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Current Status and Preparation for the Production of ^{48}Ca by Laser Isotope Separation (LIS)

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[5]ILE, Osaka Univ.

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[7]Inst. Laser Tech.

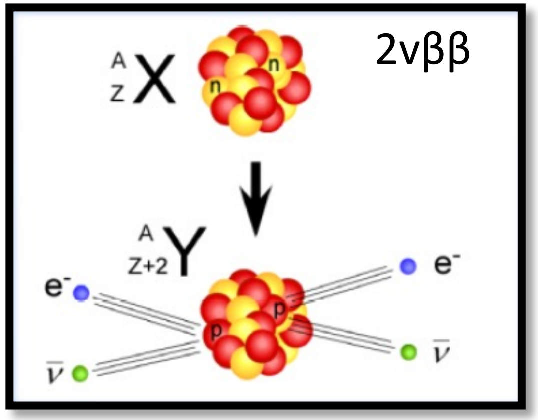
CAlcium fluoride for studies of **N**eutrino and **D**ark matters by **L**ow **E**nergy **S**pectrometer (CANDLES)

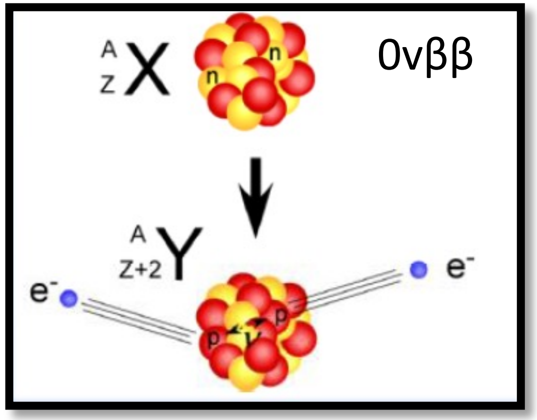
Candles

Majorana particle:
Particle identical to its antiparticle
E. Majorana, 1937

Dirac
$$\mathcal{L}_D = -m_D \overline{\nu_R^0} \nu_L^0 + \text{ h. c.}$$

Majorana
$$\mathcal{L}_{m_L} = -\frac{m_L}{2} \overline{(\nu_L^0)^c} \nu_L^0 + \text{ h. c.}$$



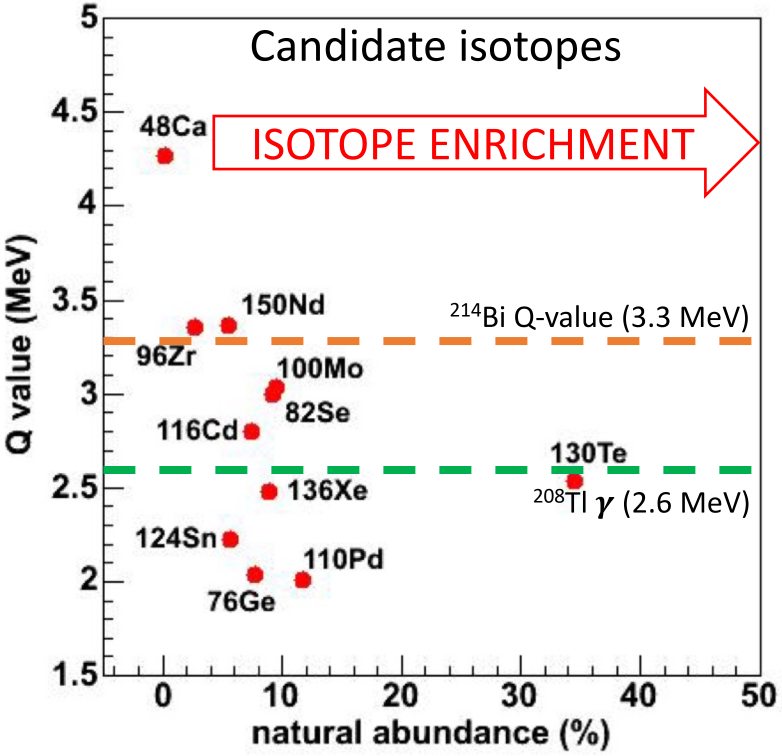


2vββ
Within standard model

0vββ
Beyond the standard model !!

Violates the lepton number conservation law
- Majorana particle
- Particle <-> Anti-particle
- Matter dominated universe

Absolute neutrino mass

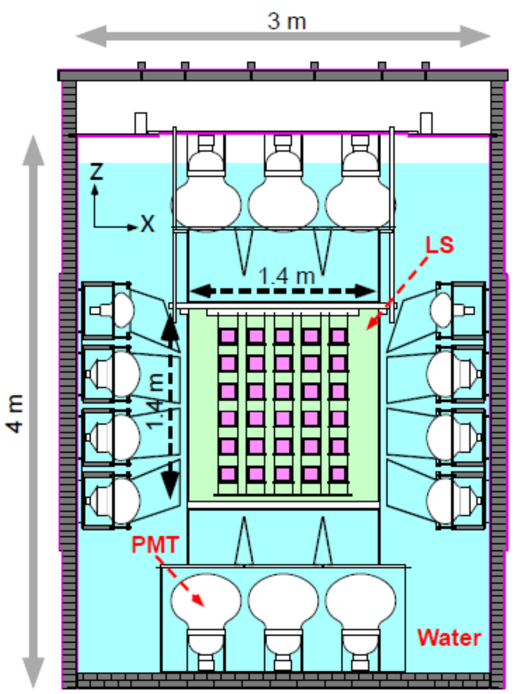


https://www-conf.slac.stanford.edu/ssi/2010/Hall080610.pdf

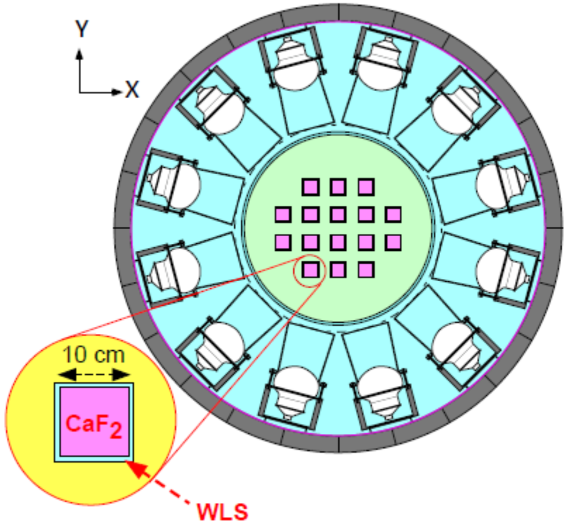
For better sensitivity
- Low background
  - Measurement and analysis
- Higher Q-value
  - Overcome natural radioactive BG
  - ^{48}Ca = 4.3 MeV
- A large amount of double beta decay nuclide
  - Isotope enrichment

CAlcium fluoride for studies of **N**eutrino and **D**ark matters by **L**ow **E**nergy **S**pectrometer (CANDLES)

Candles

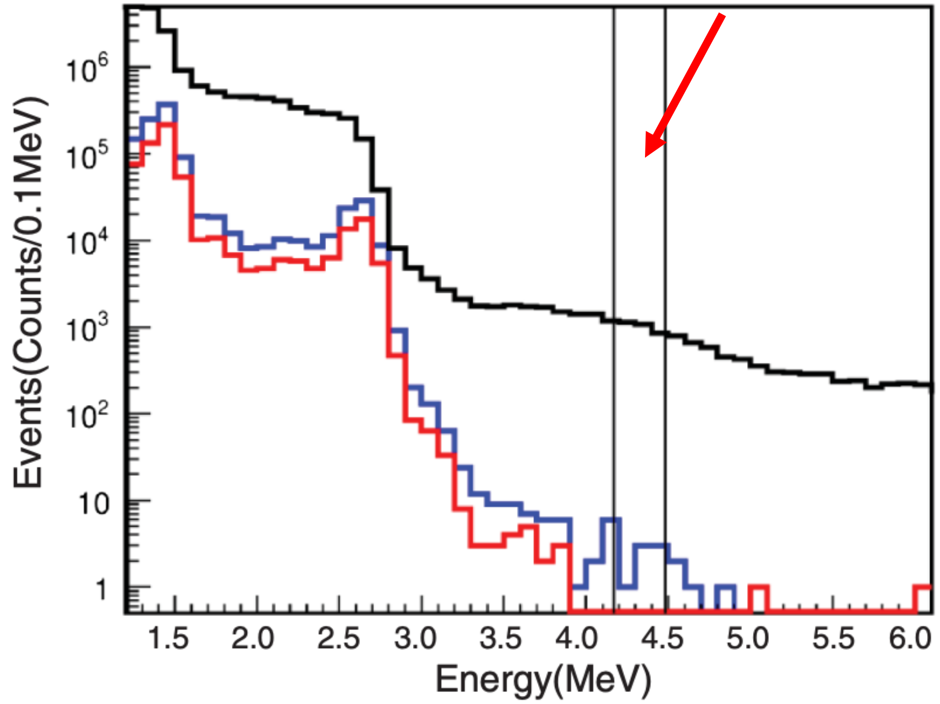


S. Ajimura, et al. *Physical Review D* 103.9 (2021): 092008.

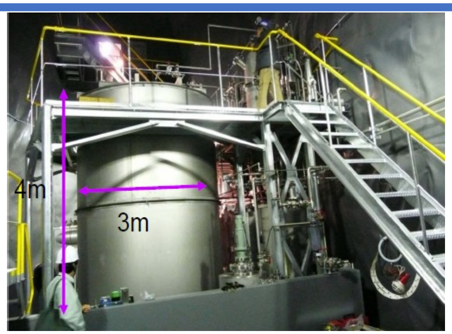


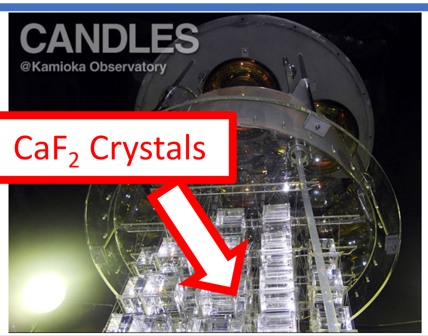
^{48}Ca N.A. = 0.187 %

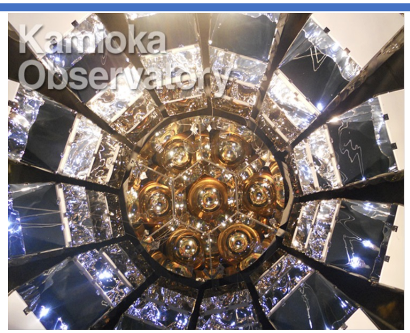
^{48}Ca ROI
4.17 – 4.48 MeV



CANDLES III – 305 kg **CaF_2 Crystals** (3.2 kg × 96 pieces) **0.35 kg ^{48}Ca**







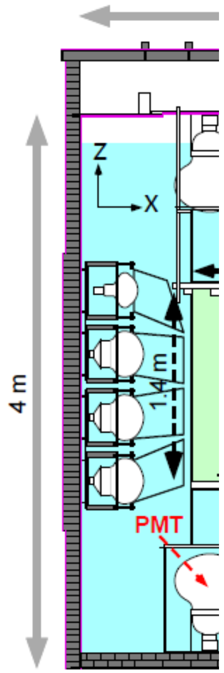
10^{-3} events/keV/yr/(kg of ^{nat}Ca)

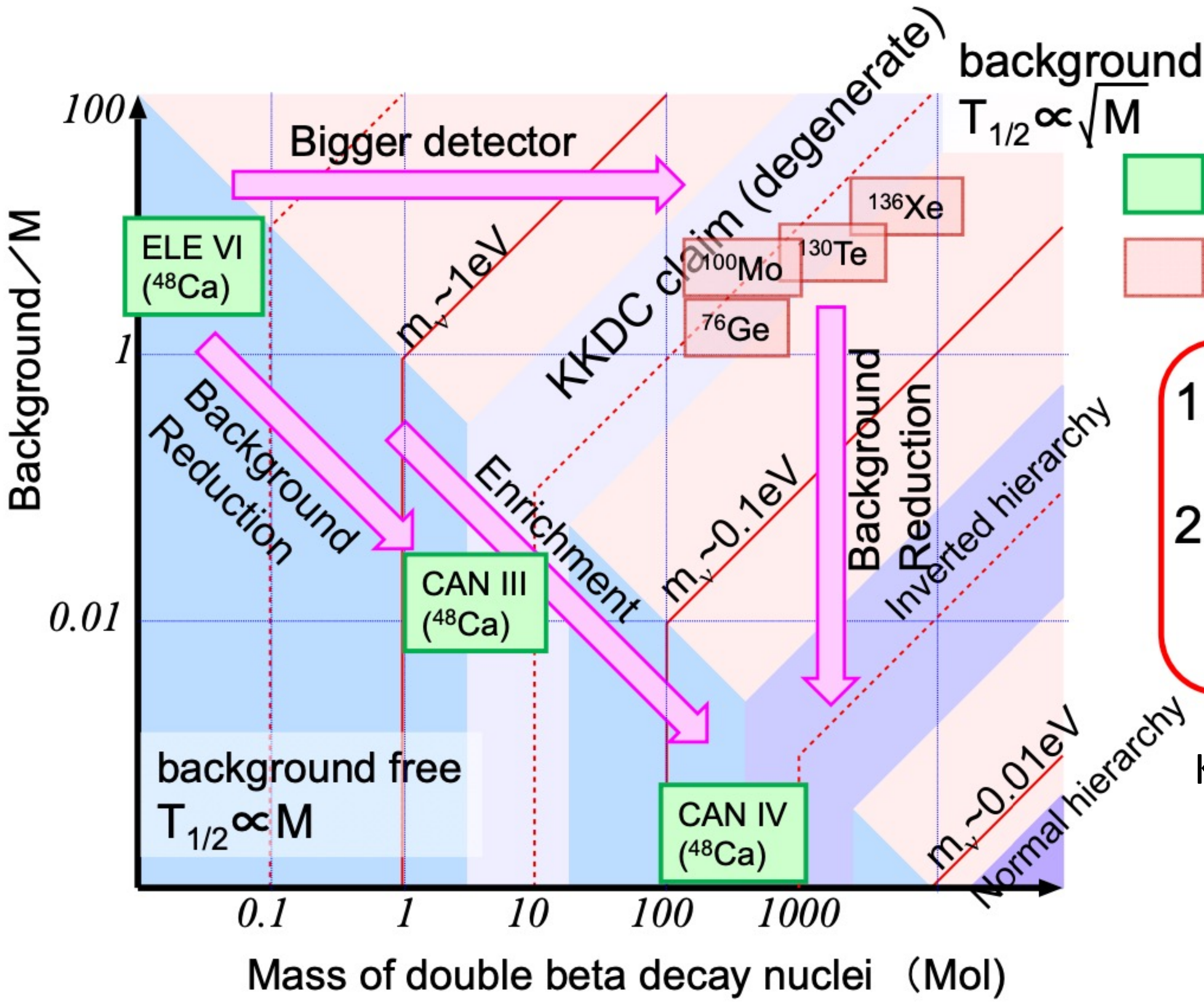
$$T^{1/2}_{0\nu\beta\beta}\ \mathrm{Ca-48} = 5.6\times10^{22}\ \mathrm{years}\ (90\%\ \mathrm{C.L.})$$

Majorana neutrino mass (m_ν) = ≤ 2.9 – 16 eV (90% C. L.)

CAlcium … ters

Our strategy



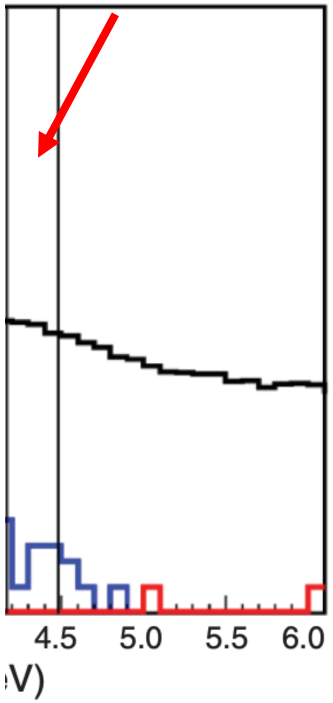


1. Achieve BG free measurement
2. Scale up
   Enrichment
   Improve S/N

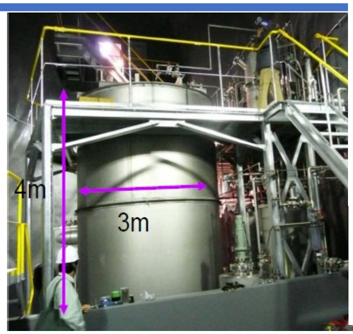
Kishimoto ICRR 2014

Ca ROI

17 – 4.48 MeV



CANDLES III – …



; of ^{nat}Ca)

ears (90% C. L.)

) = ≤ 2.9 – 16 eV

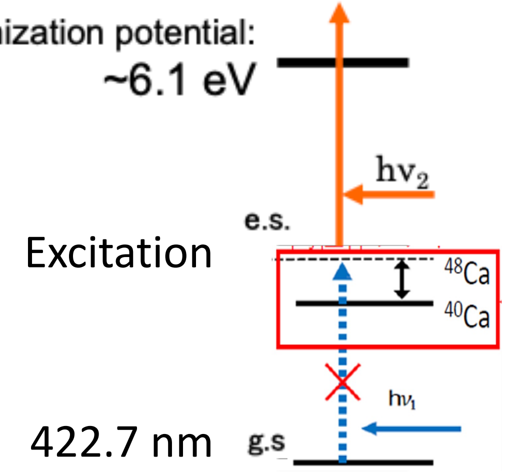
Isotope enrichment of ^{48}Ca is the key for the search of 0νββ

Calcium-48 [N.A. = 0.187%]

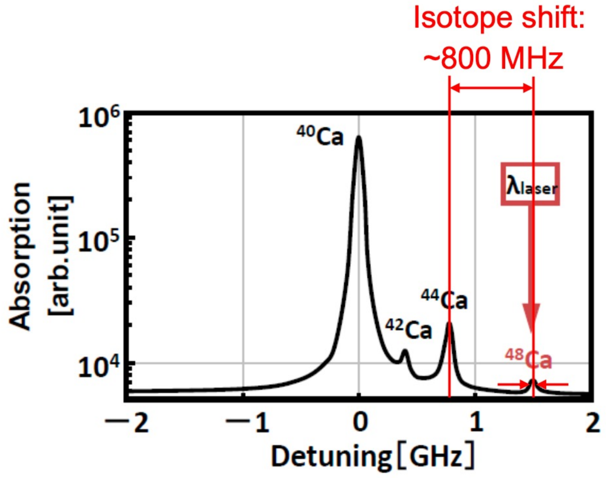
- **Ca has no gaseous compound**
- Can be enriched by electromagnetic separator (Expensive)
  - ^{48}Ca 10 grams/year (By MS) -> 1,000,000 $/g **(~1億円/g)**

Isotope separation technique	Separation Coefficient	Production efficiency (y^{-1})	Cost	Limitation
Electromagnetic separator	High	Ten of grams	High	○ High power consumption ○ Low productivity
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 ○ Solubility problem ○ Development of the cascade enrichment is required
Ion exchange chromatography	Small	Hundred of gram	Low	○ Time consumption ○ Low conversion
Laser isotope separation	High	Kiligrams	Middle	○ Development of the high-power laser, irradiation unit, and collection system

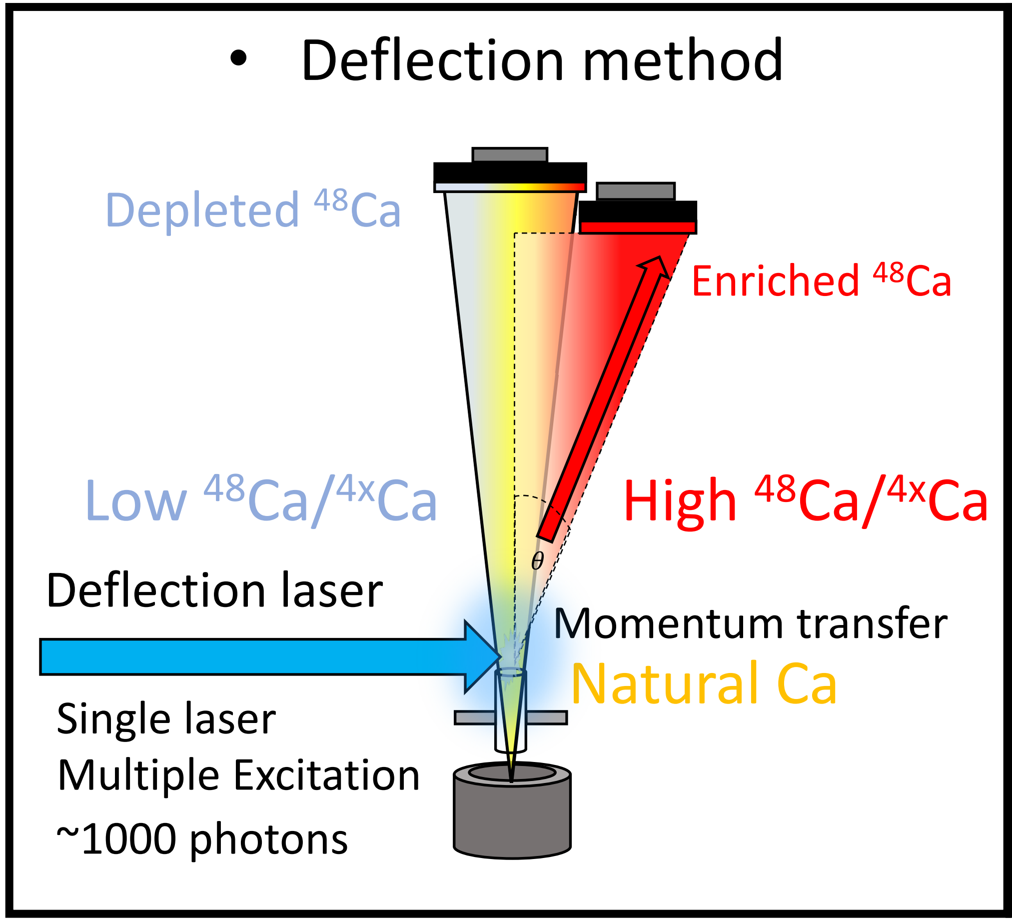
Laser Isotope Separation (LIS)

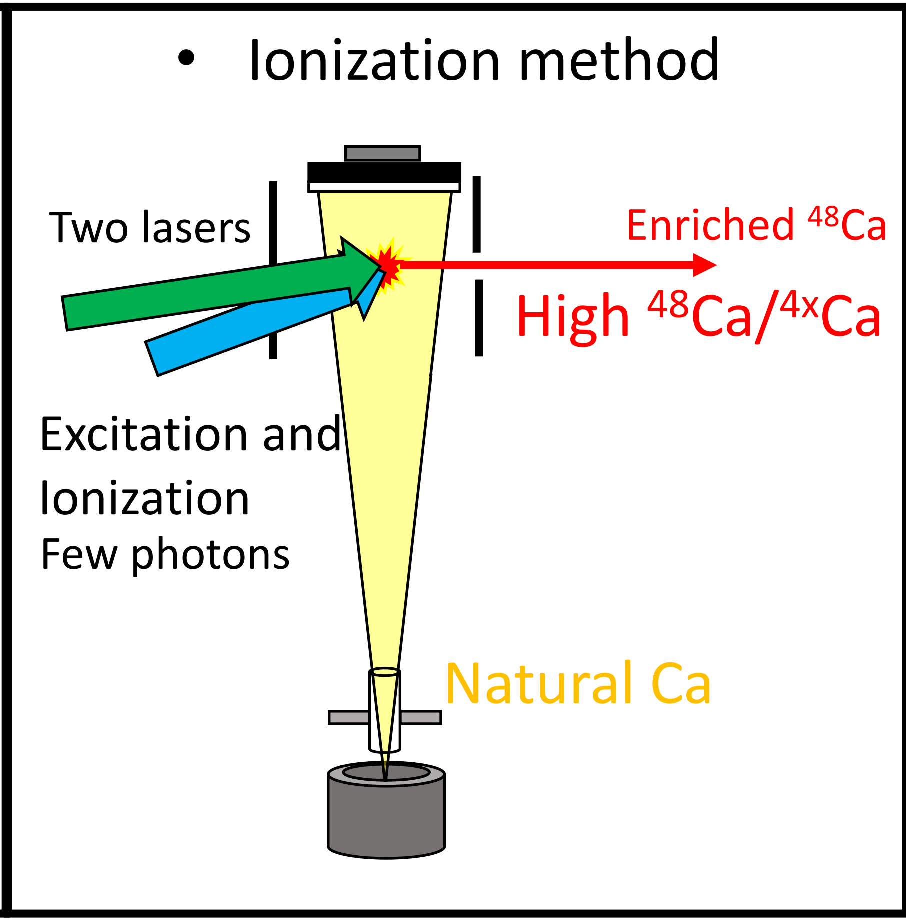


Isotope shift of Ca at 422.7 nm



Absorption spectrum of Ca at 423nm





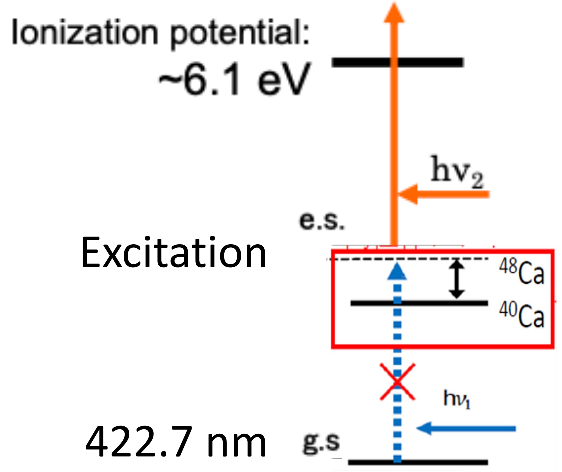
Ionization = high enrichment coefficient, low productivity

Deflection = moderate enrichment coefficient, high productivity

The DEFLECTION method was applicable for mass production

K Matsuoka *et al* 2020 *J. Phys.: Conf. Ser.* **1468** 012199

Laser Isotope Separation (LIS)



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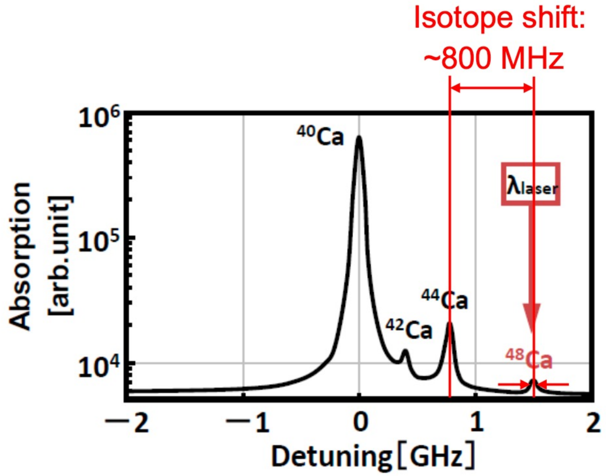
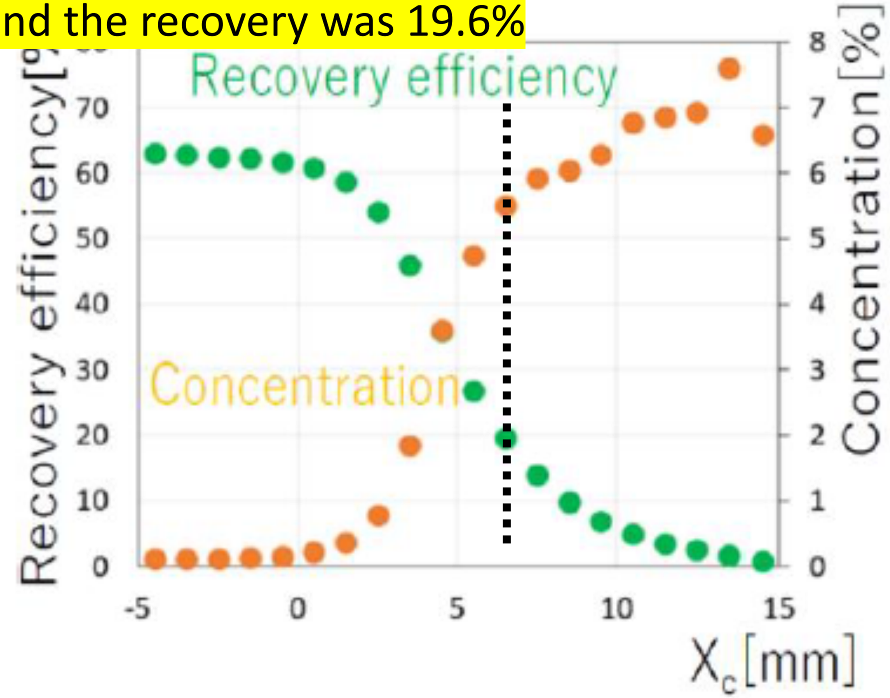


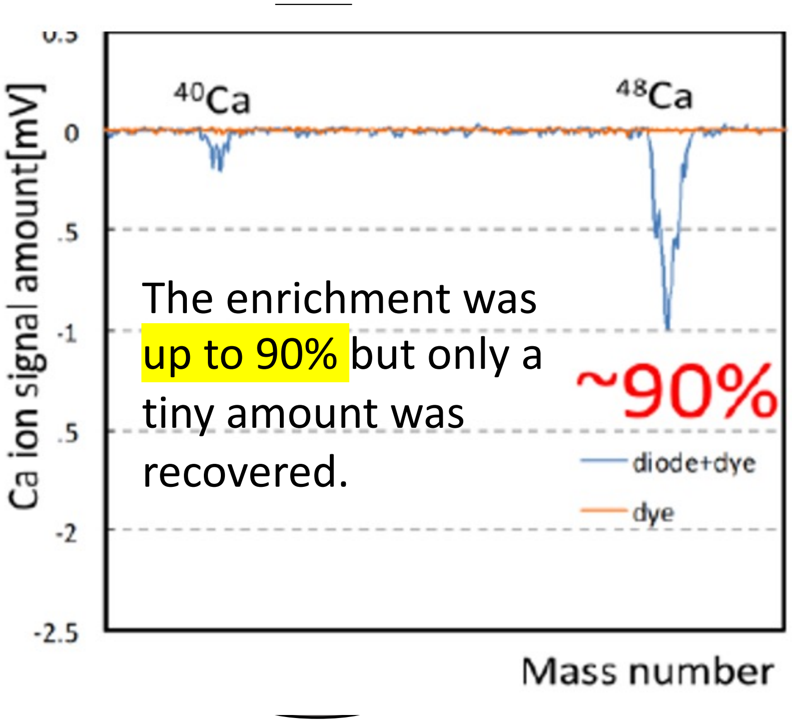
Absorption spectrum of Ca at 423nm

- Deflection method

The enrichment was 5.5% and the recovery was 19.6%



- Ionization method



The enrichment was up to 90% but only a tiny amount was recovered.

Ionization = high enrichment coefficient, low productivity
Deflection = moderate enrichment coefficient, high productivity

→ The DEFLECTION method was applicable for mass production

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Research strategies and requirements

University of Fukui

Proof of principle
- Small scale chamber and single laser system
- TOF measurement, deposition meter

Atomic beam system
- Small scale chamber
- Increase the tube number to make a sheet-like atomic beam system
- Collimator effect

Institute for Laser Technology
Institute of Laser Engineering

Laser system
- Single frequency laser
- Power-scalable laser
- SOA, multiple slave laser
- Long-term operation
- Stable laser system

Research Center for Nuclear Physics

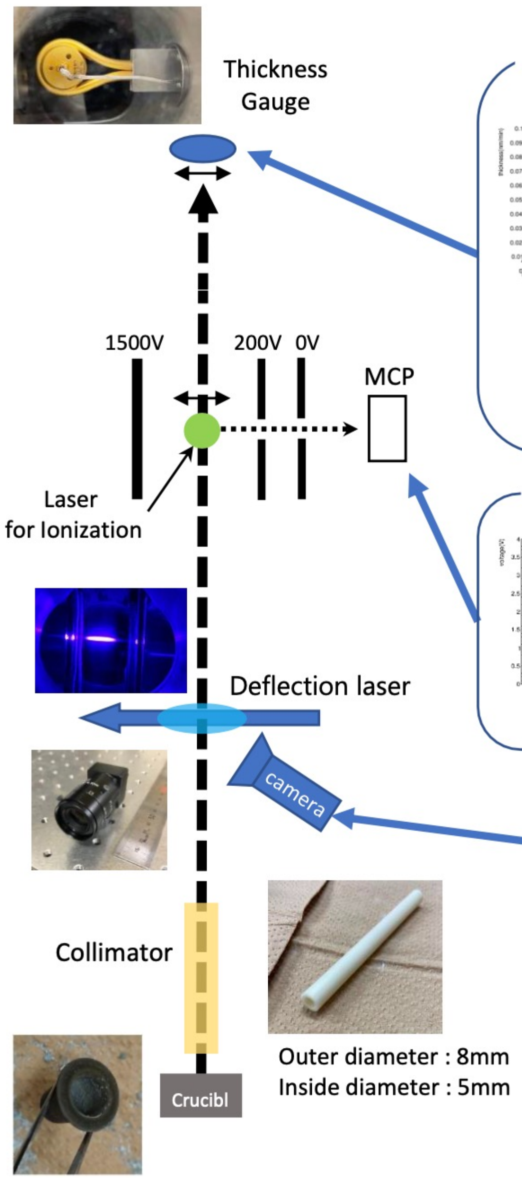
Collection and monitor system
- Collection plate
- Recovery system

Research Center for Nuclear Physics

Production system of ^{48}Ca
- Large scale chamber
- 2W laser × 6 ports → ~10g/year
- High production rate (1 mol/year)
- Automation system

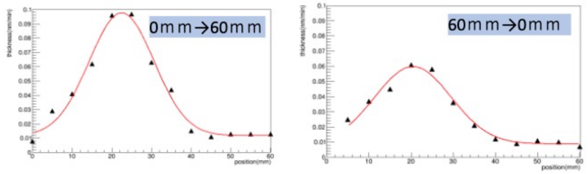
Future development
- Stable operation
- Increase the production rate by multiple 6 ports units
- 0.3 g ^{48}Ca/day
- Ton scale production

Development of atomic beam system



Thickness Gauge

Spatial Distribution



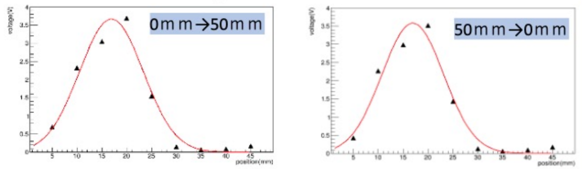
$\sigma\ =\ 8\text{mm}$ $\quad$ $\sigma\ =\ 9\text{mm}$

$\sigma^{※} =\ 5\text{mm}$ $\quad$ $\sigma^{※} =\ 5\text{mm}$

Accuracy is not good because of the BG.

TOF

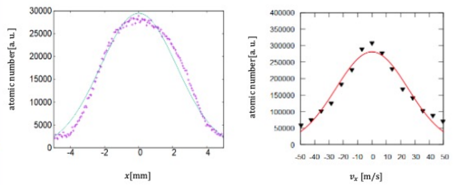
Spatial Distribution



σ = 6.422mm $\quad$ σ = 6.202mm

Fluorescence measurement

Spatial Distribution $\quad$ Velocity Distribution



x : Position coordinates perpendicular to the atomic beam

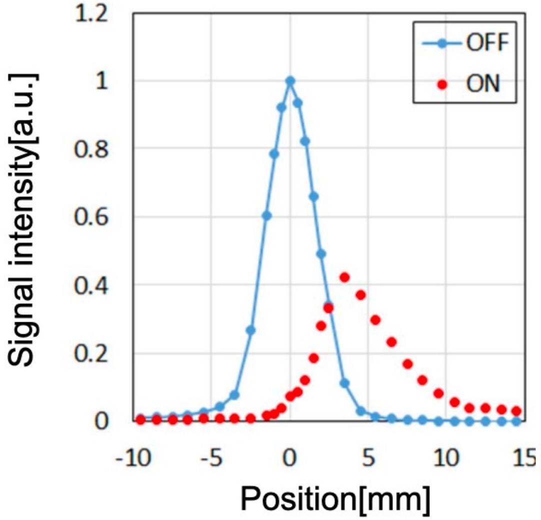
$\sigma\ =\ 2.24\text{mm}$

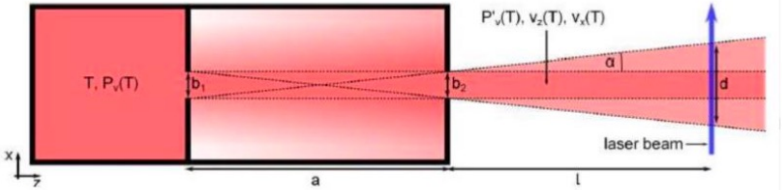
$\sigma^{※} =\ 19.1\text{mm}$

$\sigma_{v_x} = 25.1\text{m/s}$ (Measured value)

$\sigma_{v_x} = 9.2\text{m/s}$ (Calculated value)

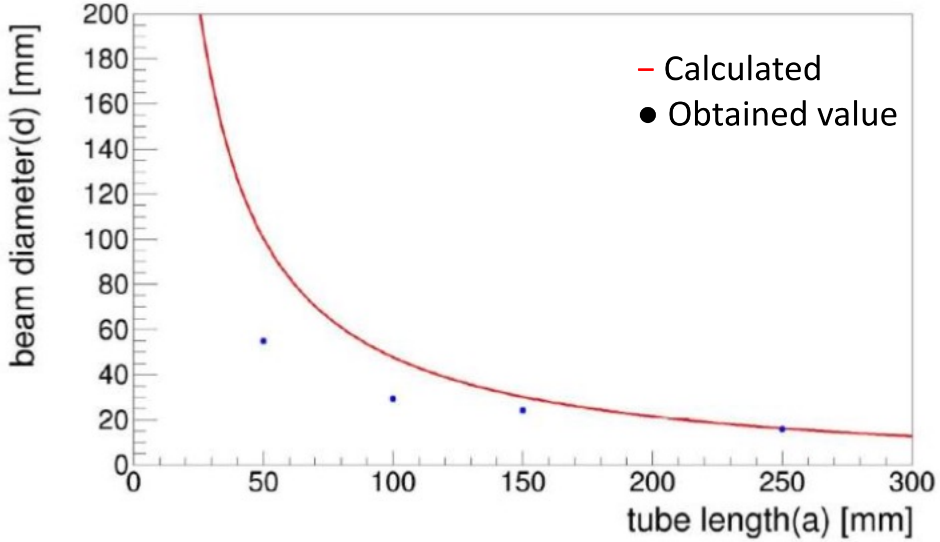
Further studies are needed.





$$d = 2\left((a+l)\frac{{b_1}/{2} + {b_2}/{2}}{a} - \frac{b_1}{2}\right)$$

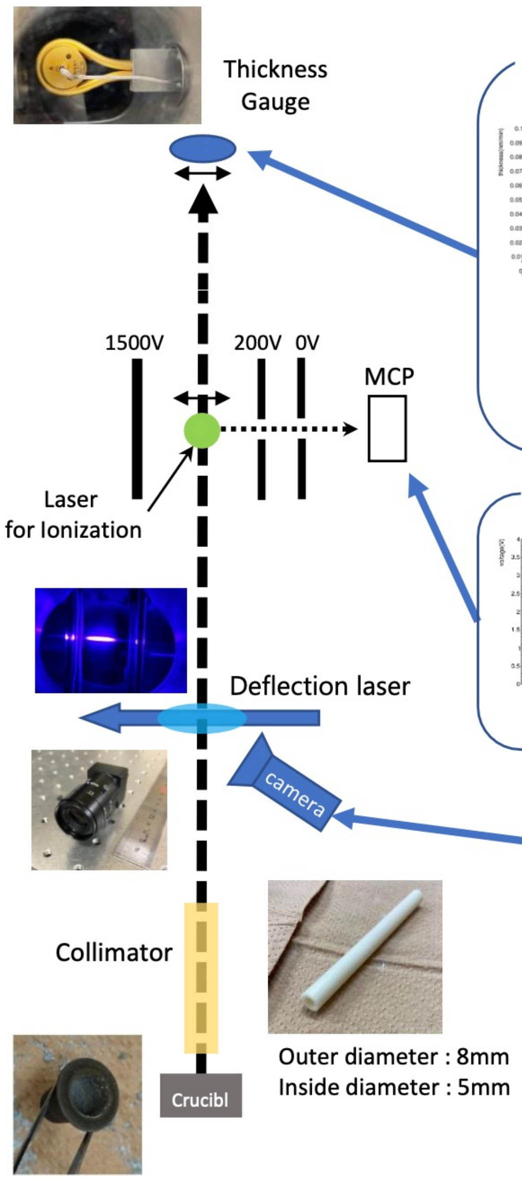
Collimator effect



The longer tube results in the narrower beam diameter

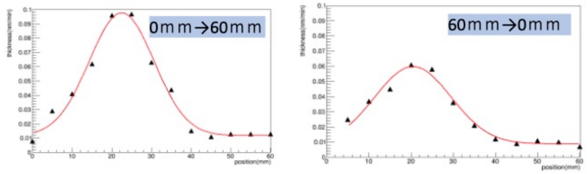
Ogawa *et al* 2022 *J. Phys.: Conf. Ser.* **2147** 012012

Development of atomic beam system



Thickness Gauge

Spatial Distribution



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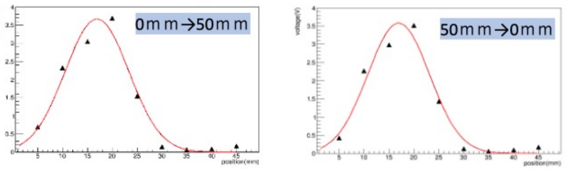
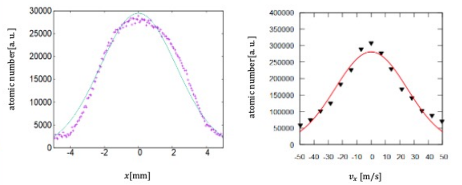
Spatial Distribution



σ = 6.422mm   σ = 6.202mm

Fluorescence measurement

Spatial Distribution | Velocity Distribution



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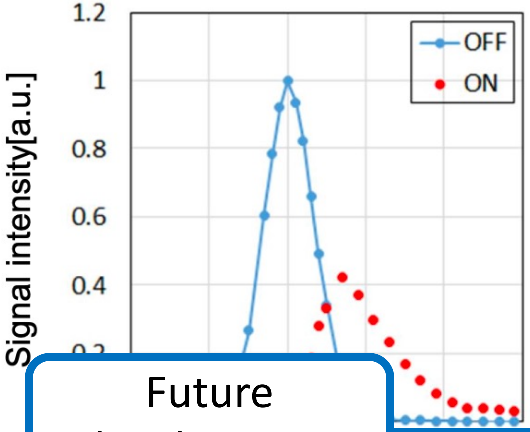
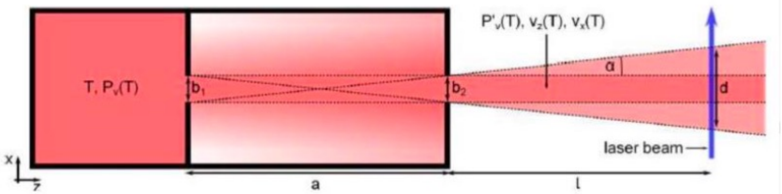
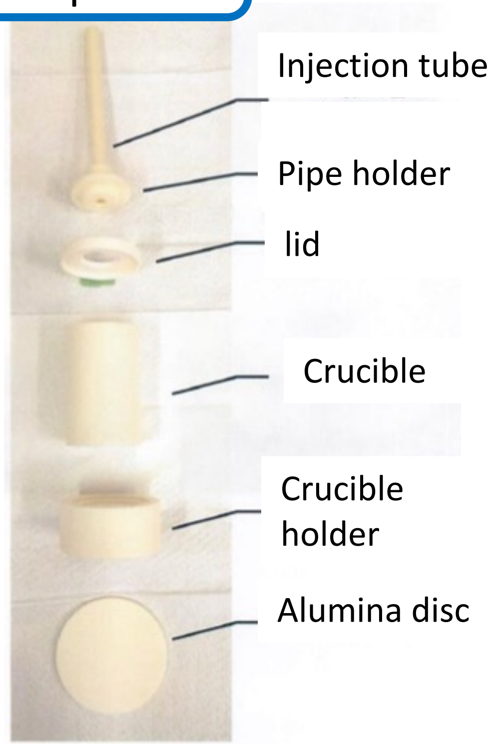
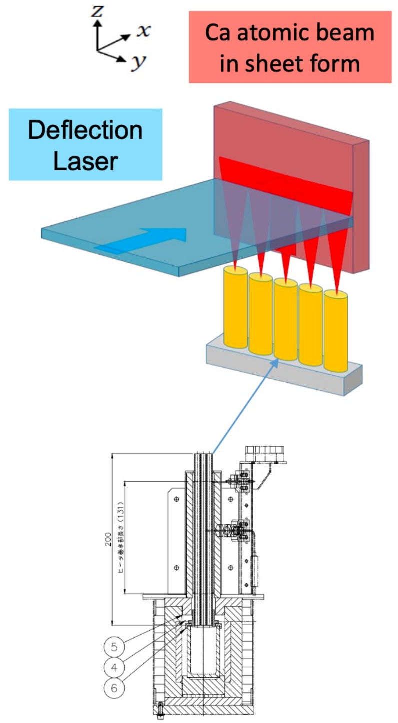




$$d = 2\left((a+l)\frac{{}^{b_1}/_2 + {}^{b_2}/_2}{a} - \frac{b_1}{2}\right)$$

Future development





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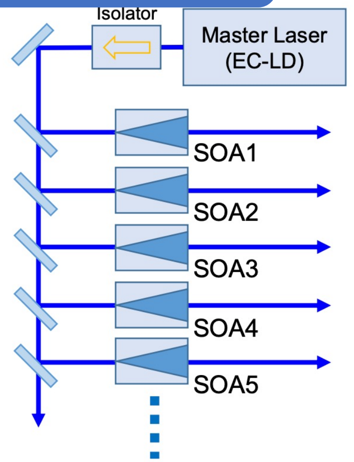
Development of the laser system

Requirements

- ✓ Wavelength: ~422.792 nm
- ✓ Frequency stability: <2 MHz rms
- ✓ Power scalability: >100 W (1 unit)
- ✓ Long life time: >30,000 hours
- ✓ Continuous wave (CW)
- ✓ High efficiency
- ✓ Low cost

MOPA- SOAs



Stable, single-frequency and high power

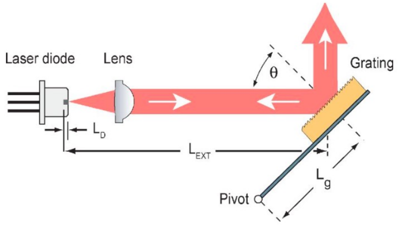
Expensive

Under development by Prof. Uemukai, Osaka University

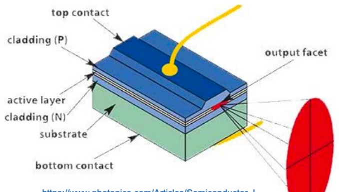
EC-LD + FP-LD [injection locking technique]

External cavity LD vs. Fabry-Perot cavity LD

Master laser | Slave laser



https://doi.org/10.1364/AO.48.006692

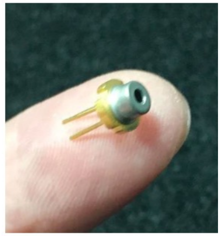


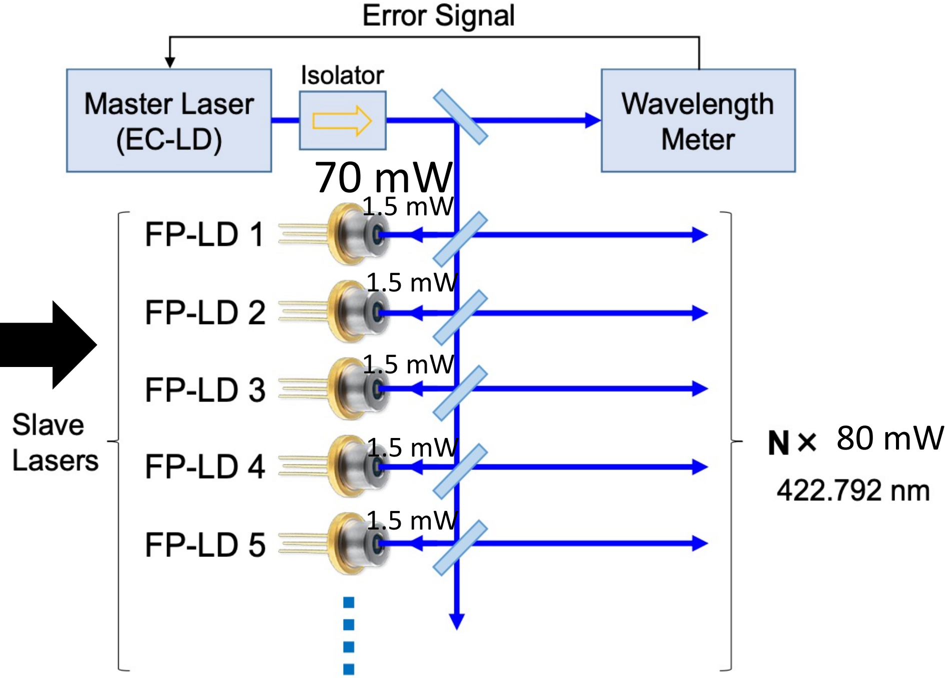
https://www.photonics.com/Articles/Semiconductor_Lasers_An_Overview_of_Commercial/a25099

- Single longitudinal-mode (Line width: <1 MHz)
- Wavelength tunable

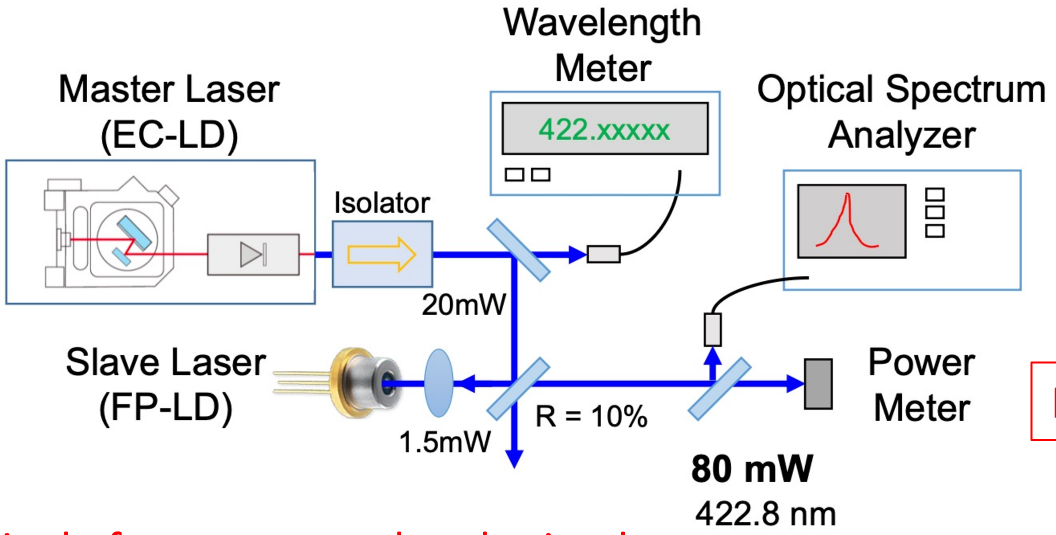
↔

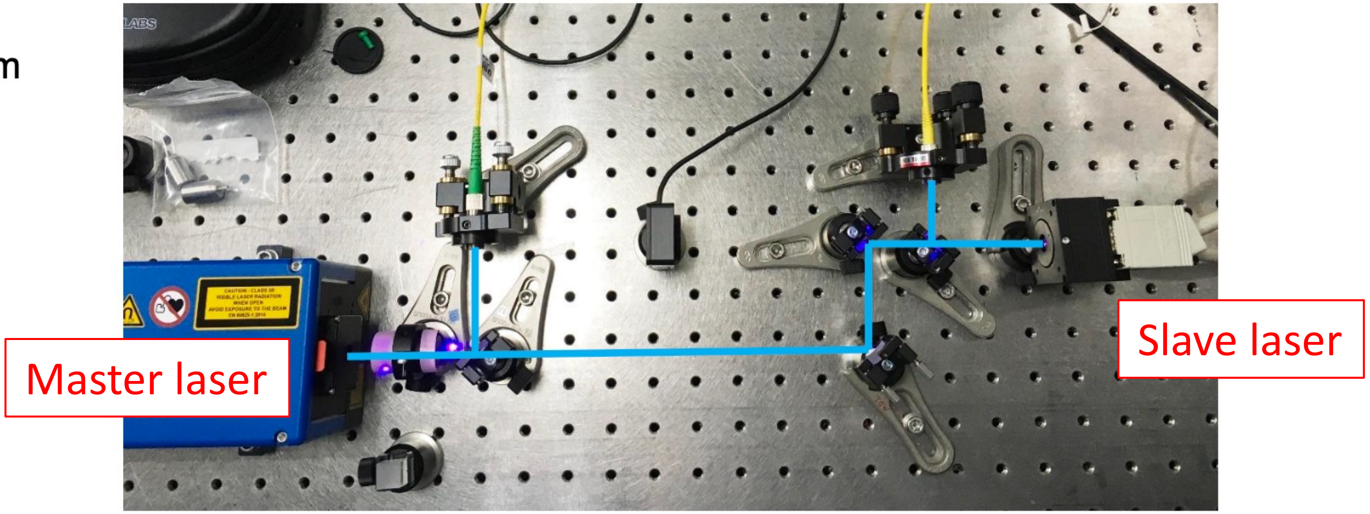
- Multi longitudinal-mode (Wide spectral width)
- Low cost
- Compact
- High efficiency



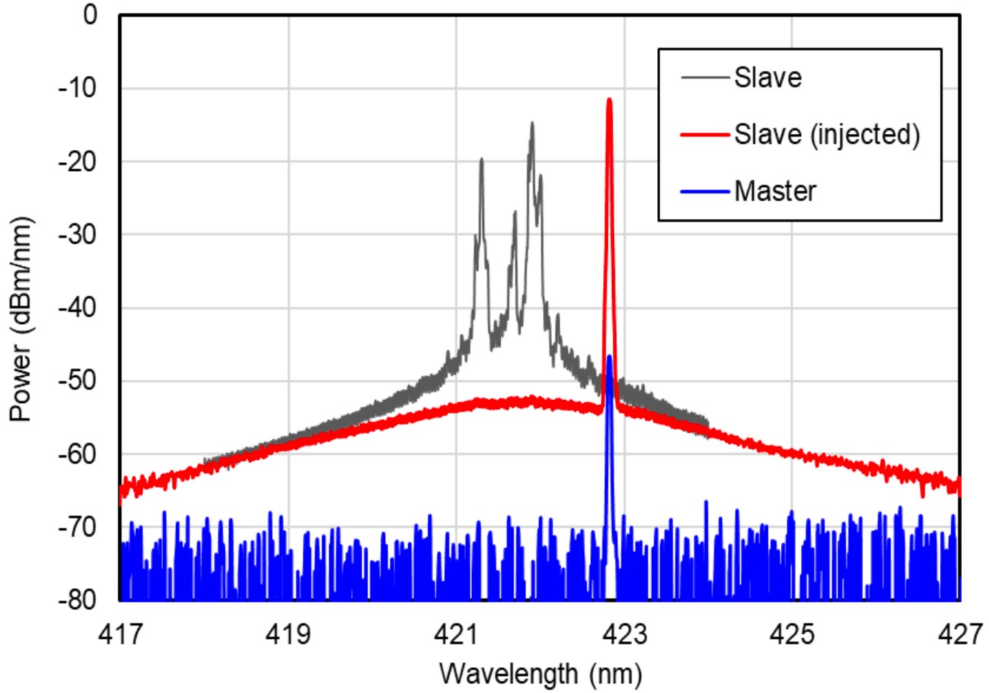


Development of the laser system EC-LD and FP-LD

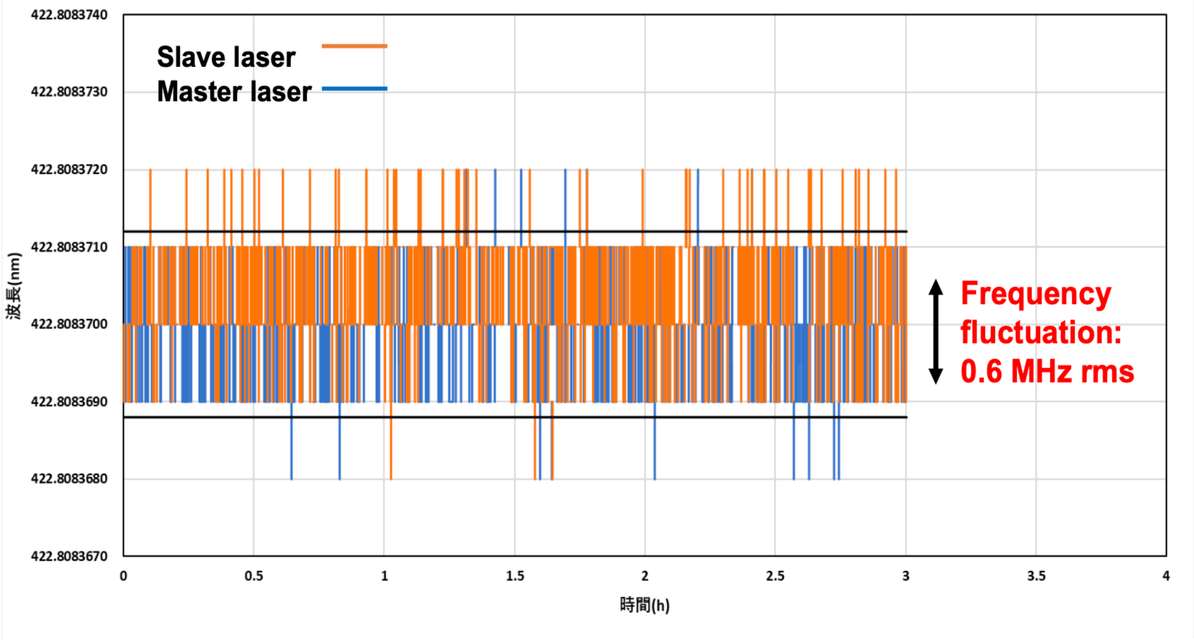




Single frequency can be obtained.



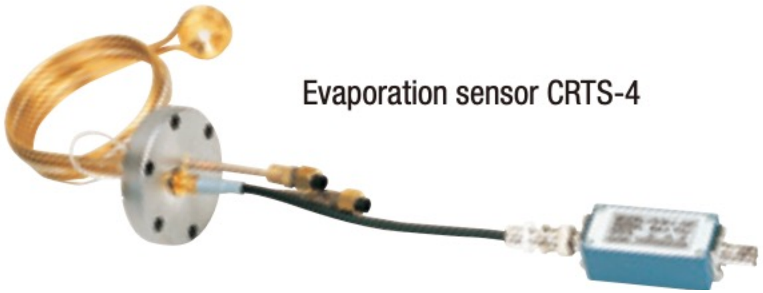
< 2MHz rms can be obtained

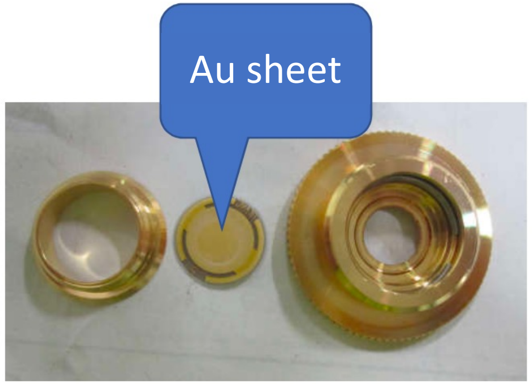


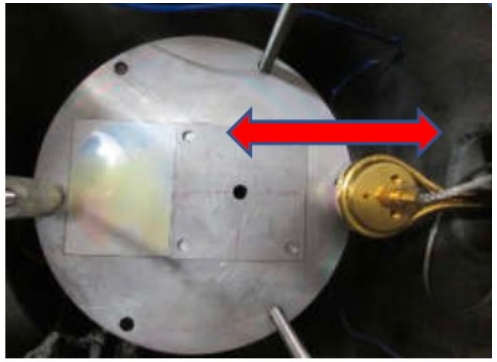
Development of production system

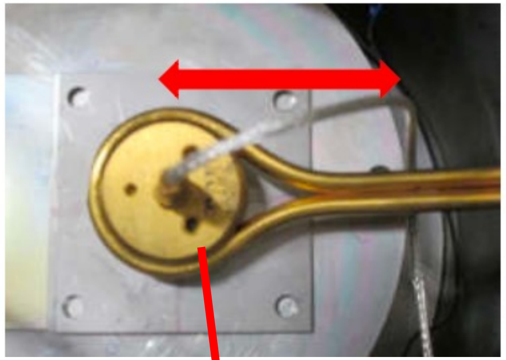
- To monitor atomic beam

Deposition meter
CRTM9200 + CRTS-4

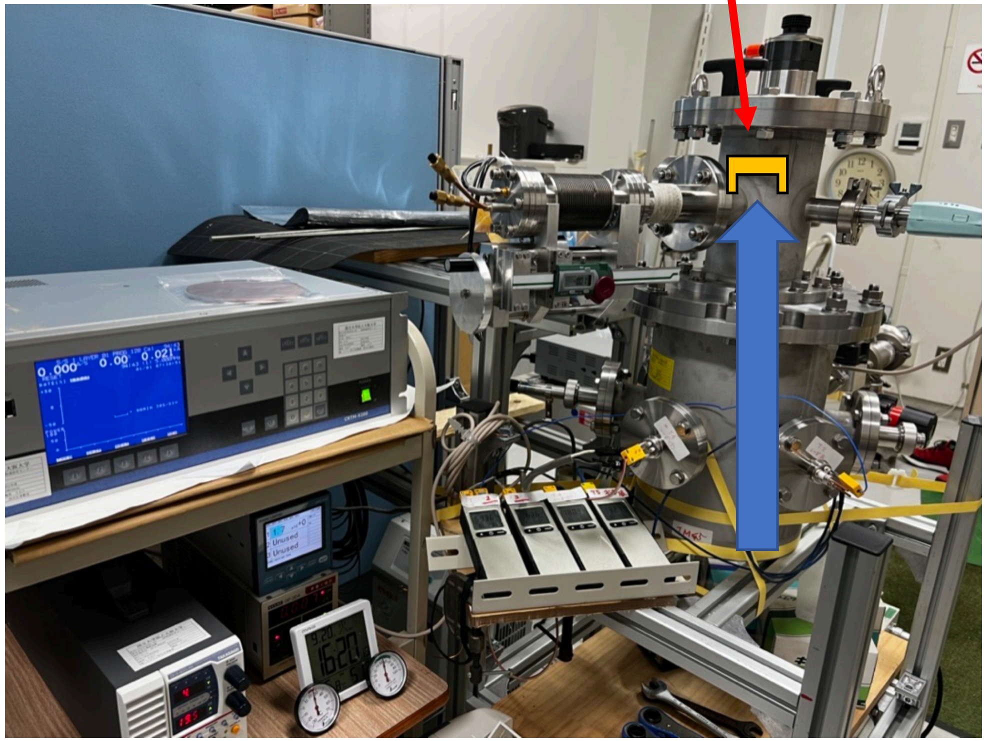




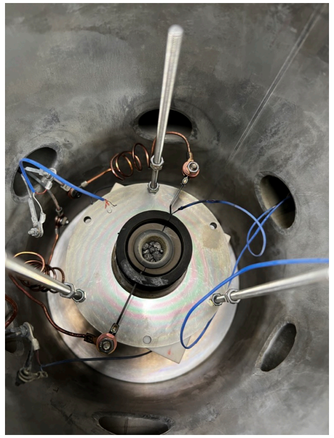




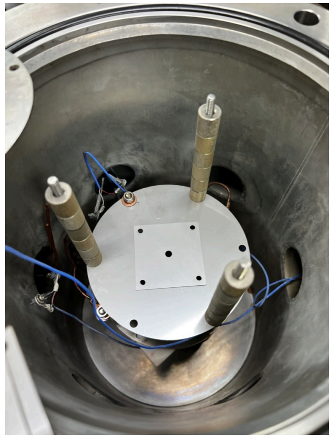
50 mm moving range, 1 mm/step



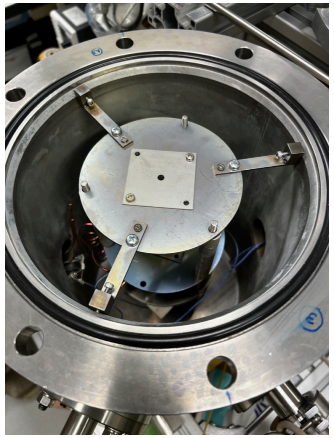
Current experimental setup



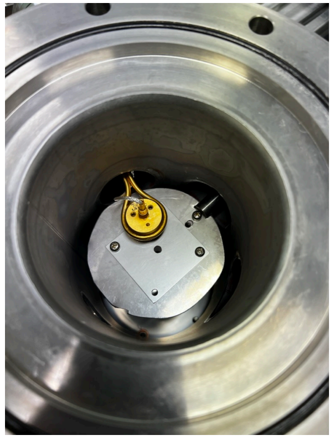
Crucible, ~1g Ca



#1Slit



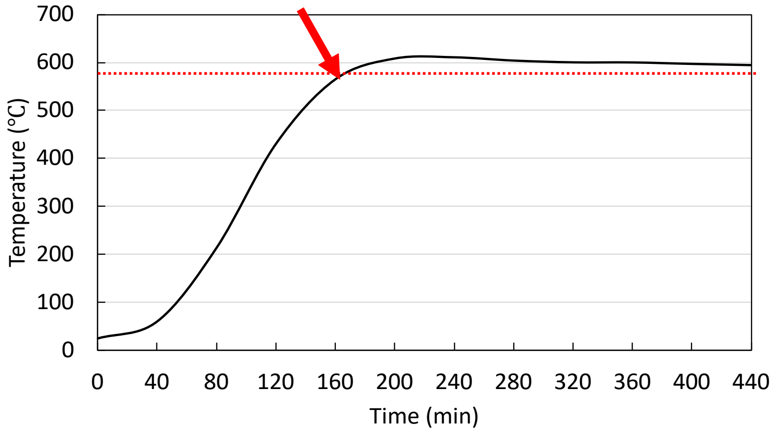
#2Slit

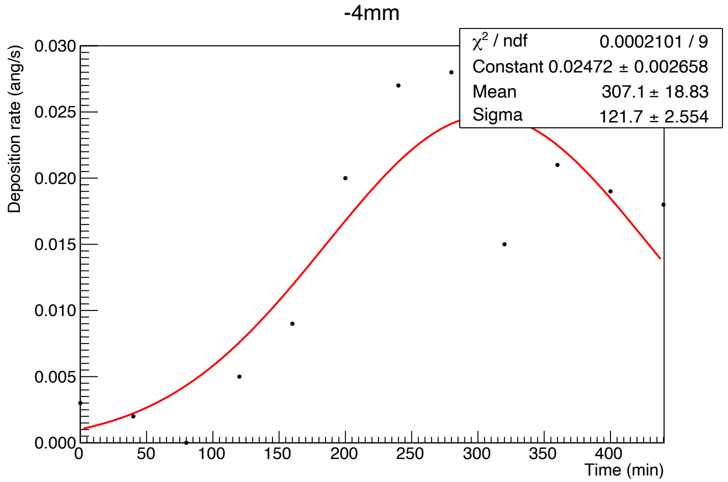


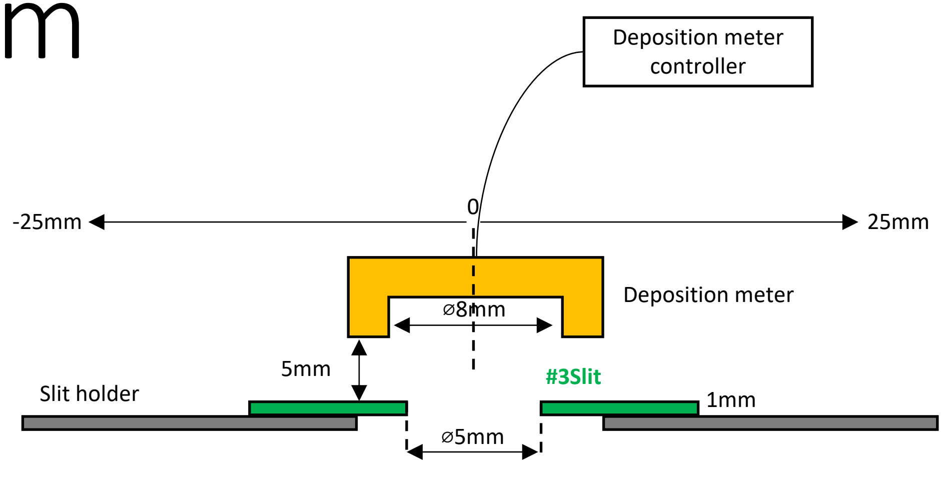
#3Slit

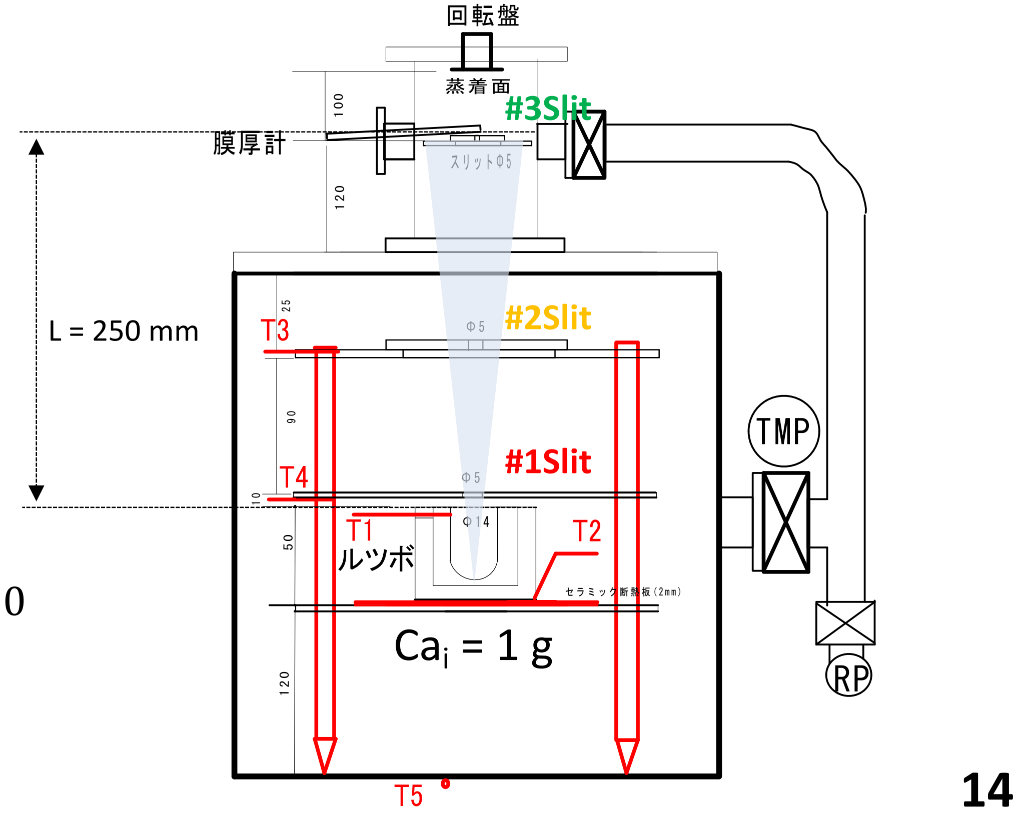
Development of production system

Calcium starts to evaporate
~580 °C at 1 Pa

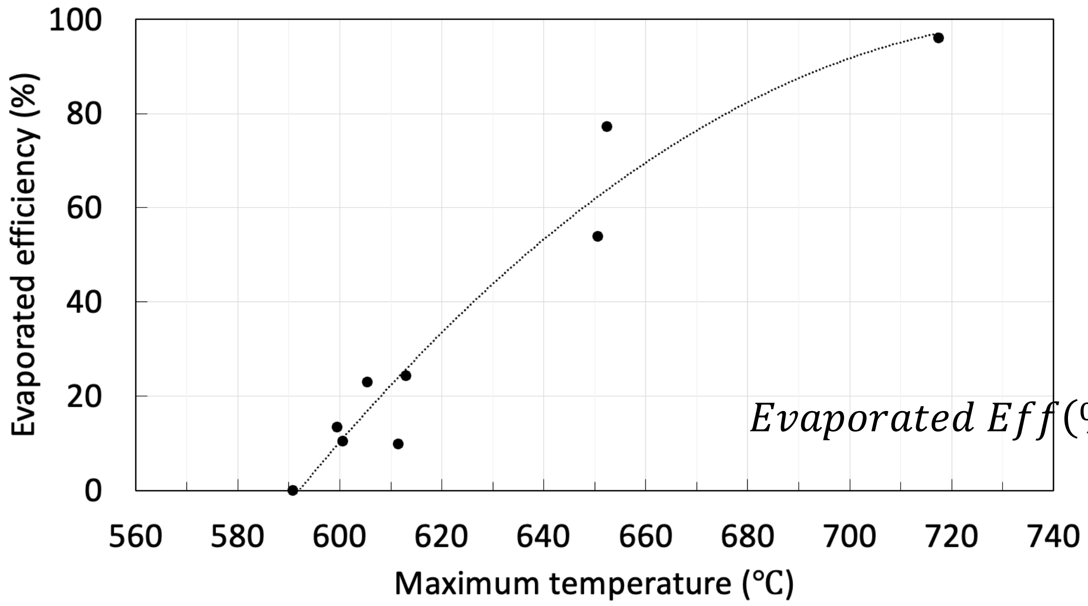








Temperature vs Evaporated Ca

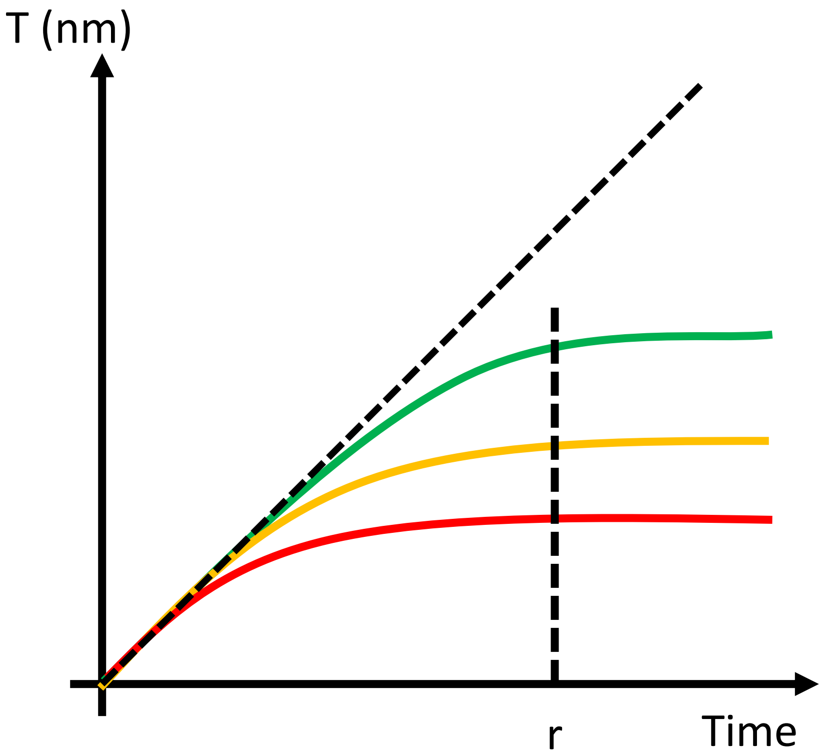


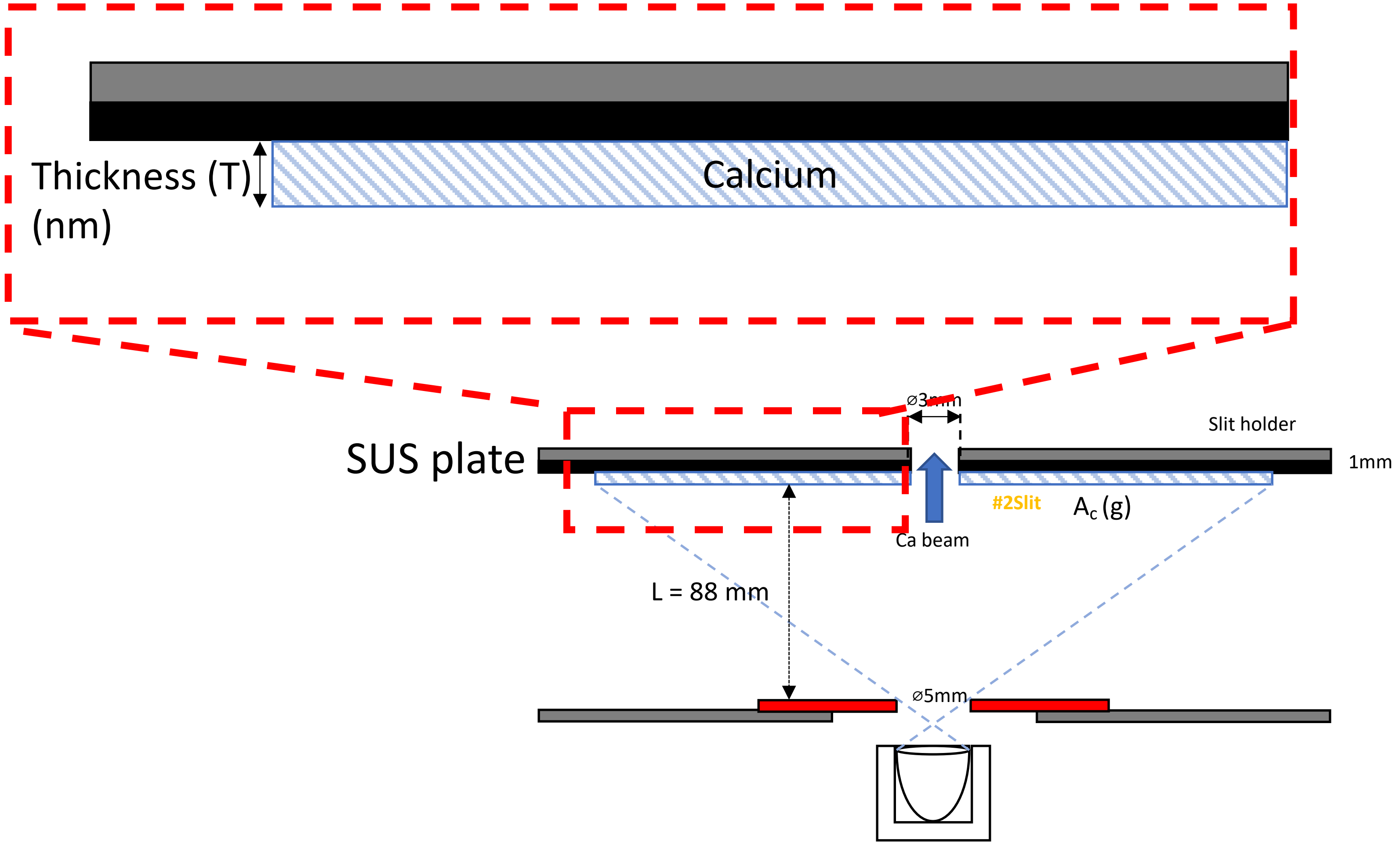
$$Evaporated\ Eff(\%) = \frac{Ca_i - Ca_f}{Ca_i} \times 100$$

Development of production system

Thin layer analysis method

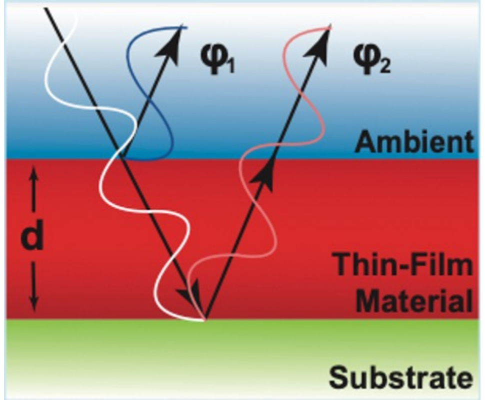
- Optical method ✅
- SEM-EDS ?
- Micro XRF ?





- Find the most appropriate material exposure to the same condition of the Ca atomic beam.
- Find the maximum capacity of the collection material.

Thickness measurement (optical method)



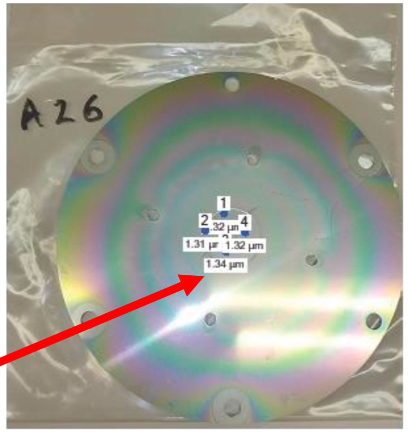
⌀120 mm SUS304 Collection plate

(Max temperature = 700℃)

A26-1

Ave = 1323±14 nm

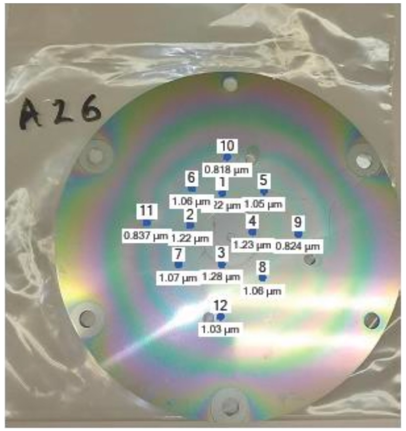
4 positions



A26-2

Ave = 1058±163 nm

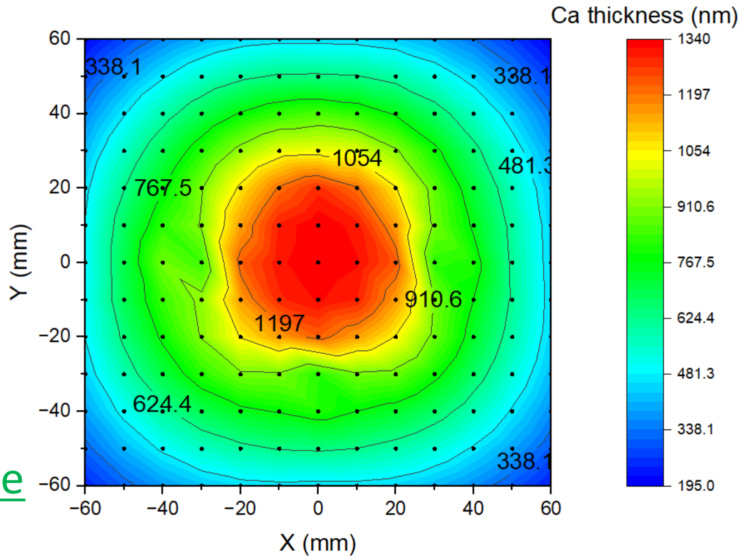
12 positions

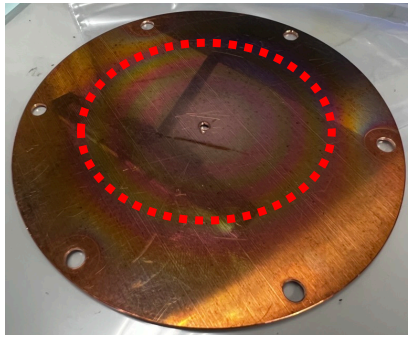


A26-3

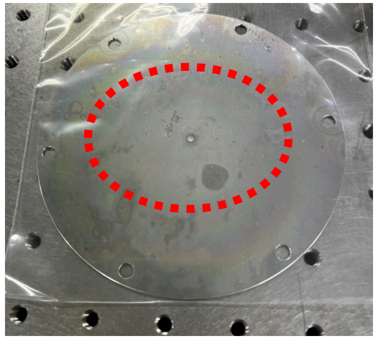
Ave = 650±164 nm

12 positions





Cu



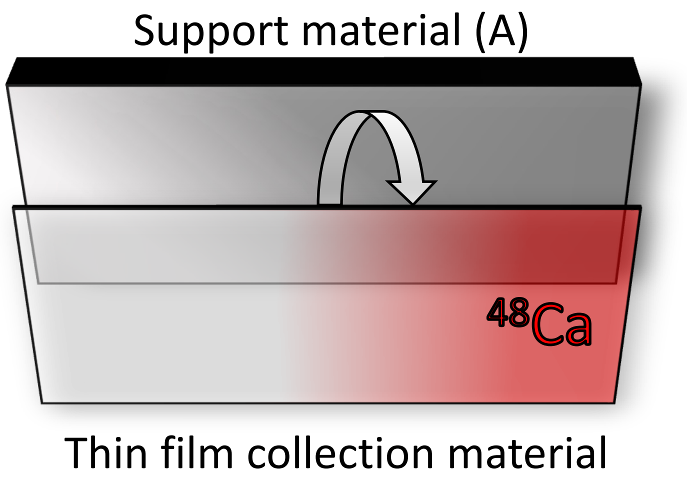
Al

- Consider the recovery method
  - Physical, chemical
  - Low radioactive contamination

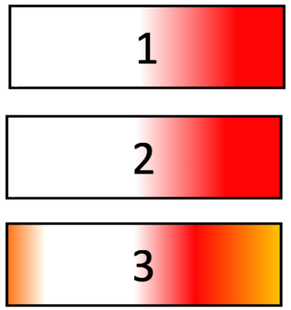
Development of production system

Important parameters

- Deflection angle (θ)
- Travel distance (L)
- Surface area (A)
- Ca Isotope composititon
- Ca recovered



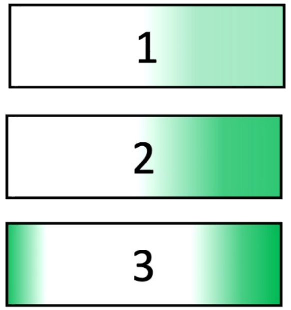
Ca isotope composition



1, 2 → Higher ^{48}Ca/^{4x}Ca ✅

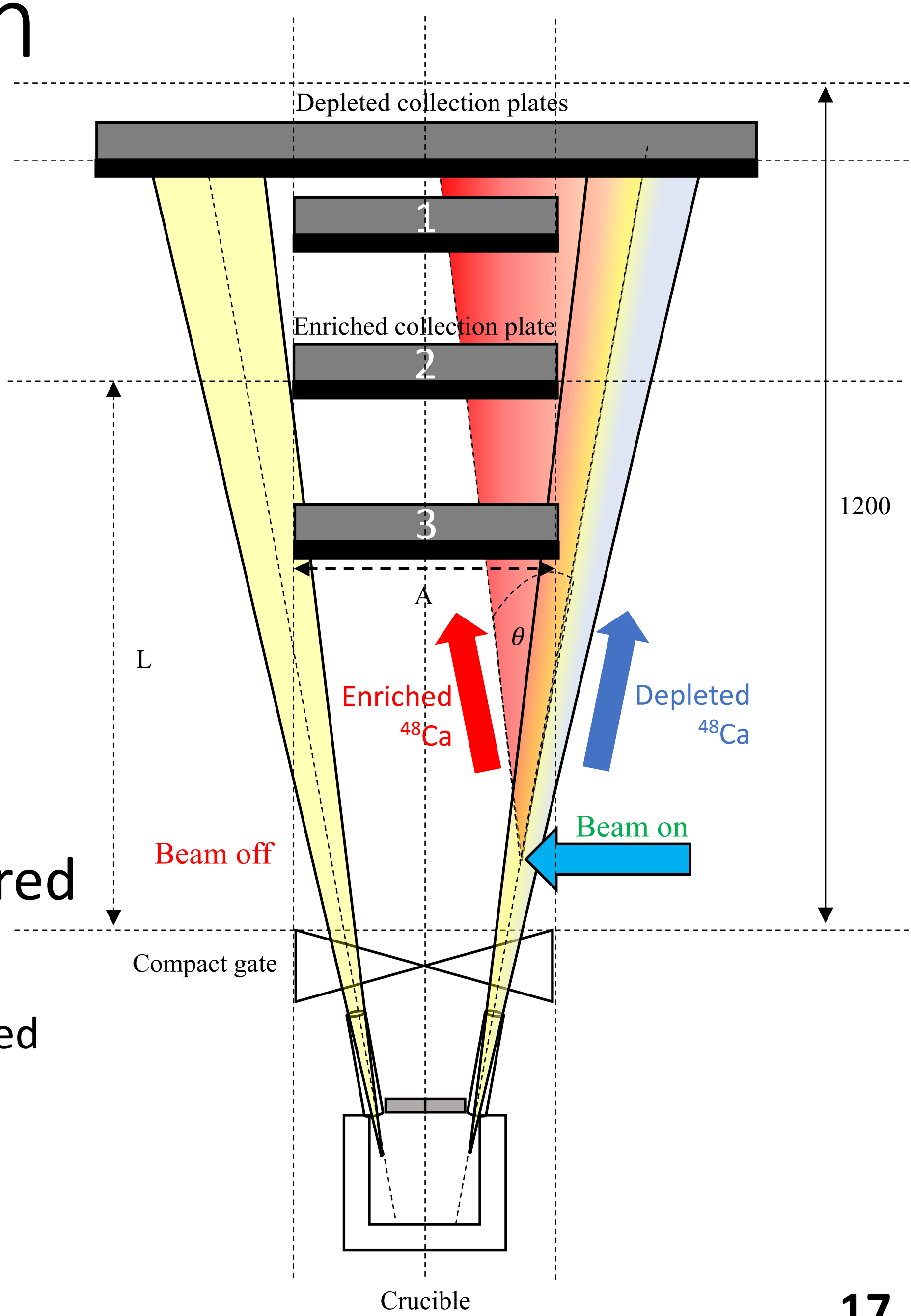
3 → Contaminated to original beam ❌

Ca Recovered



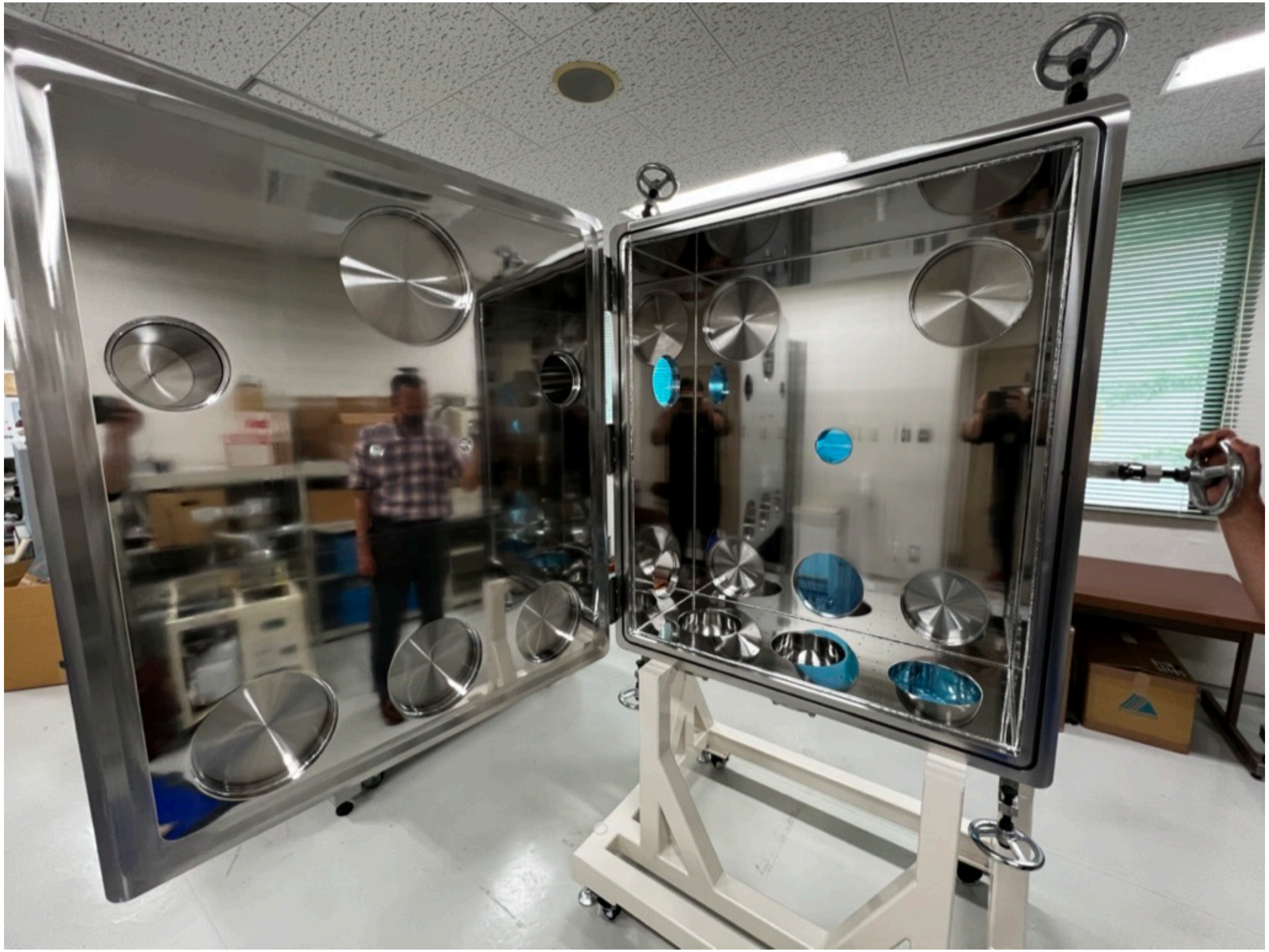
1, 2 → Lower recovered

3 → Highest recovered

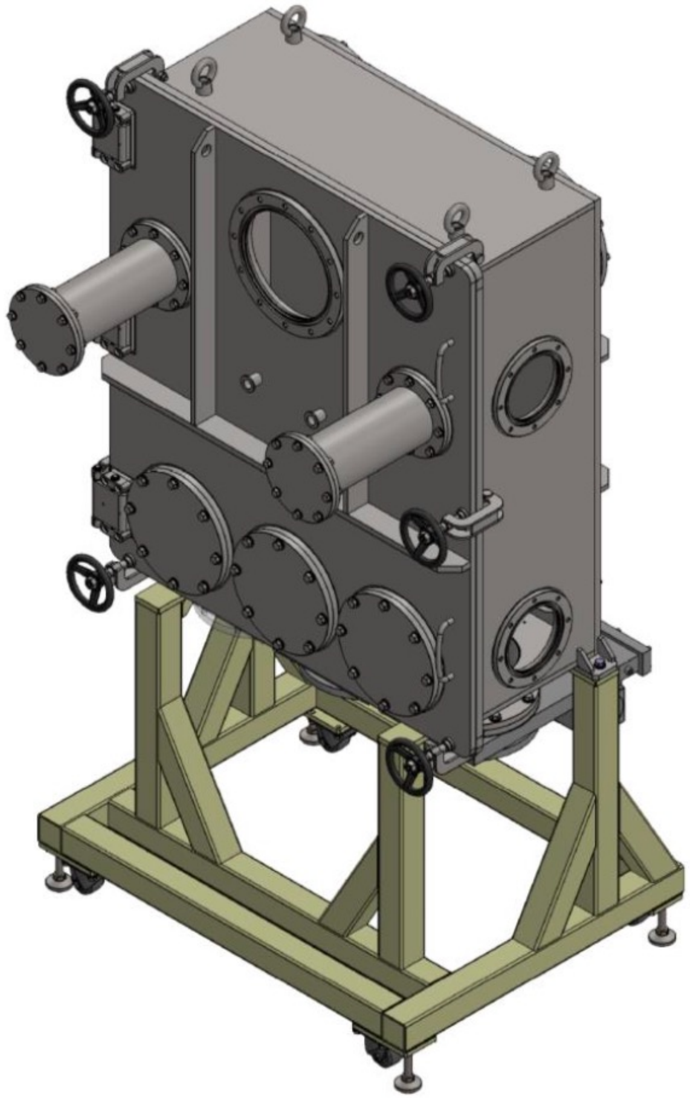


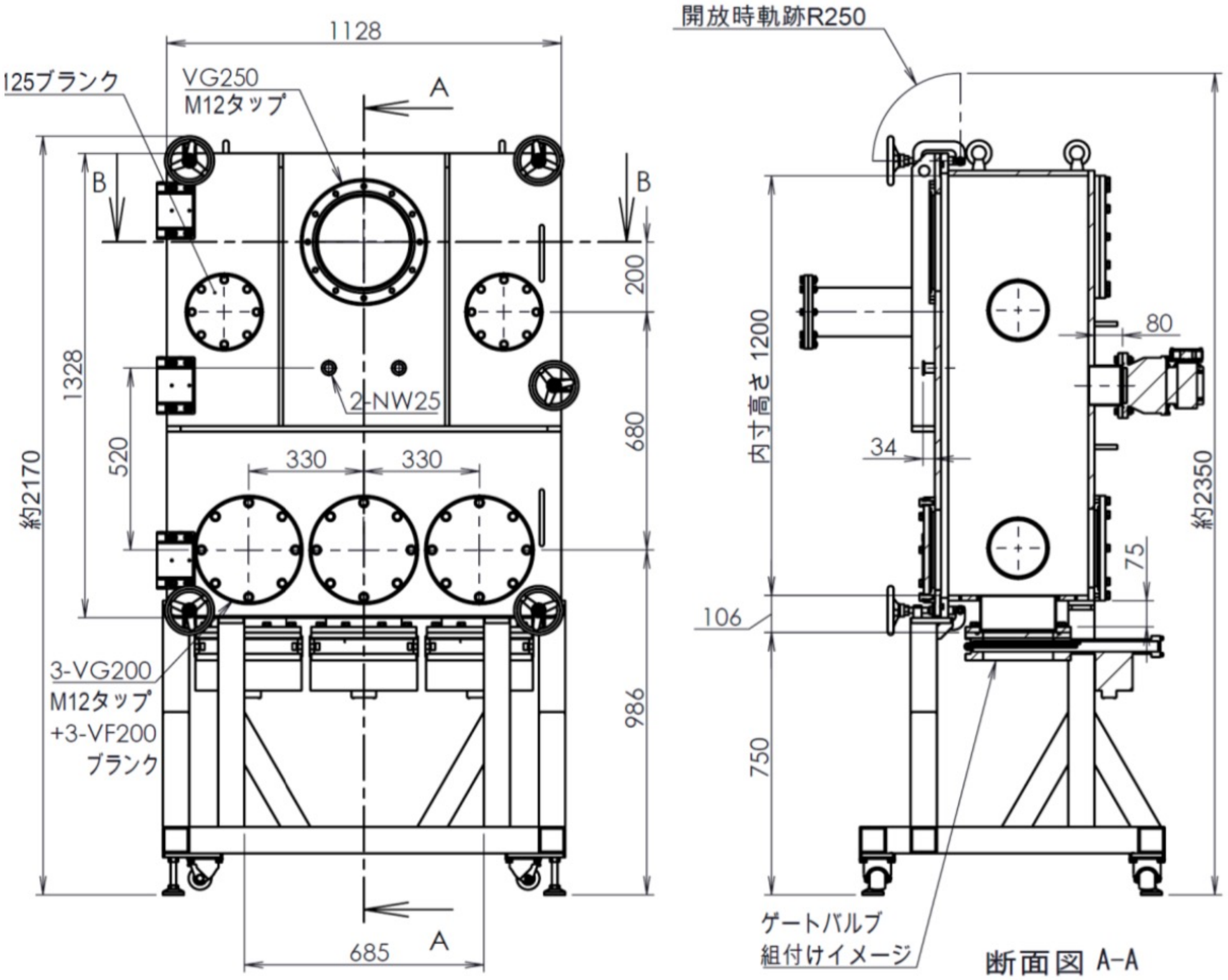
- If A is constant, L ∝ 1/^{48}Ca ∝ Recovered Ca
- Find optimal parameters for mass production

Newly design vacuum chamber

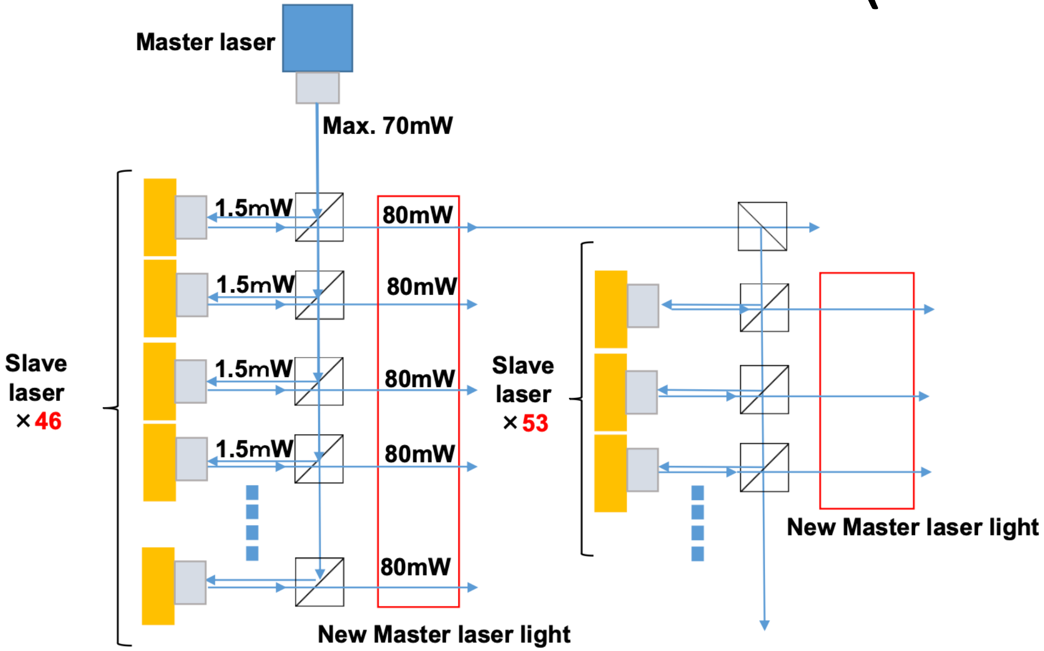


Newly design vacuum chamber

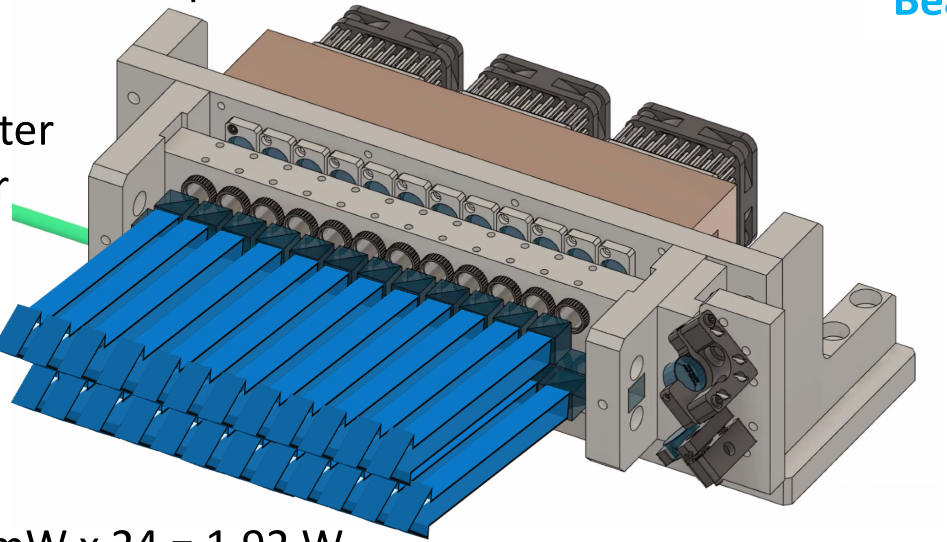




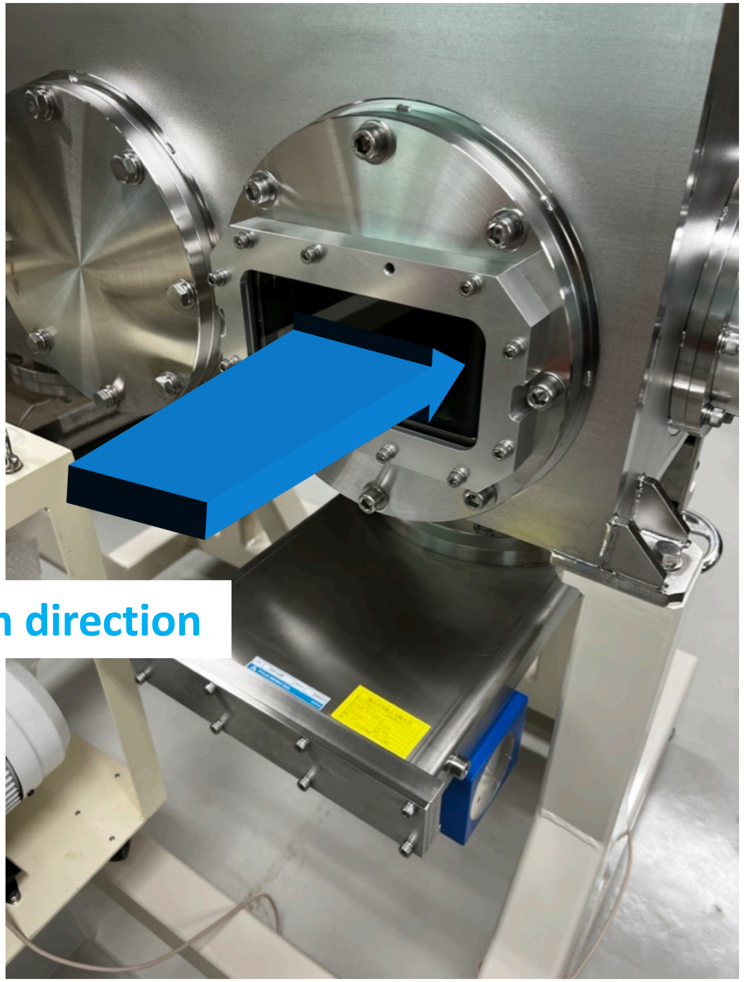
vacuum chamber (1 mol/year)



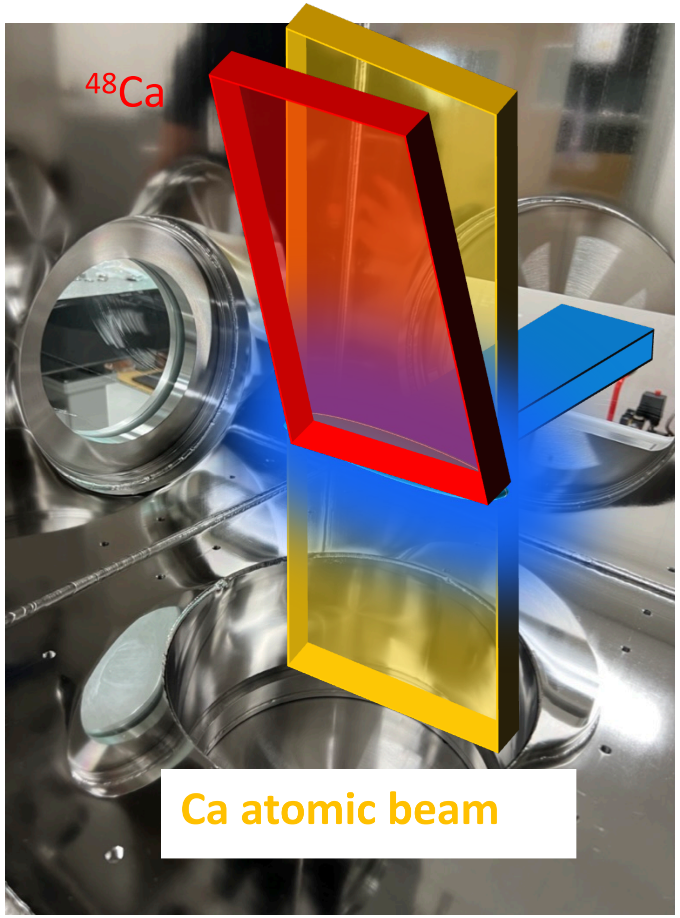
Beam spliter



80 mW x 24 = 1.92 W

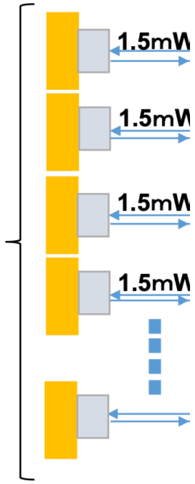


Inside

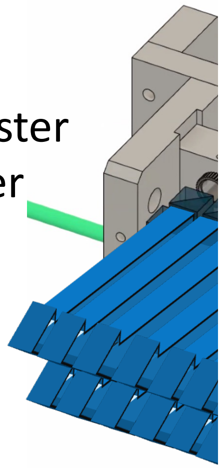


200 W can be realized by 2500 slave lasers and 1 master laser

vacuu



Beam sp



80 mW x 24 =

What is the required laser power?

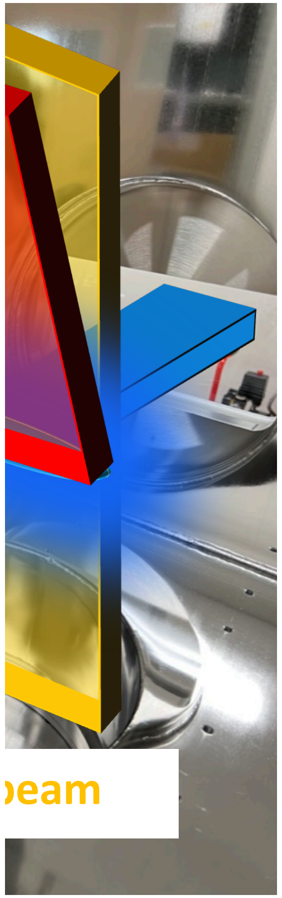
Photon energy: 4.7×10^{-19} J @ 423 nm

Number of photons absorbed by 1 atom: 1,000

→ Number of ^{48}Ca produced by 1 W laser: 2×10^{15} sec^{-1} → ~5 g/W/year

>200 W of laser power produces ^{48}Ca of 1 kg/year.

Current	FY2023	Near future	Future
100 mW →	2 W →	2 kW →	60 kW
	(~10 g / year)	(~10 kg / year)	(~300 kg / year)



ster laser

Future developments

FY2022, Preparation

- TOF assemble
- Vacuum stability of the main chamber✅
- Monitor and control system development✅
- Collection material study (in progress)

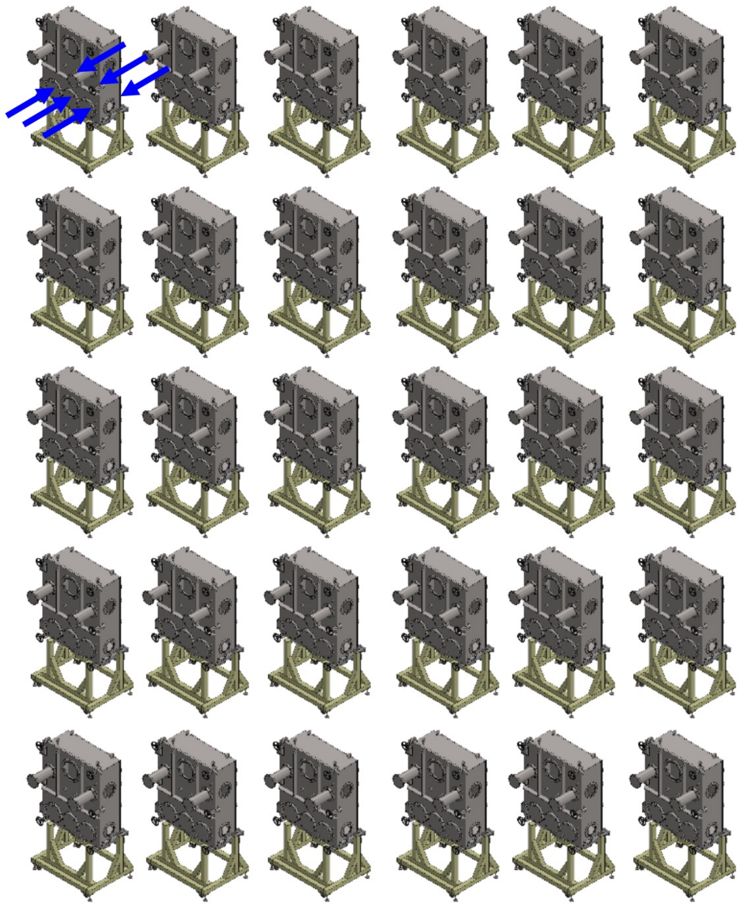
FY2023, Installation, and investigation

- First crucible installation
- First laser installation (EC-LD + FP-LD, SOA)
- Investigation of ^{48}Ca isotope separation
- Collection and recovery system installation
- Monitor and control system improvement
- Improvement and identification of problems during the long time operation

FY2024, Production

- Fully operation of 6 ports
- Scale up the mass production + automation system
- Multiple chambers

300 kg/years production plan



Vacuum chamber (30 units
6 ports/chamber
180 laser units

Power/LD => 1 W
Optical power => 1.7 kW/port
Number of LDs/port = 1700
Total optical power:
51 kW/unit -> ~300kW/30 units

Summary

- LIS for 48Ca is developed to find the cost-effective manner for large-scale production toward the study of 0νββ by CANDLES.
- Development of the LIS
  - Atomic beam system (Univ. Fukui)
    - Collimator and slit effect, simulation
  - Power scalable laser system (ILE, ILT)
    - FP-LD + PD-LDs, SOAs
  - Collection and recovery (RCNP)
    - Collection material study
    - Physical or chemical recovery method
  - Large-scale production system (RCNP)
  - Monitor and control systems (RCNP)
- The 1st milestone is the production rate of 1 mol/year by FY2023.