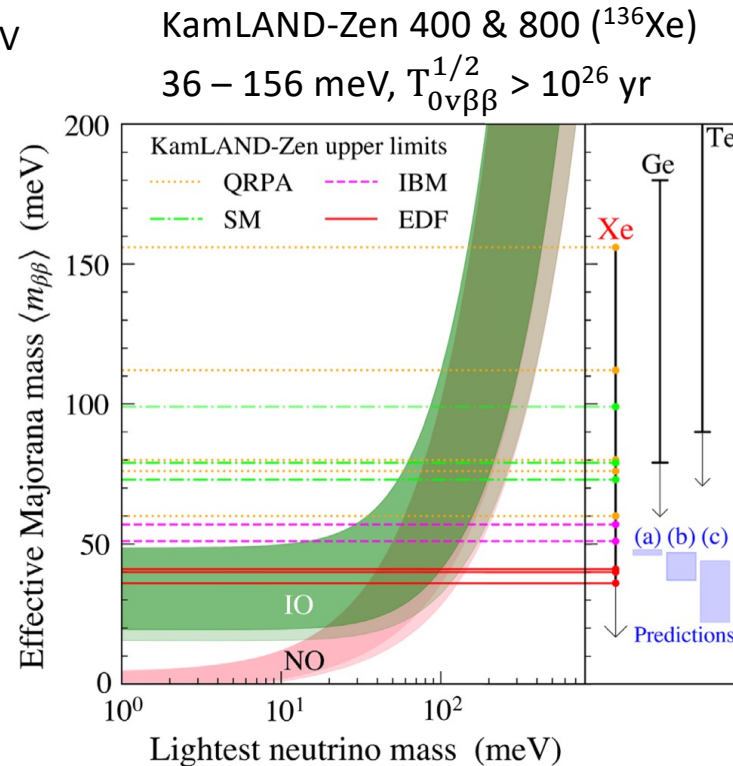


CANDLES III – $^{\text{nat}}\text{Ca}$

$$T_{0\nu\beta\beta}^{1/2} \text{ } ^{48}\text{Ca} = 5.6 \times 10^{22} \text{ years}$$

$$\langle m_{\beta\beta} \rangle \leq 2.9 - 16 \text{ eV}$$

S. Ajimura, et al. *Physical Review D* 103.9 (2021): 092008



Lightest neutrino mass (meV)

Phys. Rev. Lett. **130**, 051801

**Requires high-resolution measurements
of ^{48}CaF crystals with low impurity!**

To explore the IH region

Target: Inverse Hierachy (IH)
At $\langle m_{\beta\beta} \rangle = 0.03 \text{ eV}$

BG free measurement

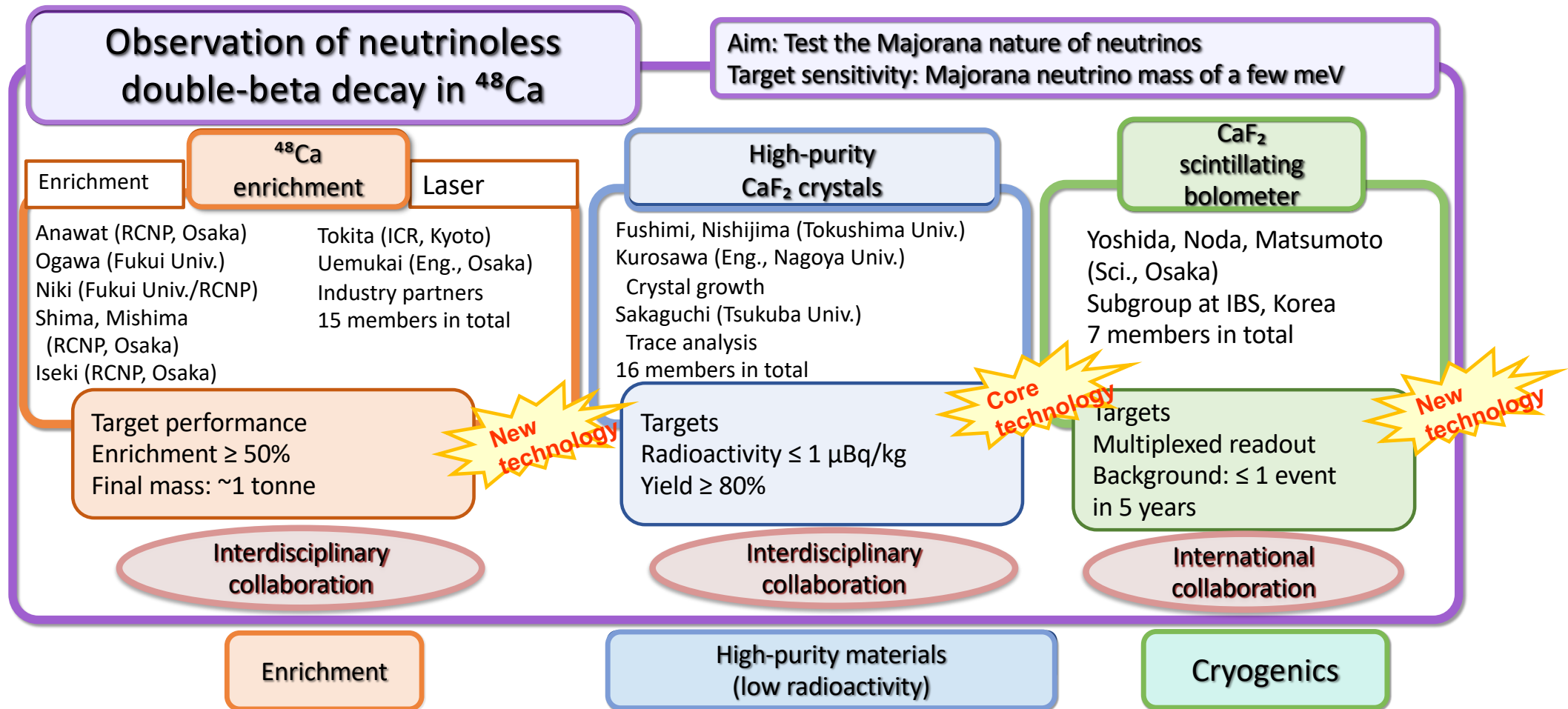
10 years of exposure time

0.8 Detection efficiency

**0.36 – 13 kmol of ^{48}Ca
~1800 × CANDLES III**

Next-generation double-beta decay detector

Developing a next-generation double-beta decay detector with emerging technologies



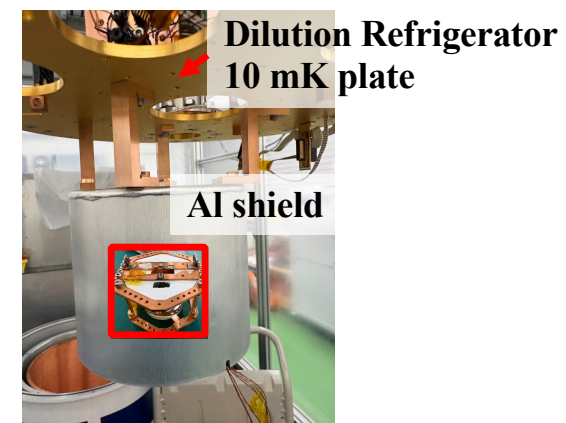
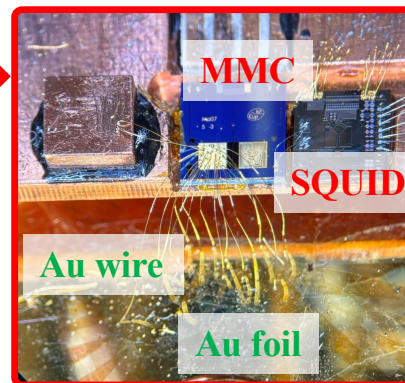
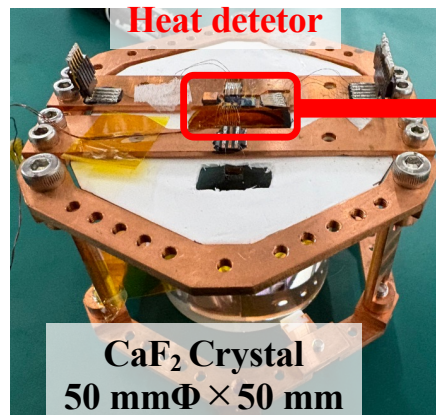
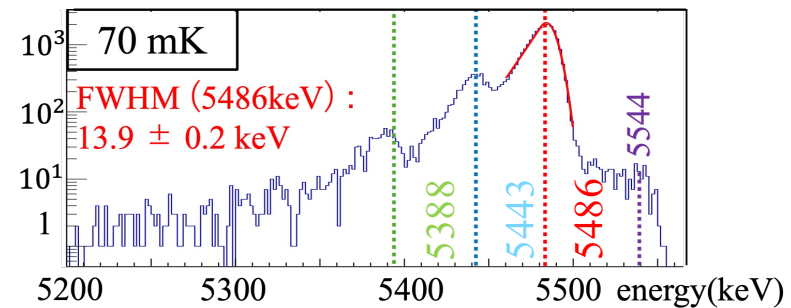
Development of the CaF₂ Scintillating Bolometer (Metallic Magnetic Calorimeter: MMC)

P9: K. Noda
Development of the CaF₂
Scintillating Calorimeter

Goal energy resolution (FWHM): 0.5% @4.27 MeV

Preliminary experiment conducted in Korea

- Sensor-only test setup without a crystal
- Measurement of ²⁴¹Am α particles
- Achieved FWHM 0.25% @5.5 MeV



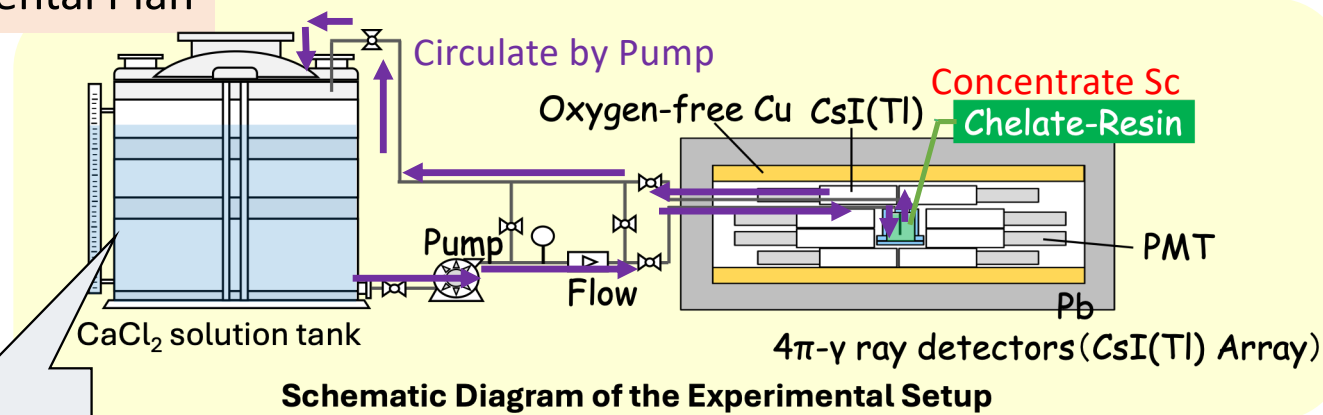
- Established sensor mounting, dilution-refrigerator operation, and the measurement environment for sensor control in Osaka

Next goal: detection of physical signals from the CaF₂ crystal

$^{48}\text{Ca} \rightarrow ^{48}\text{Sc}$ beta decay for $0\nu\beta\beta$

P6: N. Kinjo Evaluation of Chelate-Resin Adsorption Efficiency
for Collecting ^{48}Sc for ^{48}Ca Beta-Decay Half-life Measurement

Experimental Plan



630 L
260 kg CaCl_2

- ^{48}Ca decays into ^{48}Sc in the circulation path
- Use chelate resin that **selective adsorb Sc^{3+}** without Ca^{2+} and **Concentrate Sc** at the center of 30 CsI(Tl) detectors
- Measure 3γ coincidence from ^{48}Sc β -decay

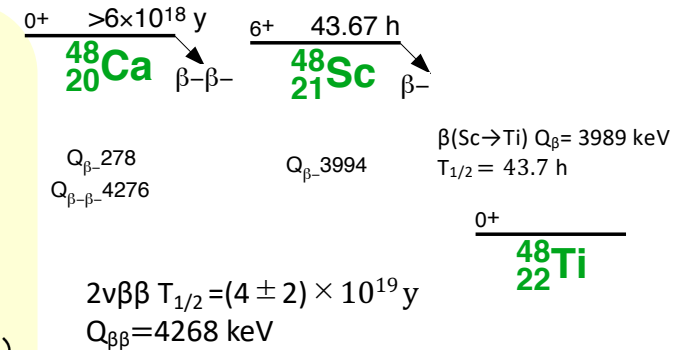
Advantages

- Large amount of Ca
- High-efficiency and low background (by 4π array and 3γ coincidence)

Current Status

- Fine-tuning the data acquisition system
- Prepare for experiments to evaluate the Sc adsorption efficiency of the chelate resin

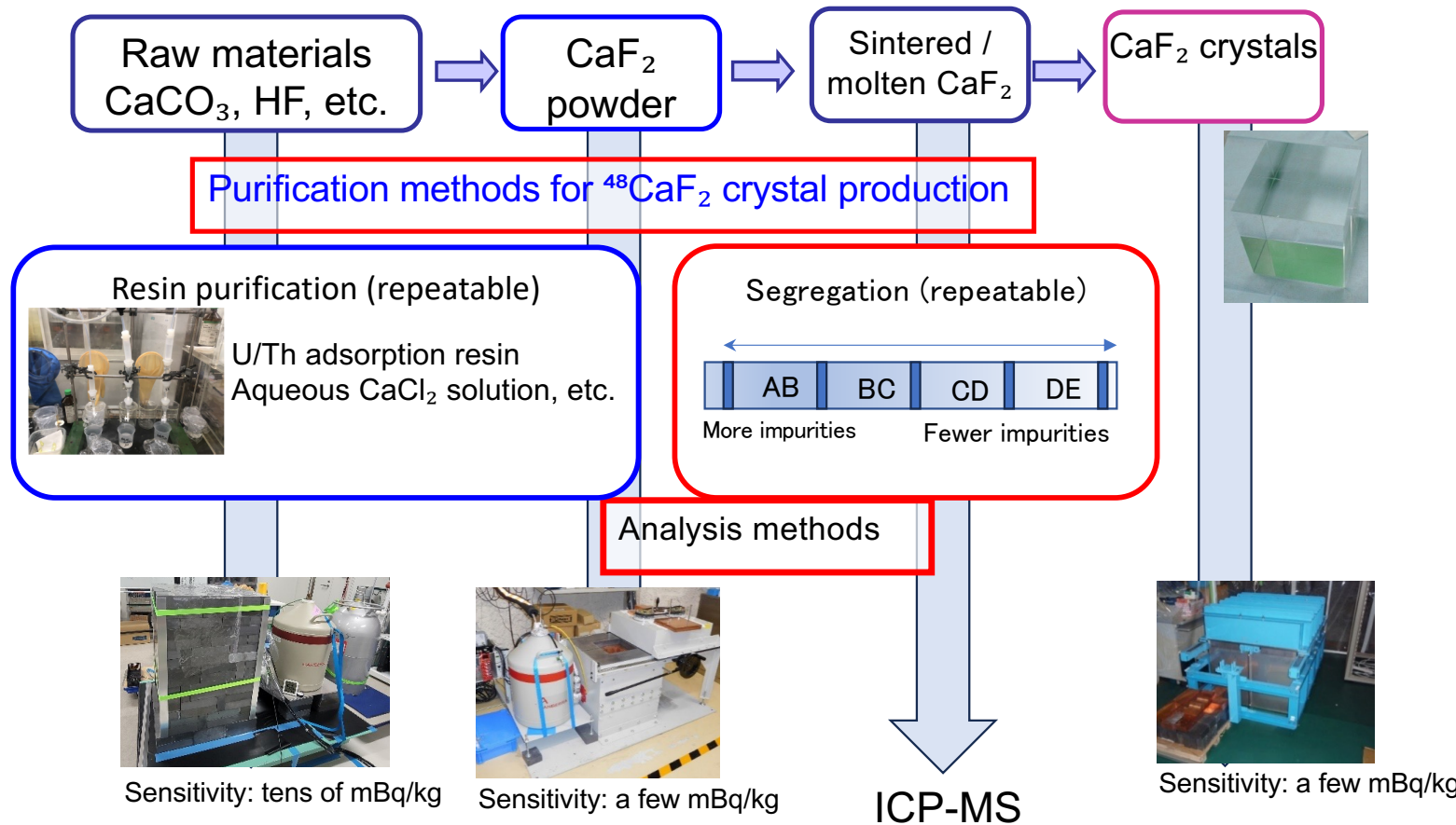
$\beta(\text{Ca} \rightarrow \text{Sc})$ $T_{1/2} = ??$
 $Q_{\beta} = 278 \text{ keV}$



Fabrication of high-purity CaF_2

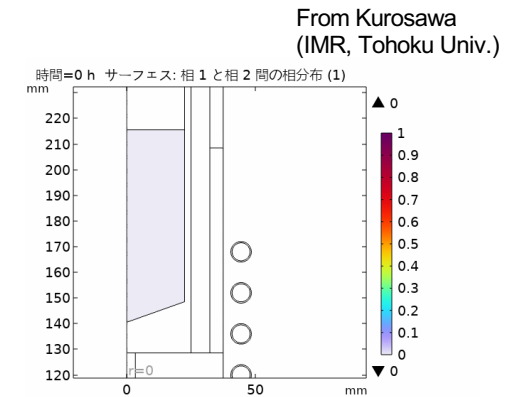
- Efficient production of high-purity CaF_2 crystals from ^{48}Ca

Requirement:
Purity $\mu\text{Bq/kg}$
Production yield $\sim 80\%$

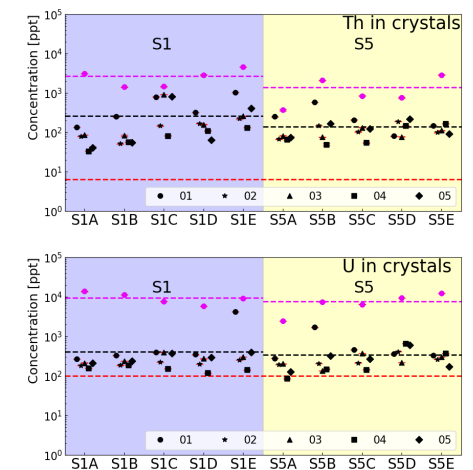


RCNP
Nishijima, Fushimi (Tokushima Univ.)
Kurosawa (Nagoya Univ.)
Sakaguchi, Takaku (Tsukuba Univ.), et al.

Crystal melting and solidification
(temperature simulation)



ICP-MS results

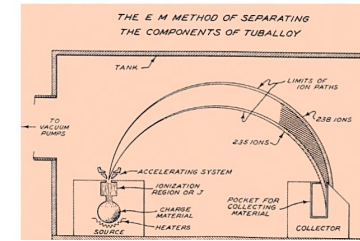


Nishijima (Tokushima Univ.) et al., JPS meeting Sep. 2026

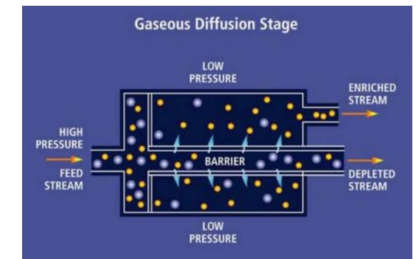
Enrichment of ^{48}Ca by Laser Isotope Separation (LIS)

Enrichment of ^{48}Ca [N.A. = 0.187%]

- **Ca has no gaseous compound.**
- ^{48}Ca 10 grams/year (By MS) $\rightarrow$ 200,000 – 1,000,000 \$/g

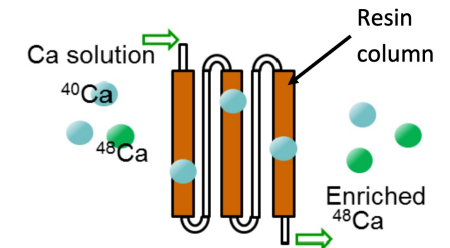


Electromagnetic

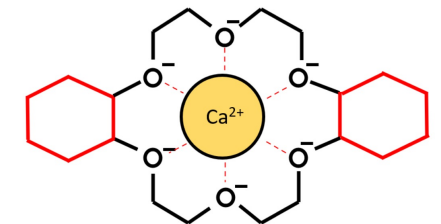


Gas diffusion

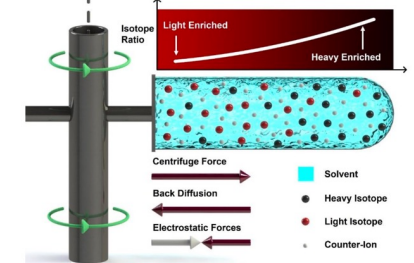
Isotope separation technique	Separation Coefficient	Production efficiency (y^{-1})	Cost	Limitation
Electromagnetic separator	High	Ten of grams	High	○ High power consumption ○ Low productivity
Liquid centrifuge	Moderate	Kilograms	Low	○ Concentration limit
Industrial isotope separation • Gas Diffusion • Gas Centrifuge	High	Thousands of tons	Low	○ Only the gas phase compound is possible ○ Compatible for U
Chemical isotope exchange	Small	Tons	Low	○ Extractant loss ○ Development of the cascade enrichment is required
Ion exchange chromatography	Small	Hundred of gram	Low	○ Time consumption ○ Low conversion
Laser isotope separation	High	Kilograms	Moderate	○ Development of the high-power laser, irradiation unit, and collection system



Chromatography



Solvent extraction

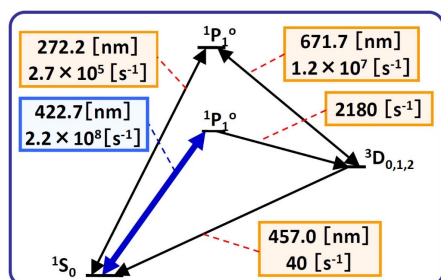


Liquid centrifuge 10

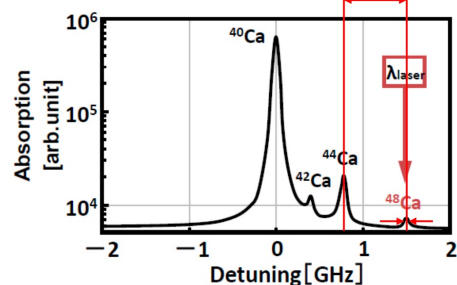
The experimental apparatus

RI Center, The University of Osaka, Toyonaka, Osaka, Japan

Oscillation wavelength

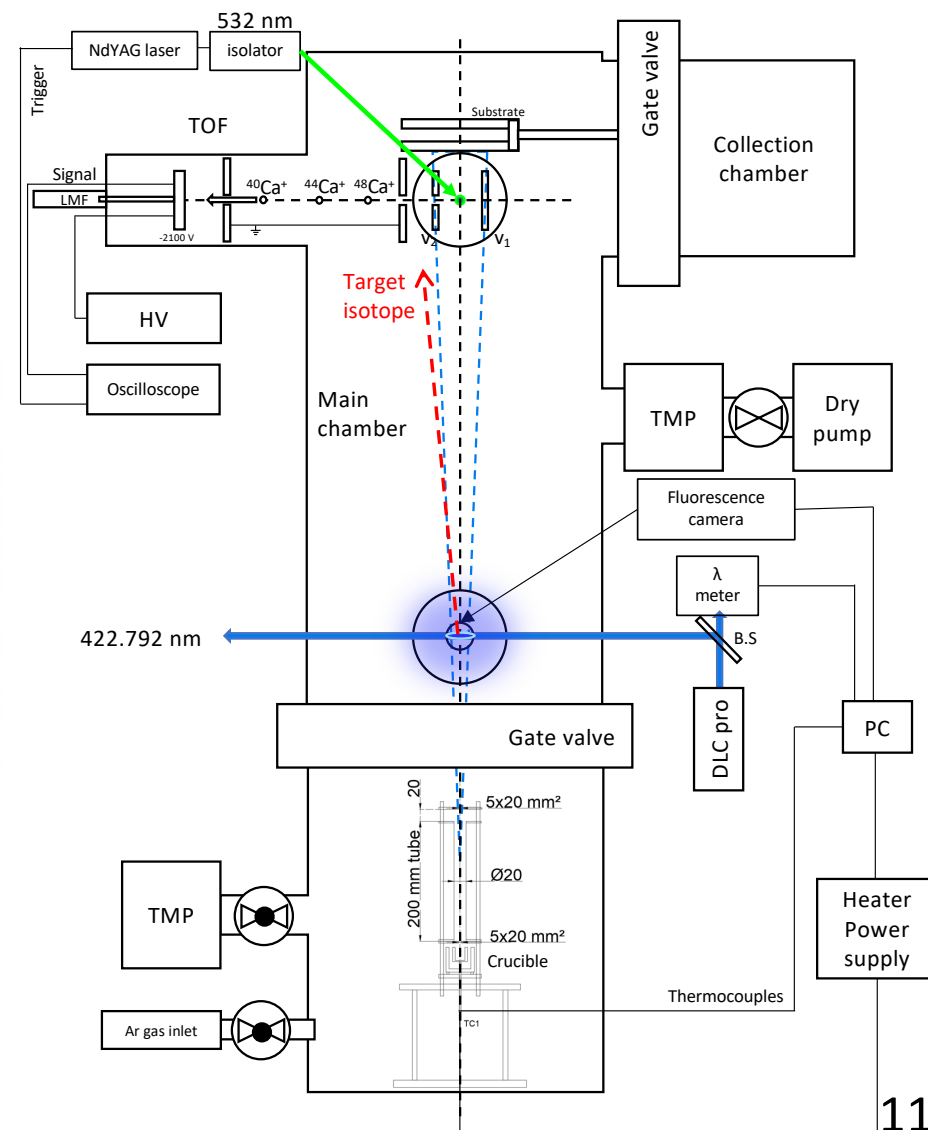
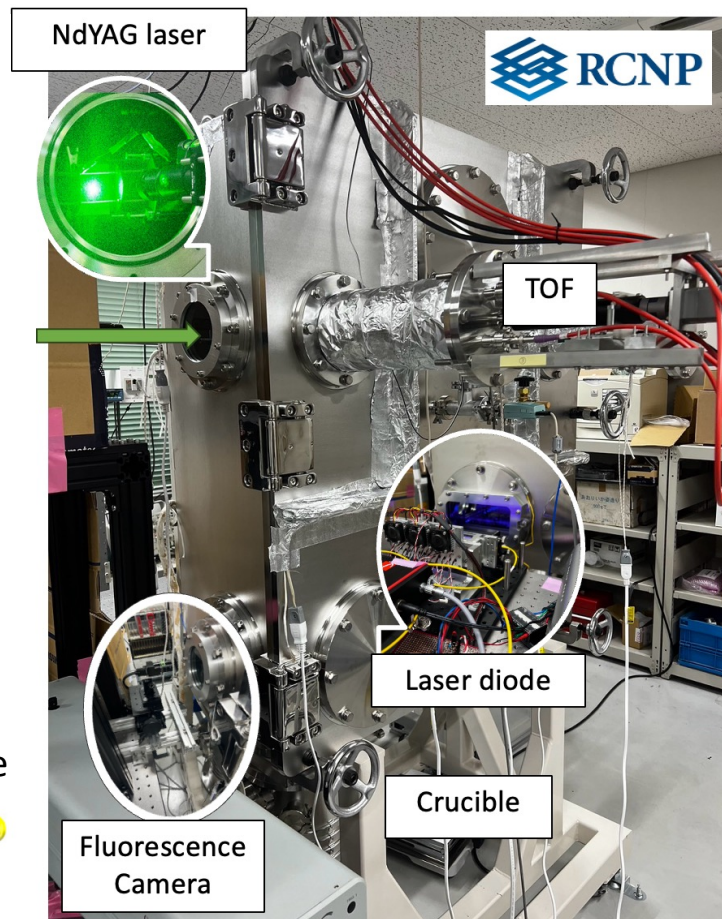
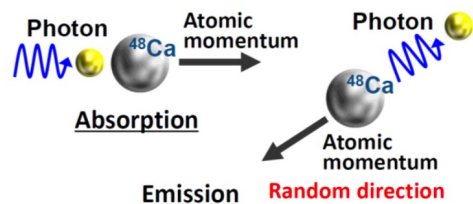


Allowed Transition Isotope shift: ~800 MHz



Absorption spectrum of Ca at 423 nm

Deflection of target isotope

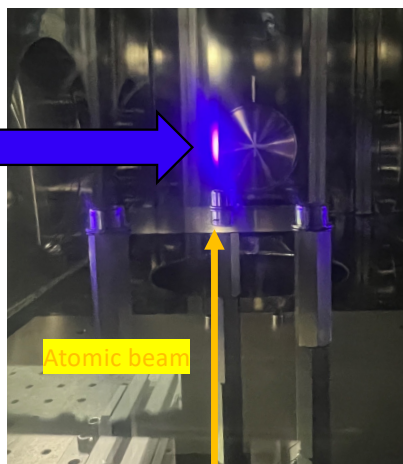


Recap on the last report (2025)

Fluorescent emission of ^{40}Ca

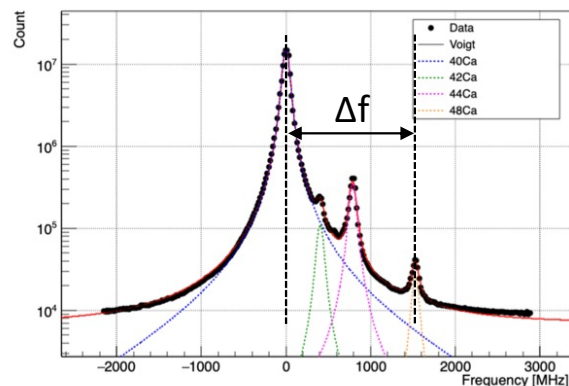
422.7918 nm

LD



Atomic beam

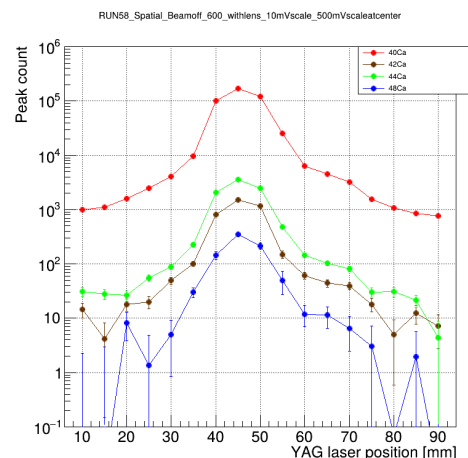
Laser-induced fluorescence measurement



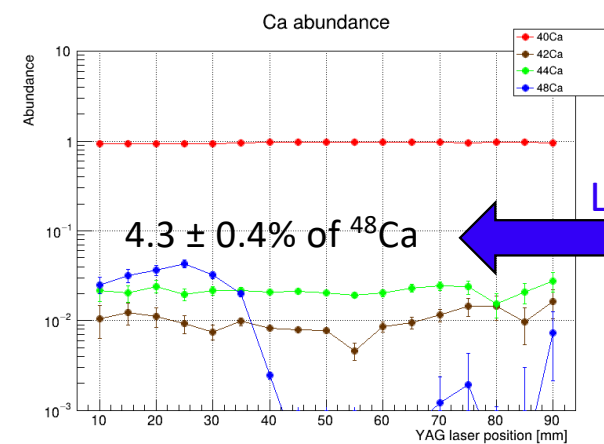
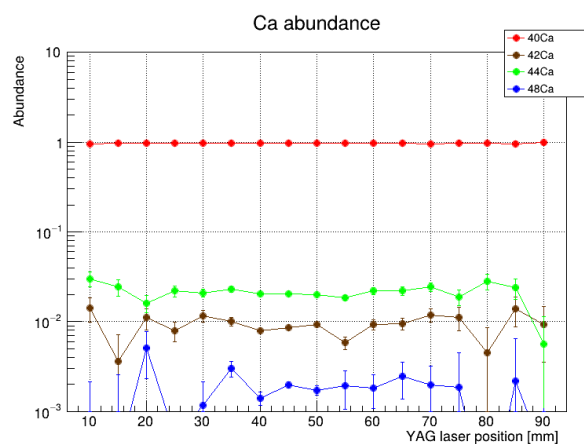
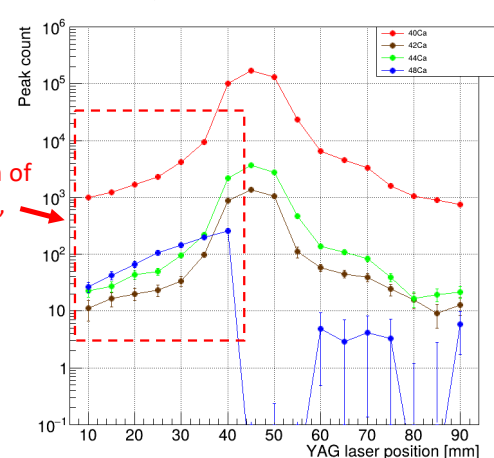
$$\Delta f = 1528.00 \pm 0.17 \text{ MHz}$$

Successfully observed the isotope shift of ^{48}Ca

Spatial distribution of the atomic beam using TOF
Beam off

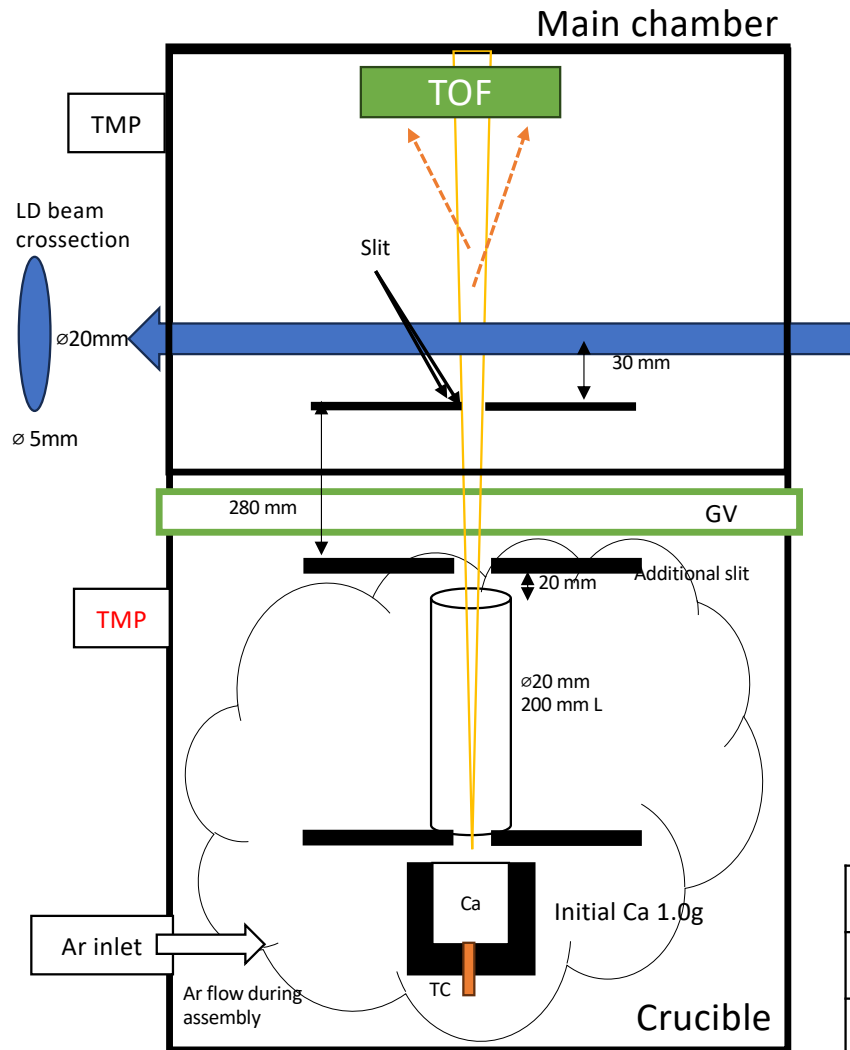


Beam on ^{48}Ca



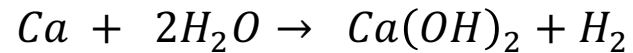
Successfully observed the enrichment of ^{48}Ca by laser deflection

Mitigate the scattering



After extensive of investigation, we found that the scattering was caused by **moisture** inside the crucible.

- Ash of 0.5 g in 1.0 g Ca grains remains.
- H₂ gas was observed during heating.

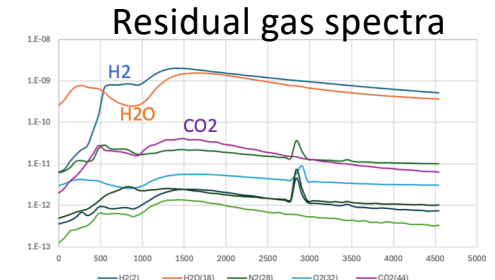


Then,

- We started to replace the Ca grains in a glove bag with an Ar, keeping the Ca away from the air.
(H₂O is from Ca grains and crucible)
- Additional TMP pump for crucible

They improve the the chamber and crucible pressure:

	Main chamber	Crucible
Before	2×10^{-4}	2×10^{-4}
After	6×10^{-5}	4×10^{-5}



Working in a glove bag



Remnant in the crucible



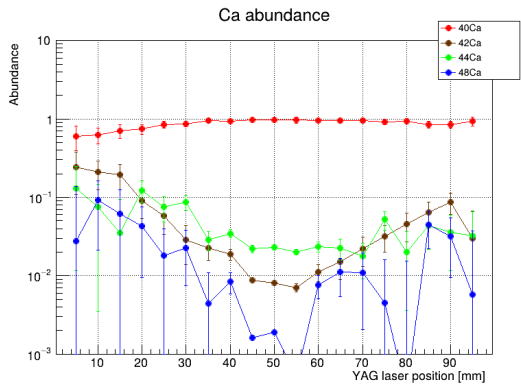
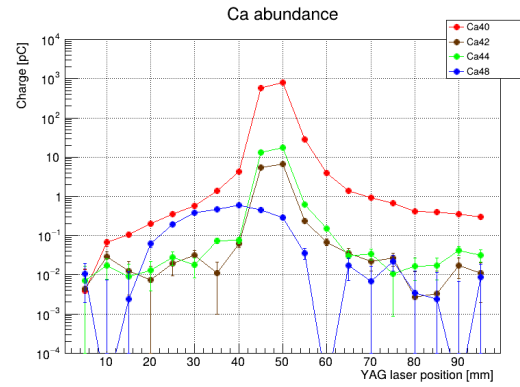
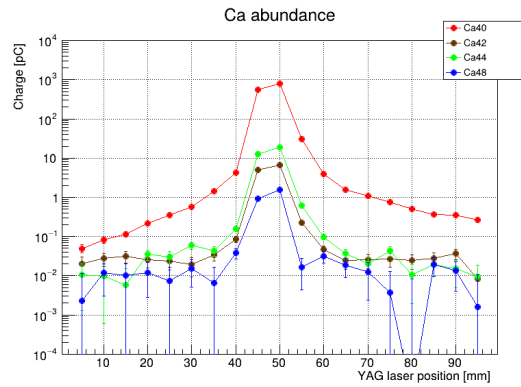
Before

Now

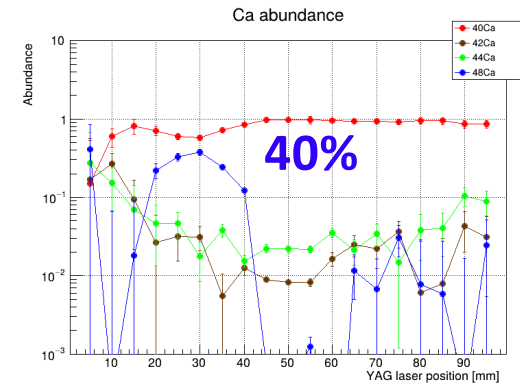
Improved LIS results (2026)

P33: A. Rittirong:

The Status of Laser Isotope Separation (LIS) of ^{48}Ca for the Study of Neutrinoless Double Beta Decay

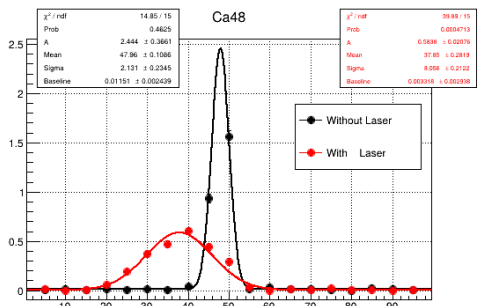
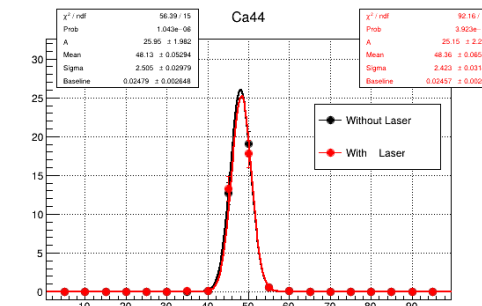
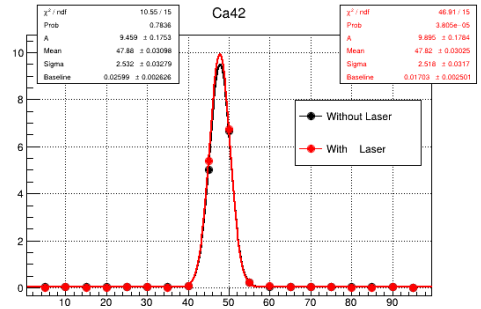
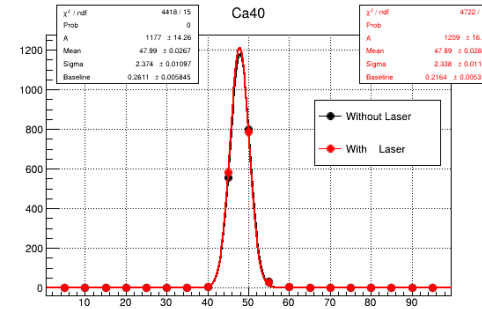


Laser off



Laser on ^{48}Ca

^{40}Ca Sigma = 2.37 ± 0.01 mm

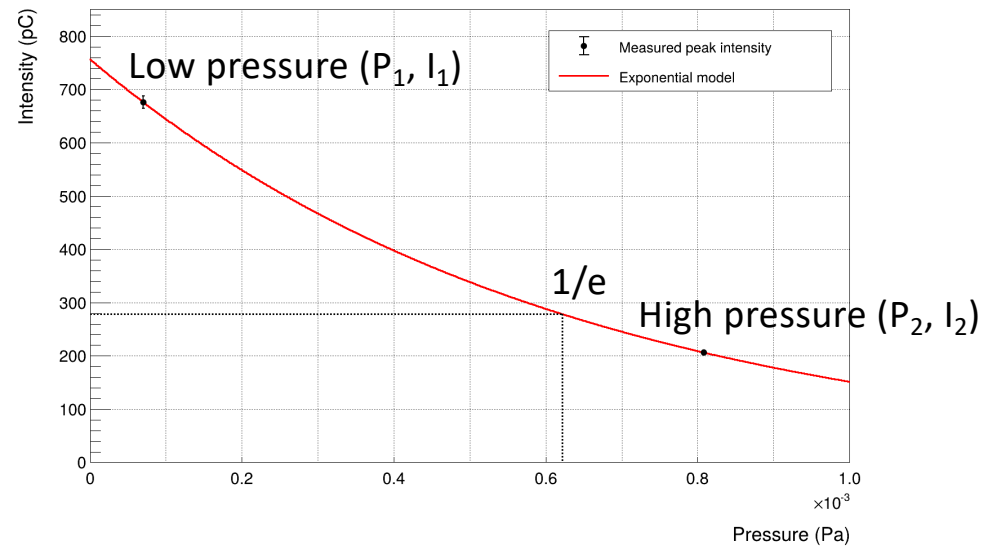
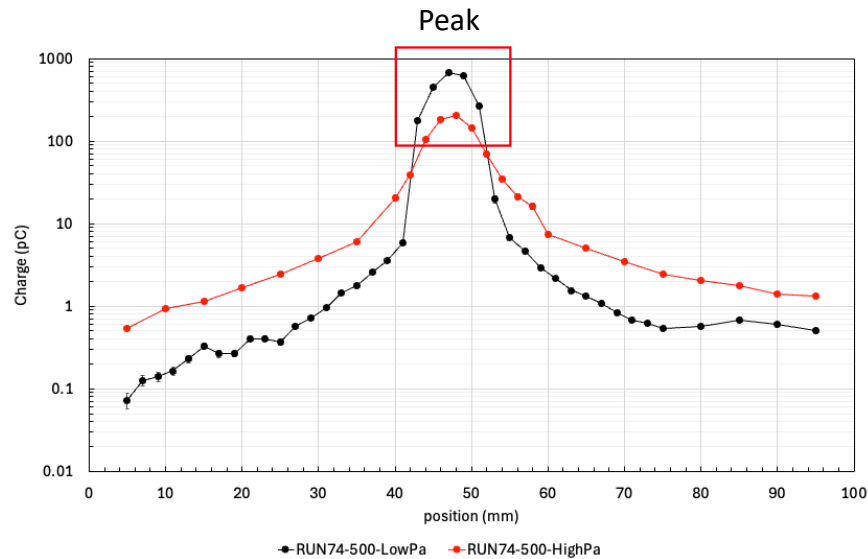


Sigma = 2.13 ± 0.23 mm, Sigma = 8.066 ± 0.21 mm
Displacement = $47.96 - 37.45 = 10.51$ mm

Crucible: 500 degC
Main chamber: 9.77E-5 Pa
Crucible: 1.50E-4 Pa

Achieved 40% ^{48}Ca enrichment, x200 times of enrichment!!

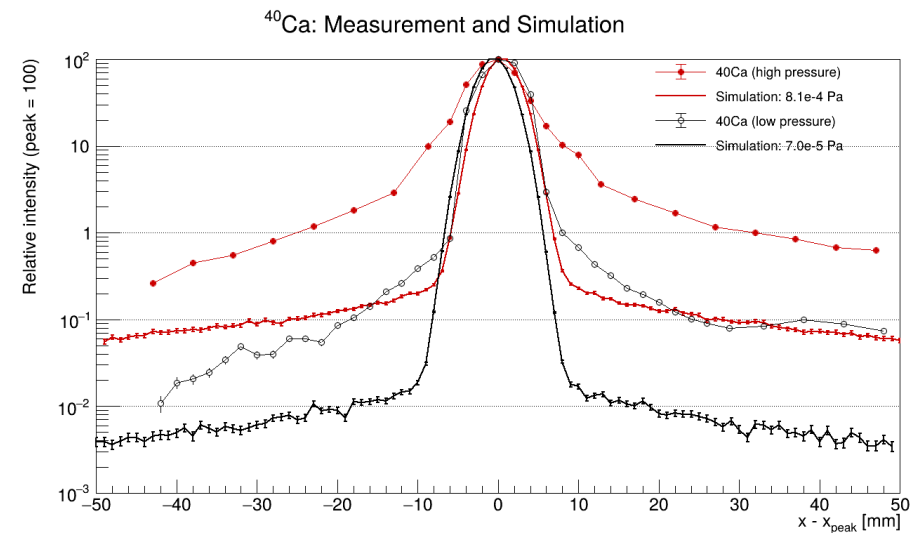
Pressure effect



Region	Low pressure: 7.0×10^{-5} Pa	High pressure: 8.1×10^{-4} Pa
Peak	676 ± 12 pC	206 ± 5 pC

$$I(P) = I_0 e^{-P/P_c} \Rightarrow I(P_c) = \frac{I_0}{e}$$

$$P_c = (6.22 \pm 0.15) \times 10^{-4} \text{ Pa}$$



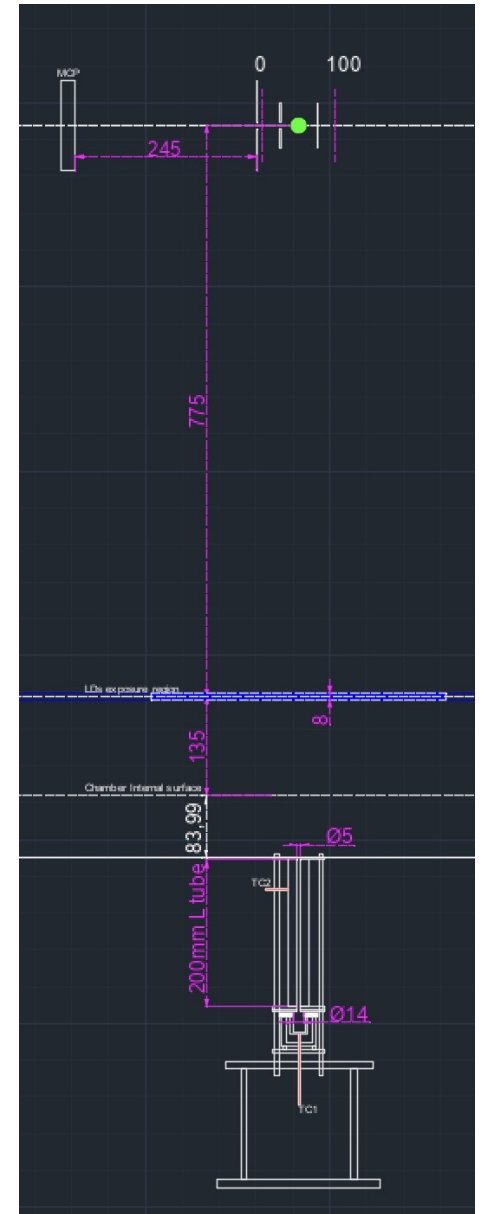
Experiments have more excess?
Need more investigation

Amount of Ca

Estimation of evaporated Ca amount

- Present Ca beam size is collimated to $2 \times 5 \text{ mm}^2$ at 380 mm above of the crucible → Only 1.1×10^{-5} of evaporated Ca is deposited on the target plate.
- Ca amount in the crucible is 1.0 g
 - 0.8 g is stacked on the slit.
 - 0.2 g is evaporated.
- Thus, 2.2 ug of $^{\text{nat}}\text{Ca}$ (13 nm) is deposited.
- The amount of ^{48}Ca is estimated as 4 ng .

The real challenge is increasing this value!
Our first goal is to make 1 mg of ^{48}Ca .

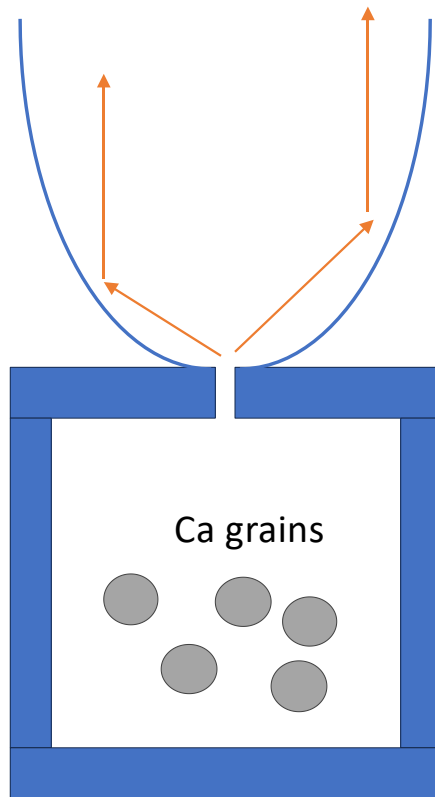


Effective use of the Ca

P37 : Rei Tanaka

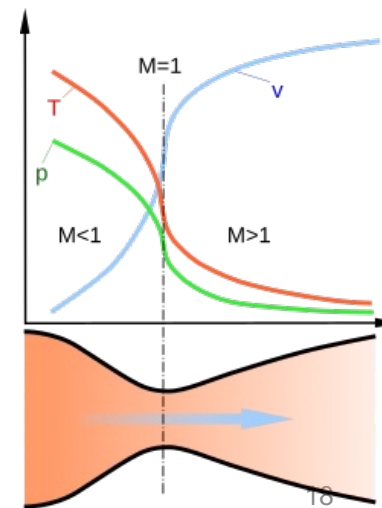
Optimization of an Atomic Beam System for Laser Isotope Separation of ^{48}Ca

Parabolic mirror



- The divergence of the emitted Ca beam must be controlled.
- In the molecular-flow regime, a pinhole combined with a parabolic reflector may provide beam collimation.
- In the viscous-flow regime, a Laval nozzle may be an effective solution.

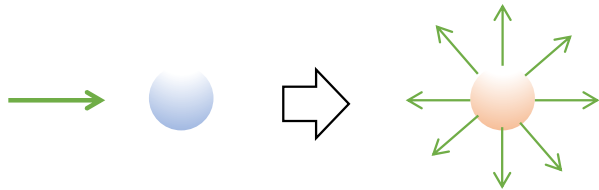
De Laval nozzle



https://en.wikipedia.org/wiki/De_Laval_nozzle

LIS with Rabi oscillation for re-use of the laser

Conventional excitation and de-excitation



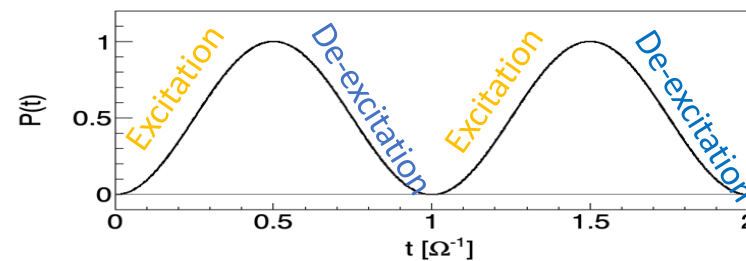
Absorption excites the atom, followed by emission in a random direction.

Present LIS system absorbs ~ 500 photons. Then, the laser light is lost in isotropic emission the process.

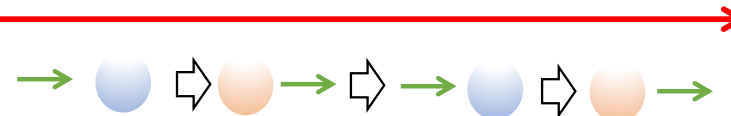
Thus, $3 \text{ eV} \times 500 = 1.5 \text{ keV}/^{48}\text{Ca}$ is required. This will be the rate-limiting factor for the mass production.

Can we re-use the laser light?

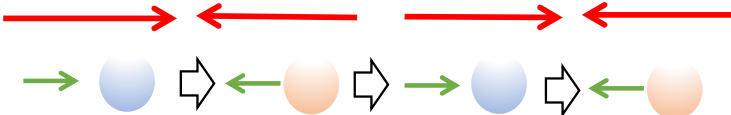
Excitation and de-excitation via Rabi oscillations



Fixed laser direction



Alternating laser direction

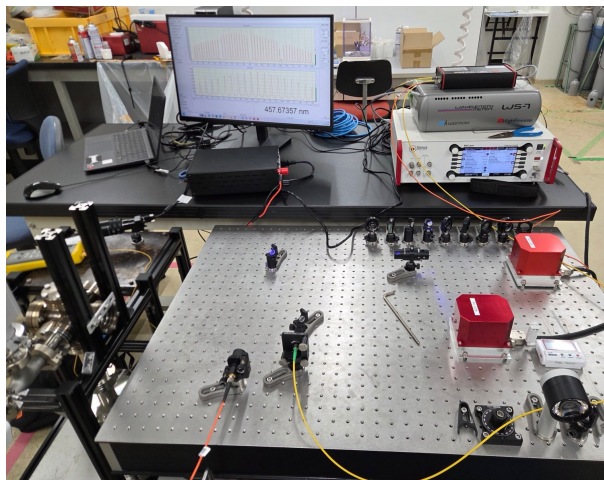
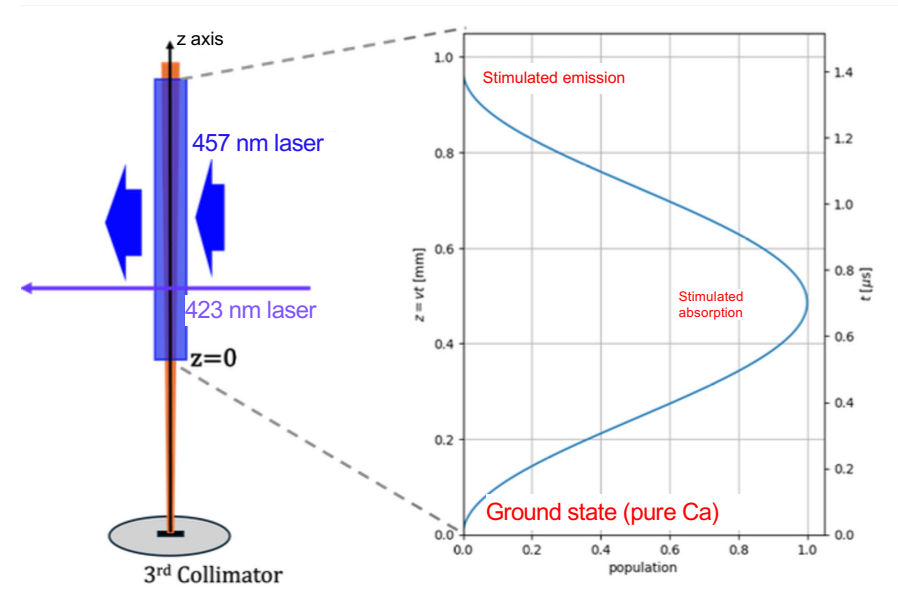
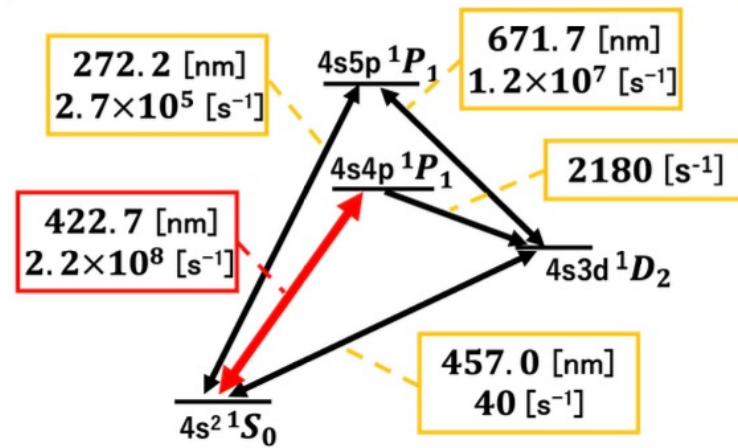


Reversing the pulsed laser direction in sync with Rabi oscillations deflects the atoms. Reflections allow repeated use of the laser light.

Proof-of-Principle Test of the Rabi Oscillation Method

P38: H. Iseki

Rabi oscillation for isotope enrichment



- Induce Rabi oscillations on the $1S_0 \rightarrow 1D_2$ transition using a 457 nm laser.
- Verify the transition using a 423 nm laser by detecting the depletion of the ground-state population.

Summary

- CANDLES experiment for neutrinoless double-beta decay ($0\nu\beta\beta$) is taking advantage of the high Q-value of ^{48}Ca (4.27 MeV).
- For the next generation experiment, we are developing:
 - MMC-based calorimeters (0.5% FWHM at 4.27 MeV)
 - Methods to achieve $\mu\text{Bq/kg}$ -level radiopurity
 - ^{48}Ca enrichment technology (natural abundance: 0.187%)
- At the University of Osaka, we are developing ^{48}Ca enrichment using laser deflection.
 - We achieved 40% ^{48}Ca enrichment. Removing water was crucial.
 - The amount of enriched ^{48}Ca obtained so far is estimated at ~ 4 ng.
 - We are working toward producing 1 mg.

Back up

Design of the new crucible

- We are currently requesting a quotation from C&A Co.

Design of the new crucible

- Size : 0.3 Liter
- Graphite heater
- Materials
 - Graphite crucible
 - Carbon radiation shield
 - Quartz spacer

