NEWS

Neutrinos Electro-Weak interactions and Symmetries

Candles

RCNP

Development of Laser Isotope Separation (LIS) System for ^{48}Ca Toward the Study of Neutrinoless Double Beta Decay of CANDLES

A. Rittirong[1*], S. Umehara[1], K. Matsuoka[1], Y. Minami[1], T. Shima[1], S. Yoshida[2], I. Ogawa[3], S. Tokita[4], H. Okuda[5], H. Niki[1,3], M. Uemukai[6], and N. Miyanaga[5,7]

[1]RCNP, Osaka Univ., [2]Dep. Sci., Osaka Univ., [3]Dep. Eng., Univ. Fukui, [4]ICR, Kyoto Univ., [5]ILE, Osaka Univ., [6]Dep. Eng. Osaka Univ., [7]Inst. Laser Tech.

anawat@rcnp.osaka-u.ac.jp

March 22nd, 2024

Research strategies and requirements

University of Fukui

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

Atomic beam system
- Small scale chamber
- Collimator effect
- Sheet-like atomic beam

ICR, Kyoto Univ., LIE, and LIT

Laser system
- Single frequency laser
- Power-scalable laser
- InGaN tunable + Tapered SOA
- Long-term operation
- Stable laser system

Research Center for Nuclear Physics (RCNP)

Collection system
- Collection plate
- Recovery system

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

RCNP, Osaka U + ICRR, Tokyo U.

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

Candles

Future development
- Stable operation 1/6 port
- Increase the production rate by multiple 6 port units
- Ton scale production
- LISSE

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

Particle identical to its antiparticle
E. Majorana, 1937

Dirac

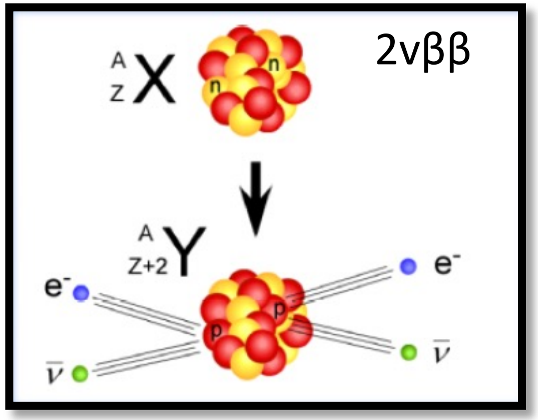
$$\mathcal{L}_D = -m_D \overline{\nu_R^0} \nu_L^0 + \text{h. c.}$$

Majorana

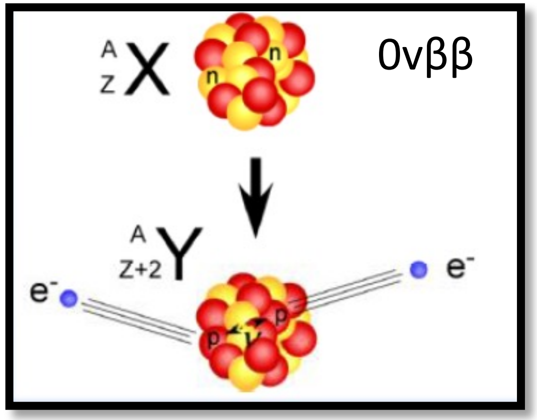
$$\mathcal{L}_{m_L} = -\frac{m_L}{2} \overline{(\nu_L^0)^c} \nu_L^0 + \text{h. c.}$$

Majorana particle

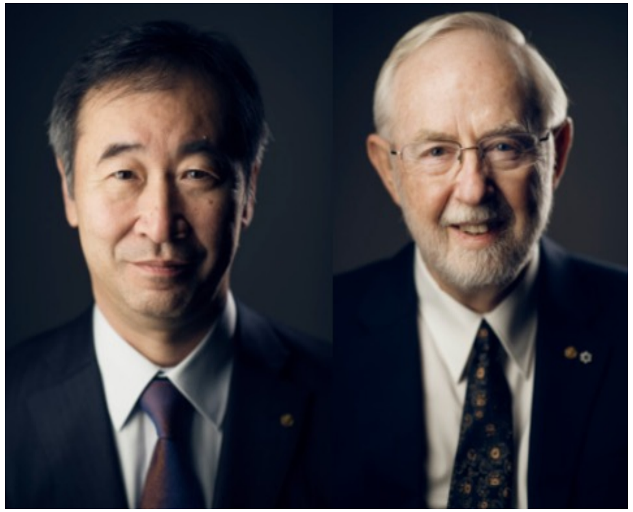
- Violates the Lepton number conservation law
- Particle <-> Anti-particle
- Matter dominated universe



2νββ
Within standard model



0νββ
Beyond the standard model !!



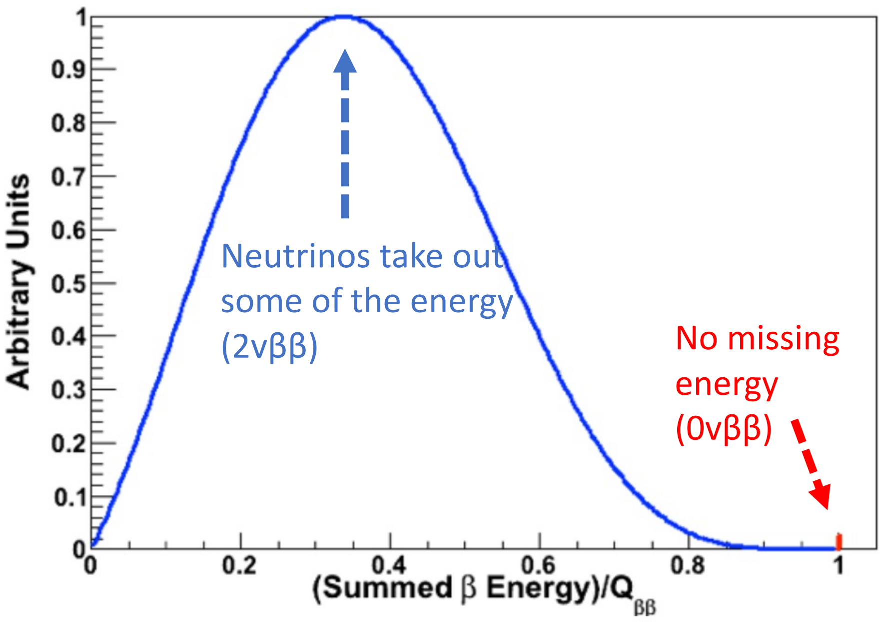
The 2015 Nobel Prize in Physics
Prof. Takaaki Kajita (ICRR)
Prof. Arthur B. McDonald (SNO)

Neutrino oscillation

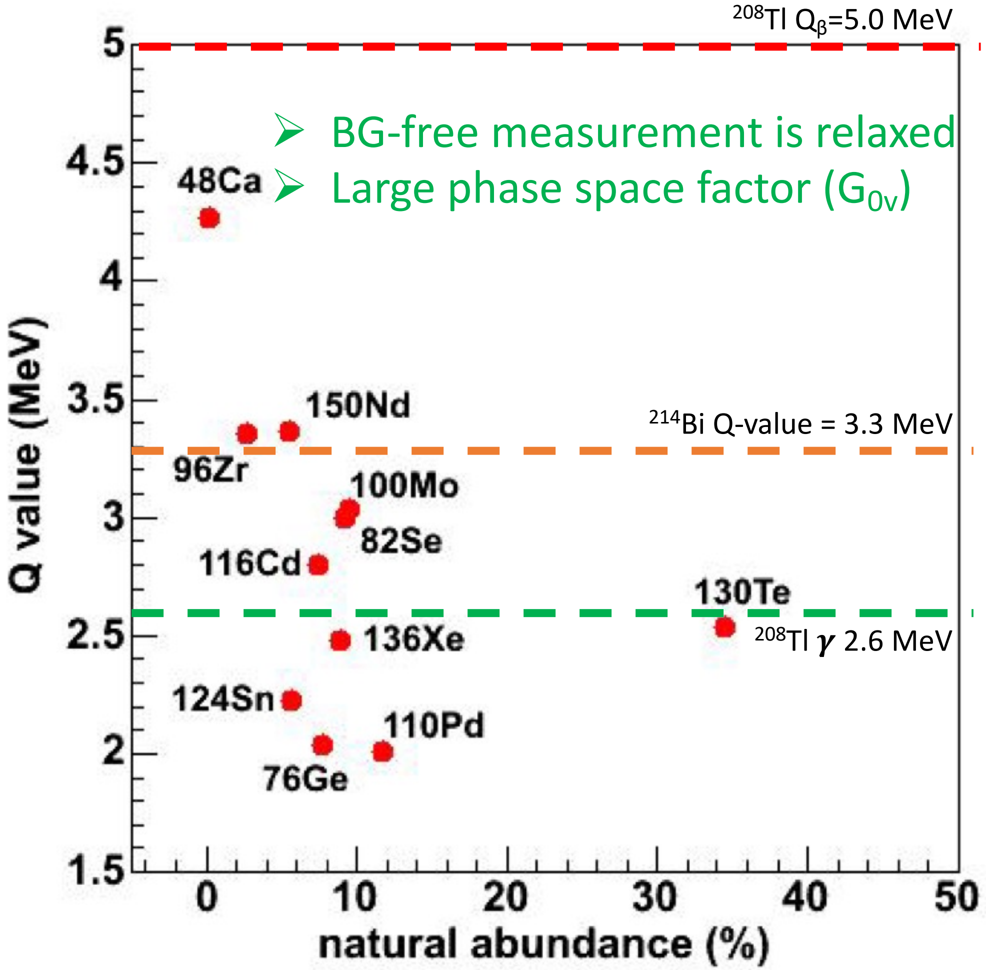
- Prove that neutrino has mass (m_ν)

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

Candles



Why ^{48}Ca?? => 4.27 MeV



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

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

$\langle m_{\beta\beta}\rangle$: Majorana neutrino mass

$T_{1/2}^{0\nu\beta\beta}$: Half-life

$G_{0\nu}$: Phase space factor

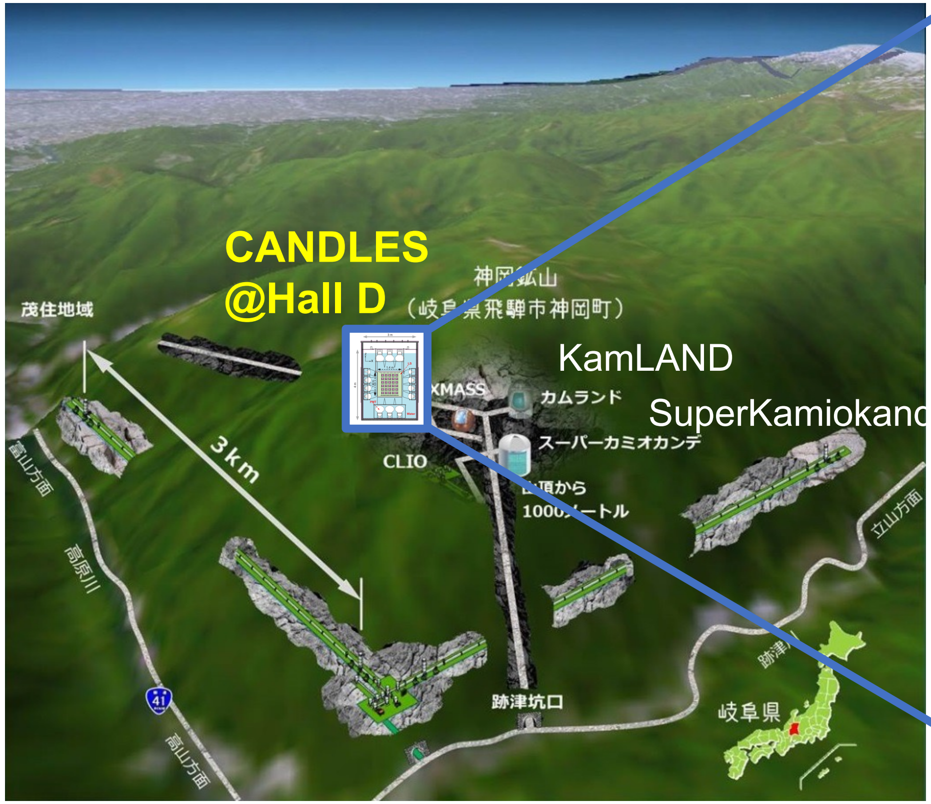
$M_{0\nu}$: Nuclear matrix element

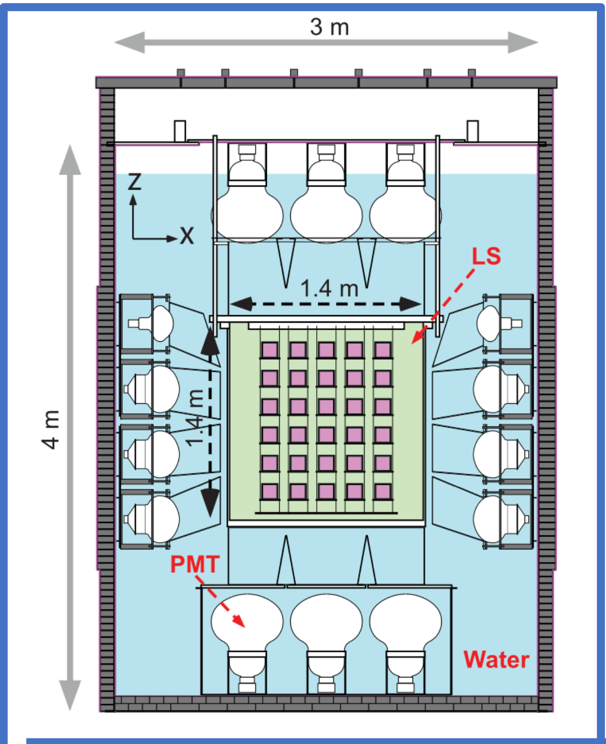
Requirement for 0νββ

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

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

Candles



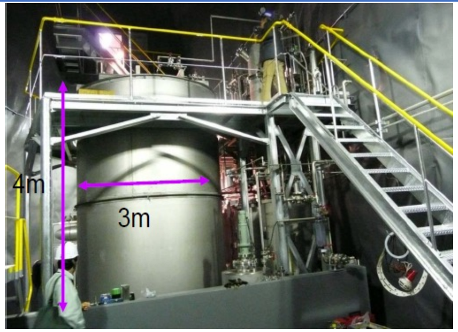


S. Ajimura, et al. *Physical Review D* 103.9 (2021): 092008.

- Liquid scintillator (LS)
  - 4π active shield(2 m^3)
- 62 Large photomultiplier tube
- Shielding system
  - Pb : 10-12cm
  - B_4C sheet : 5mm

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

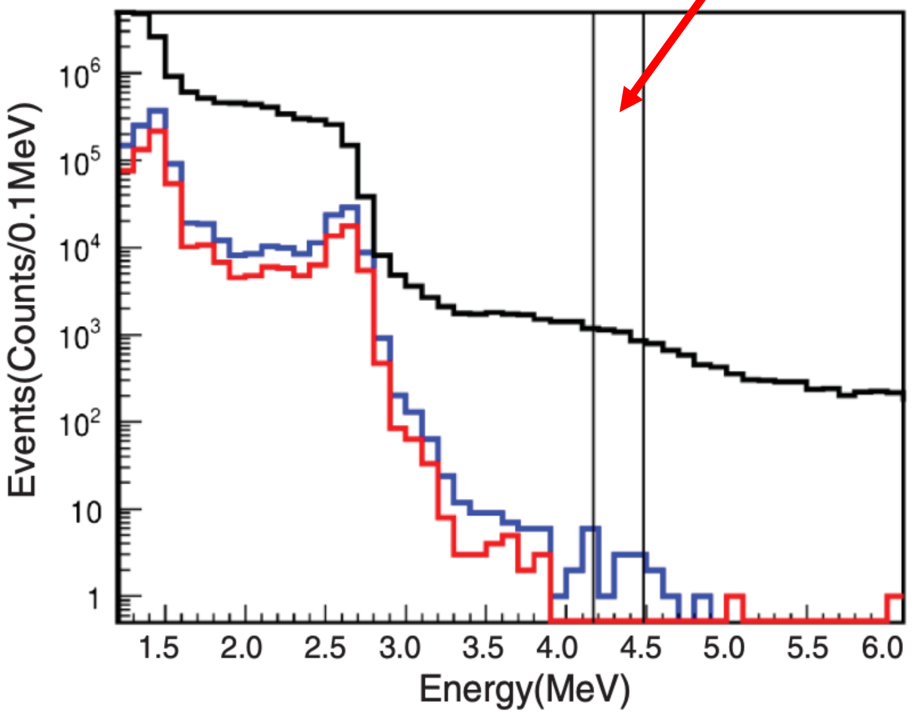
CANDLES III – 305 kg **CaF_2 Crystals** (3.2 kg × 96 pieces) ~ **0.35 kg ^{48}Ca**



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

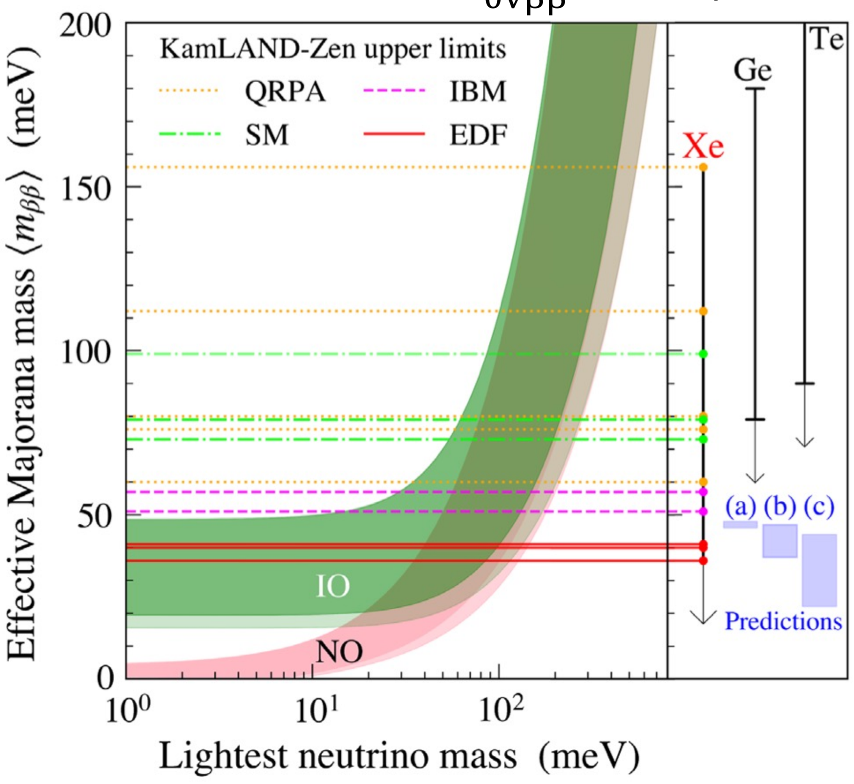
Candles

^{48}Ca ROI 4.17 – 4.48 MeV



KamLAND-Zen 400 & 800 (^{136}Xe)

36 – 156 meV, $T^{1/2}_{0\nu\beta\beta} > 10^{26}$ yr



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

To explore the IH region

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

BG free measurement

10 years of exposure time

0.8 Detection efficiency

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

CANDLES III – ^{nat}Ca

$$T^{1/2}_{0\nu\beta\beta}\ ^{48}Ca = 5.6\times10^{22} \text{ years}$$

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

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

^{48}Ca isotope enrichment is the key!!

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

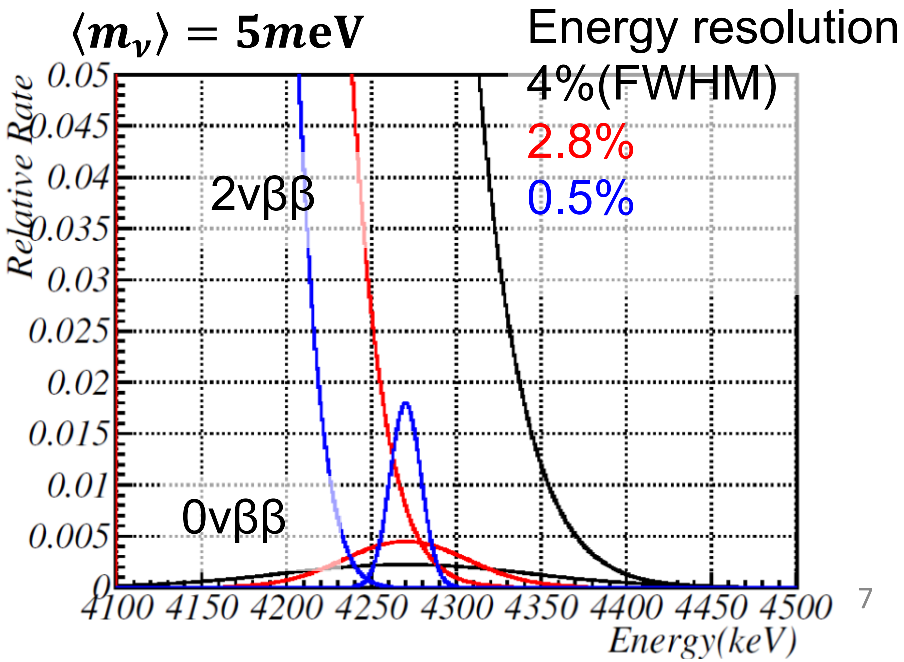
Candles

Future CANDLES

	CANDLES III	Next detector system
^{48}Ca Abundance	0.187%	**50%**
^{48}Ca Weight	0.35 kg	600 kg ~
Energy Resolution	6%	1.0% (required)
$\langle m_\nu \rangle$ sensitivity	500meV	~5 meV
Feature	Cooling CaF2 Low BG	Enrichment of ^{48}Ca Scintillating bolometer

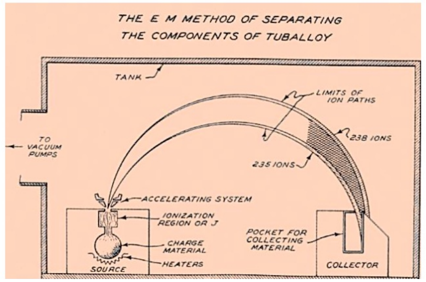
- Improvement of Analysis
  - $^{212}Bi^{212}Po$ rejection by CNN
  - ^{208}Tl rejection by likelihood analysis
  - Position distribution of ^{208}Tl

- A large amount of ^{48}Ca
  - For high sensitivity :
    - Increase by enrichment
- Higher energy resolution
  - To reduce $2\nu\beta\beta$ events

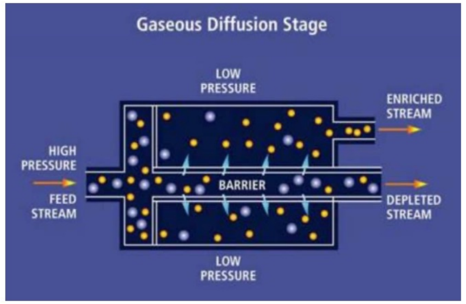


Calcium-48 [N.A. = 0.187%]

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

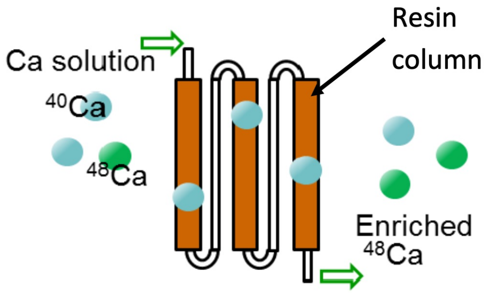


Electromagnetic

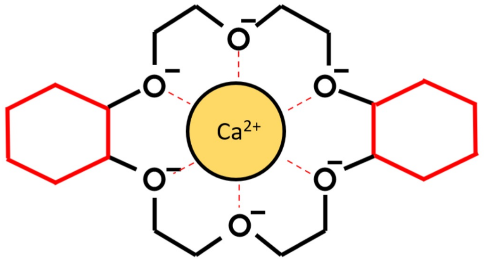


Gas diffusion

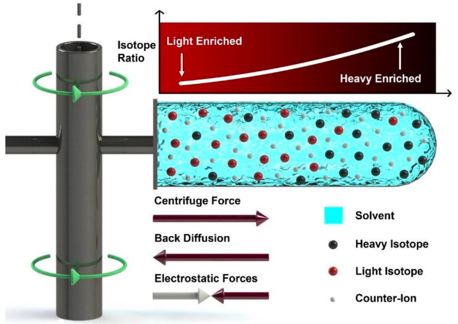
Isotope separation technique	Separation Coefficient	Production efficiency (y^{-1})	Cost	Limitation
Electromagnetic separator	High	Ten of grams	High	○ High power consumption ○ Low productivity
Liquid centrifuge	Middle	Kilograms	Low	○ Concentration limit
Industrial isotope separation • Gas Diffusion • 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	Middle	○ Development of the high-power laser, irradiation unit, and collection system



Chromatography



Chemical exchange



Liquid centrifuge

Research strategies and requirements

University of Fukui

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

Atomic beam system
- Small scale chamber
- Collimator effect
- Sheet-like atomic beam

Research Center for Nuclear Physics (RCNP)

Collection system
- Collection plate
- Recovery system

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

ICR, Kyoto Univ., LIE, and LIT

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

RCNP, Osaka U + ICRR, Tokyo U.

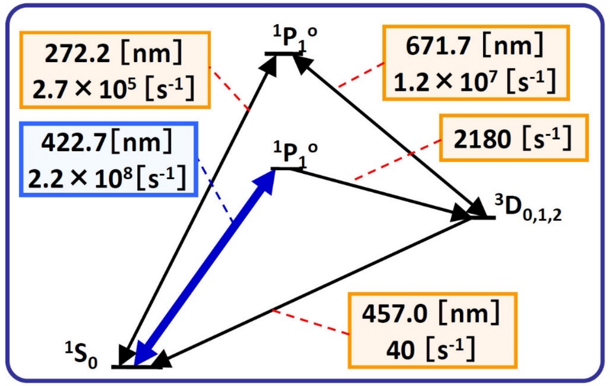
CAlcium fluoride for studies of Neutrino and Dark matters by Low Energy Spectrometer (CANDLES)

Candles

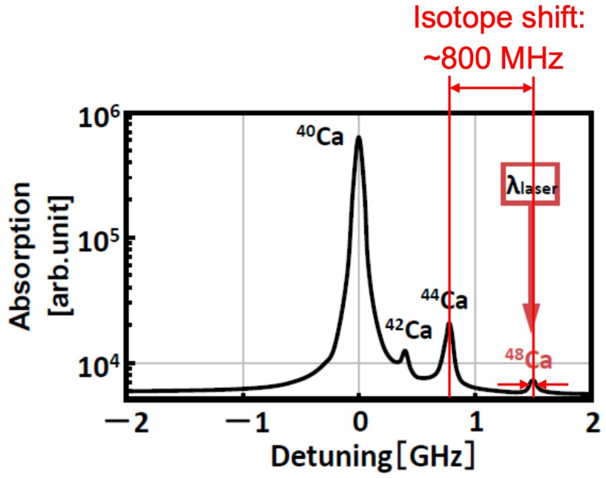
Future development
- Stable operation 1/6 port
- Increase the production rate by multiple 6 port units
- Ton scale production
- LISSE

Proof of principle for LIS

@University of Fukui, Japan



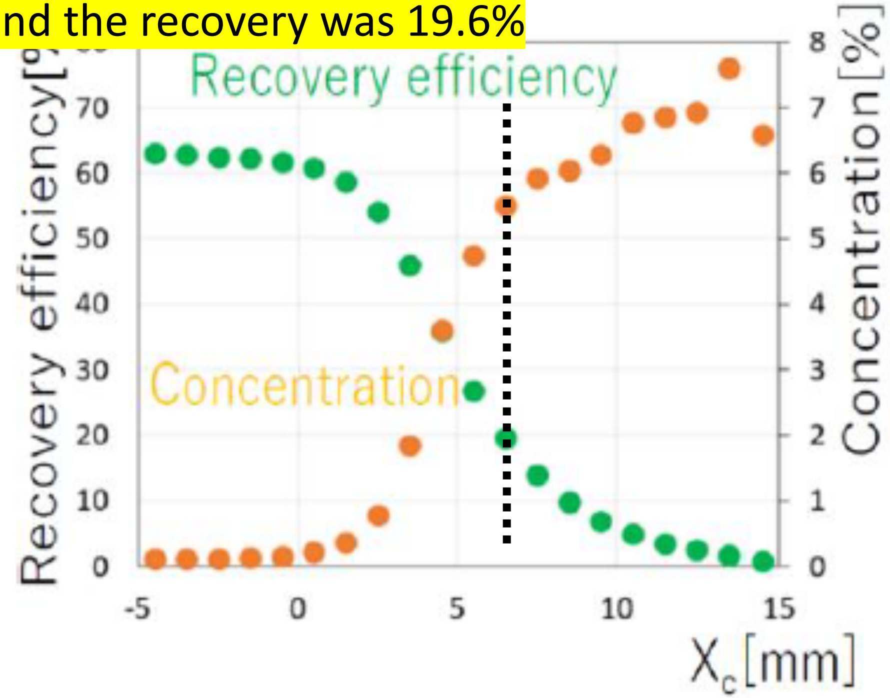
Isotope shift of Ca at 422.7 nm



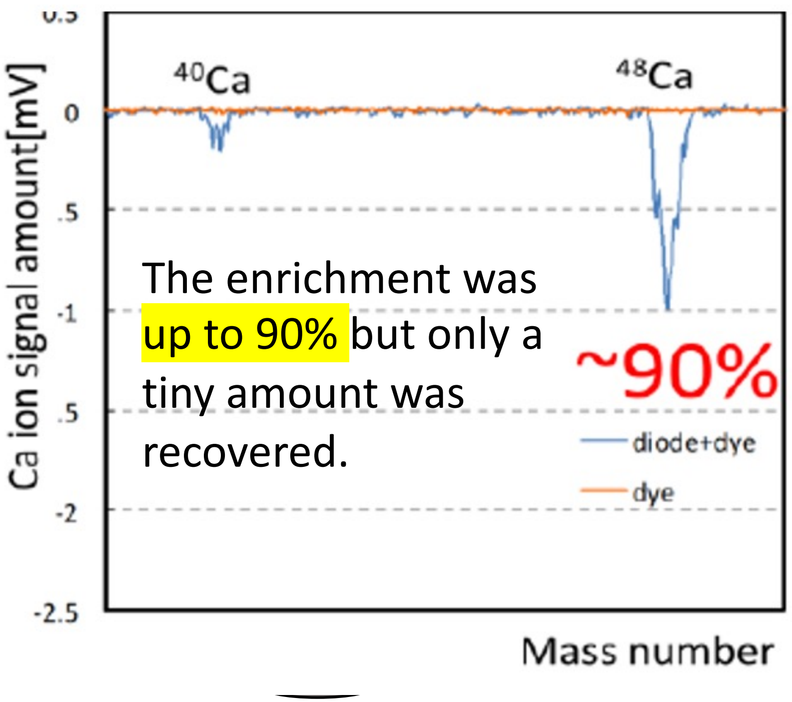
Absorption spectrum of Ca at 423nm

- 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

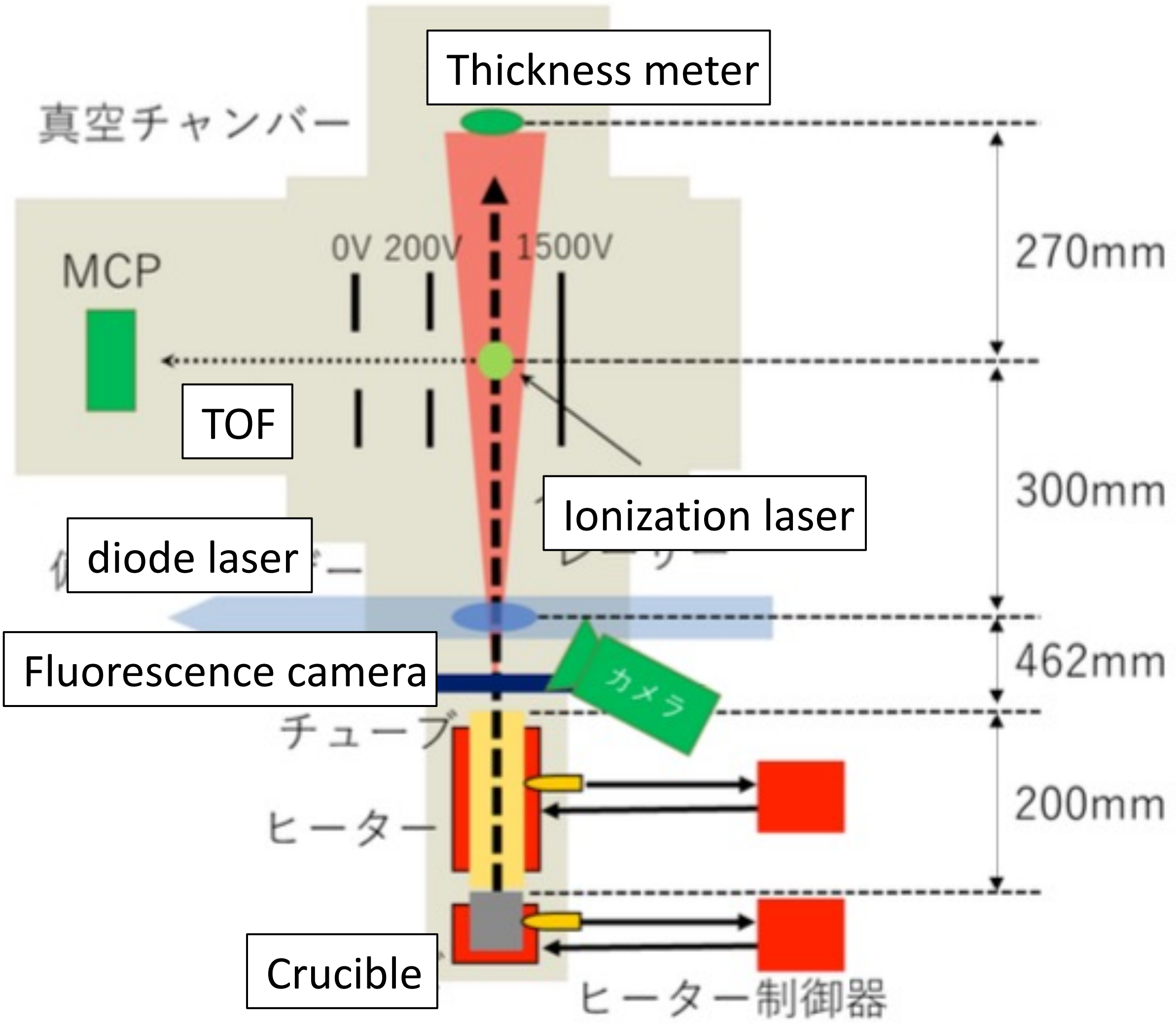
The DEFLECTION method was applicable for mass production

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

@University of Fukui, Japan

Development of atomic beam system

Atomic beam monitoring systems



How to treat the atomic beam

$$K_n = \frac{\lambda}{D}$$

Continue $K_n \sim 0$

Intermediate $(0.01 < K_n < 1)$

Molecular flow $K_n > 1$

⌀5 mm, L 200 mm tube (Previous)

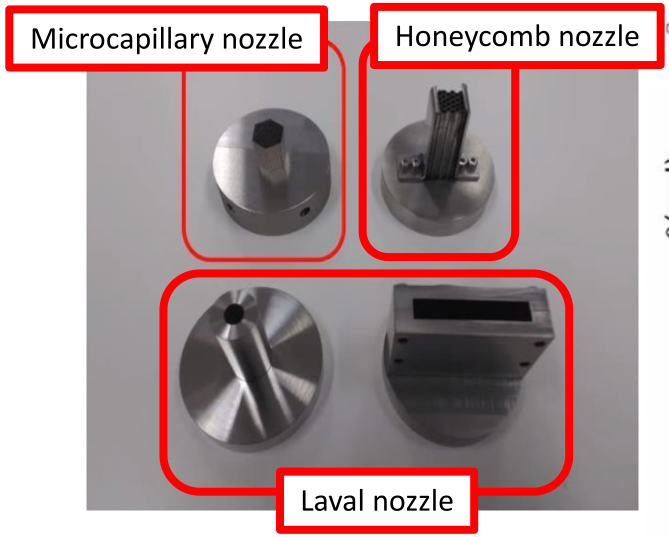
ϕ方向 $K_n = \frac{12.6\ \text{mm}}{5\ \text{mm}} = 2.52$

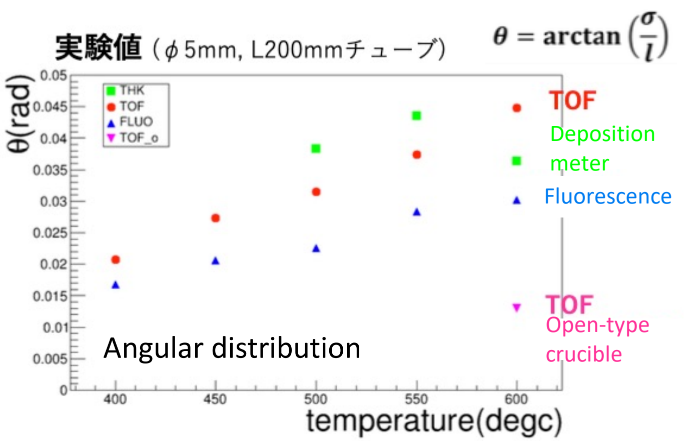
L方向 $K_n = \frac{12.6\ \text{mm}}{200\ \text{mm}} = 0.063$

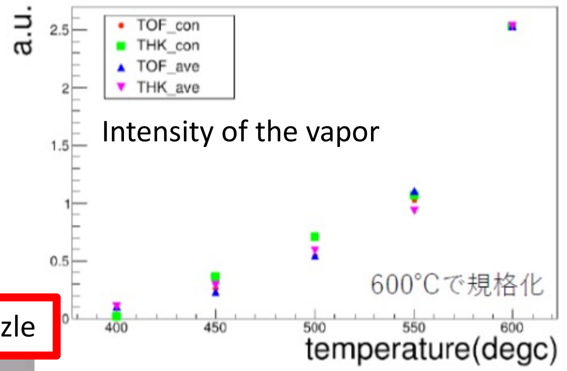
⌀0.1 mm, L 20 mm tube (Future)

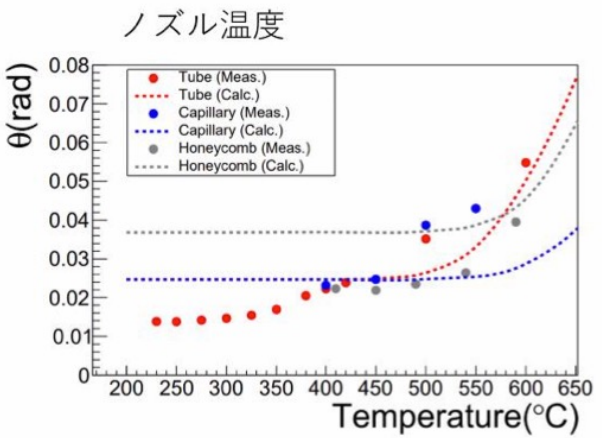
ϕ方向 $K_n = \frac{12.6\ \text{mm}}{0.1\ \text{mm}} = 126$

L方向 $K_n = \frac{12.6\ \text{mm}}{20\ \text{mm}} = 0.63$







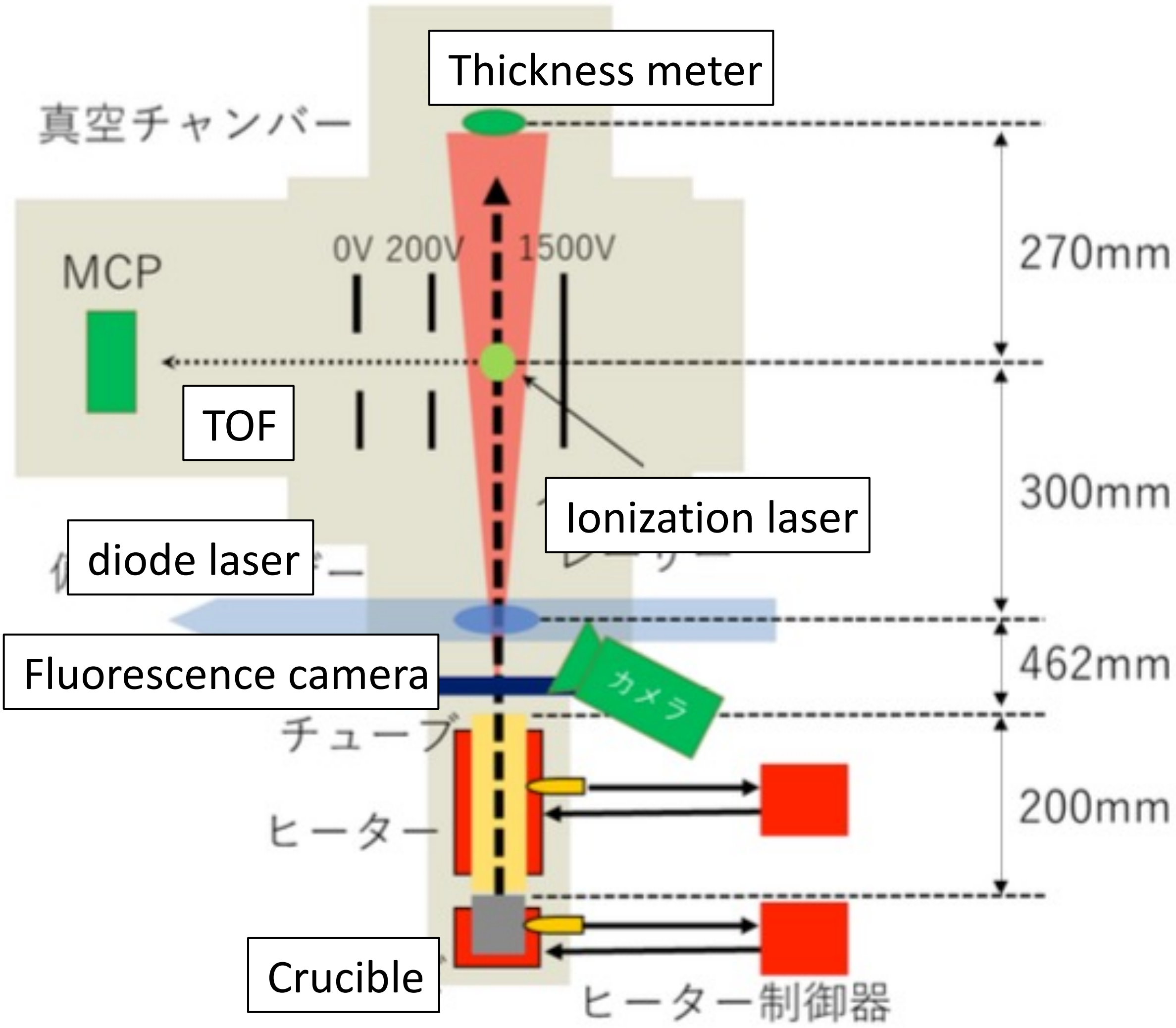


K_n increases -> smaller angular distribution, high vapor intensity

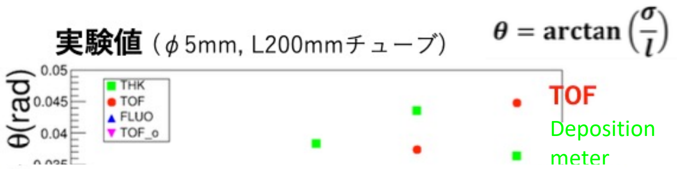
Development of atomic beam system

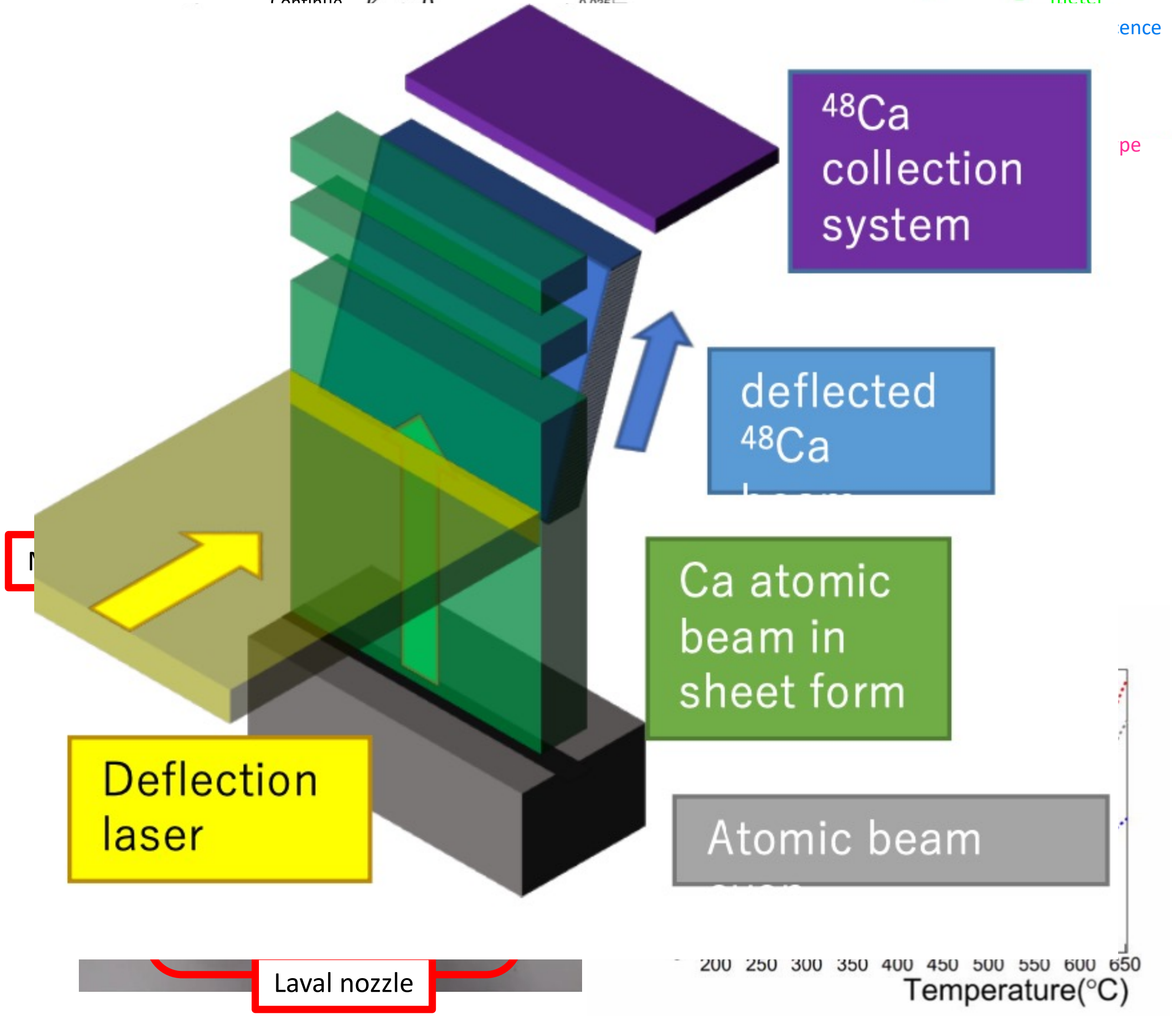
@University of Fukui, Japan

Atomic beam monitoring systems



How to treat the atomic beam





K_n increases -> smaller angular distribution, high vapor intensity

Research strategies and requirements

University of Fukui

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

Atomic beam system
- Small scale chamber
- Collimator effect
- Sheet-like atomic beam

Research Center for Nuclear Physics (RCNP)

Collection system
- Collection plate
- Recovery system

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

ICR, Kyoto Univ., LIE, and LIT

Laser system
- Single frequency laser
- Power-scalable laser
- InGaN tunable + Tapered SOA
- Long-term operation
- Stable laser system

RCNP, Osaka U + ICRR, Tokyo U.

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

Candles

Future development
- Stable operation 1/6 port
- Increase the production rate by multiple 6 port units
- Ton scale production
- LISSE

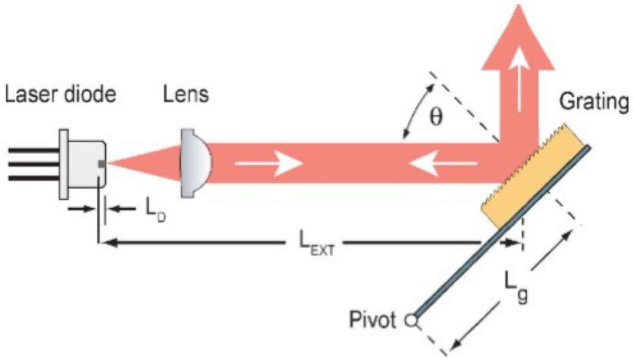
Development of the laser system

@ICR Kyoto Univ., ILE, and ILT

Laser performances required for isotope separation

- ✓ 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

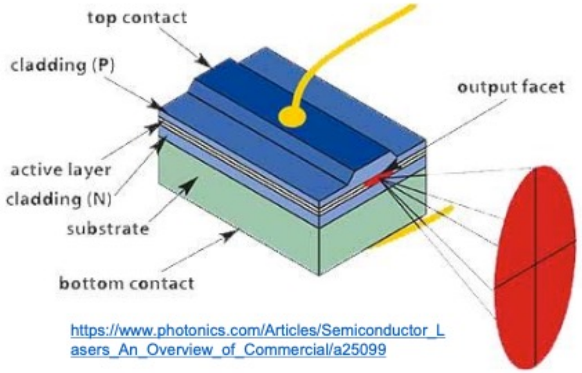
External cavity lasers



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

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

Fabry-Perot lasers



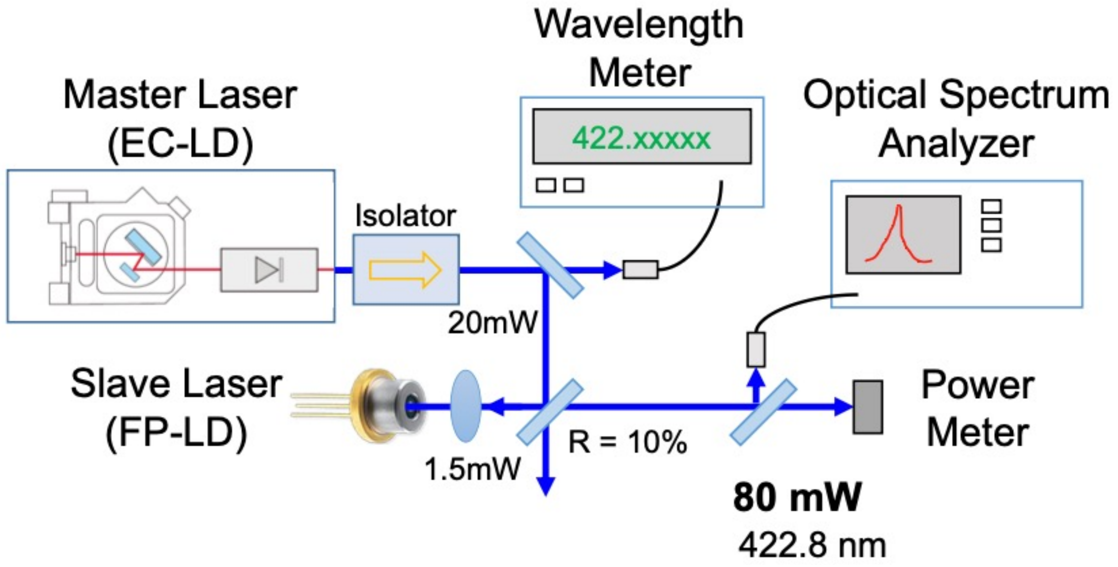
https://www.photonics.com/Articles/Semiconductor_Lasers_An_Overview_of_Commercial/a25099

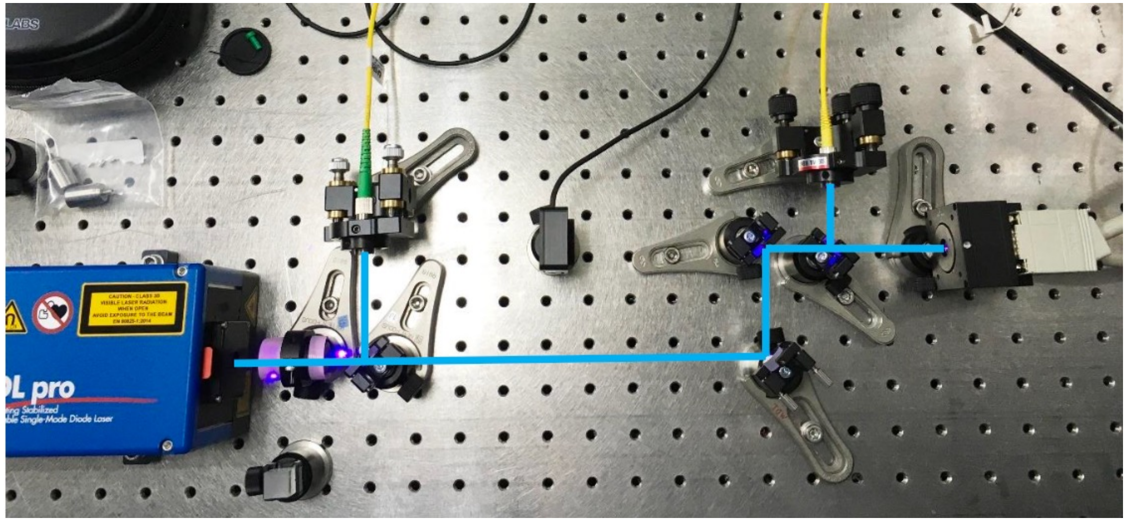
- Multi longitudinal-mode (Wide spectral width)
- Low cost
- Compact
- High efficiency

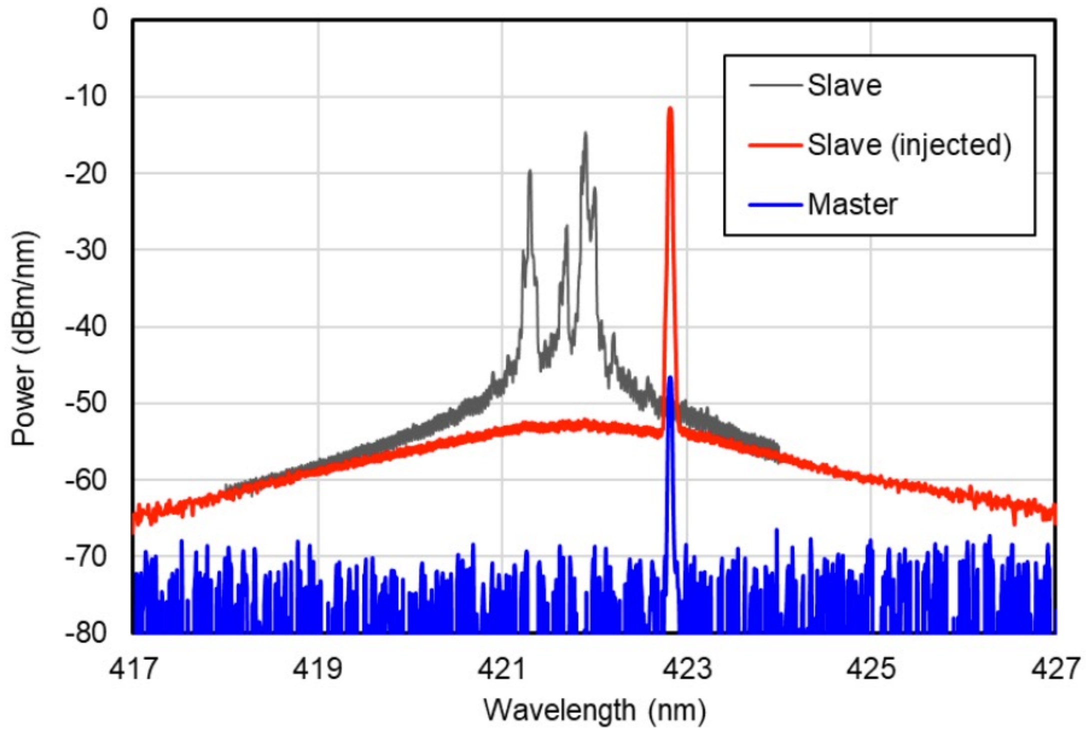
Development of the laser system

Ogawa *et al* 2022 *J. Phys.: Conf. Ser.* **2147** 012012
@ICR Kyoto Univ., ILE, and ILT

Experiment of injection locking







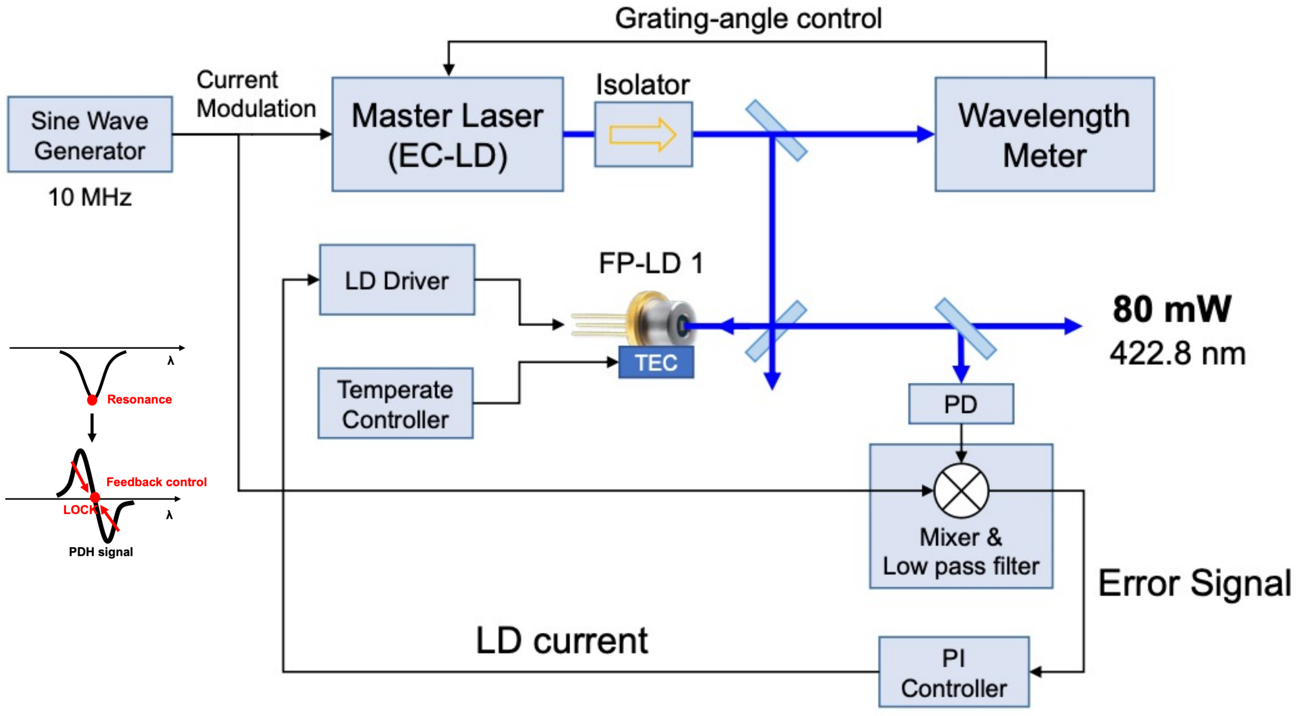
A single frequency of slave laser was obtained.

Development of the laser system

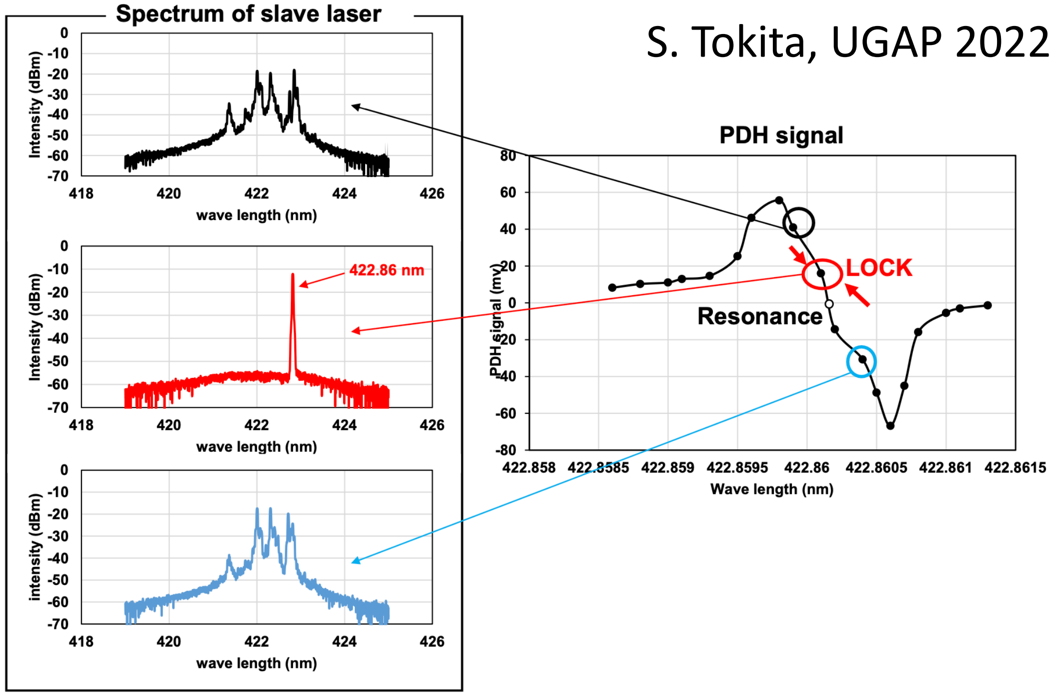
@ICR Kyoto Univ., ILE, and ILT

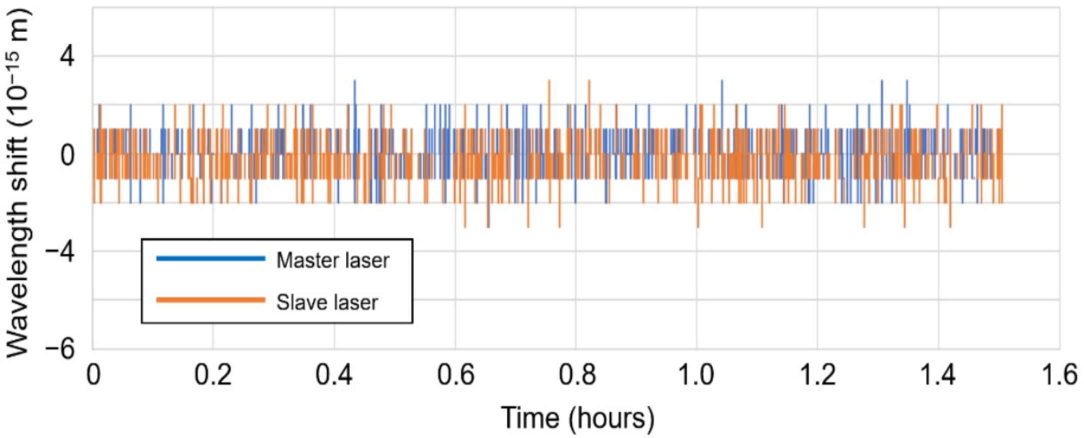
S. Tokita, UGAP 2022

Experiment of wavelength stabilization by using PDH method



Spectrum and PHD error signal





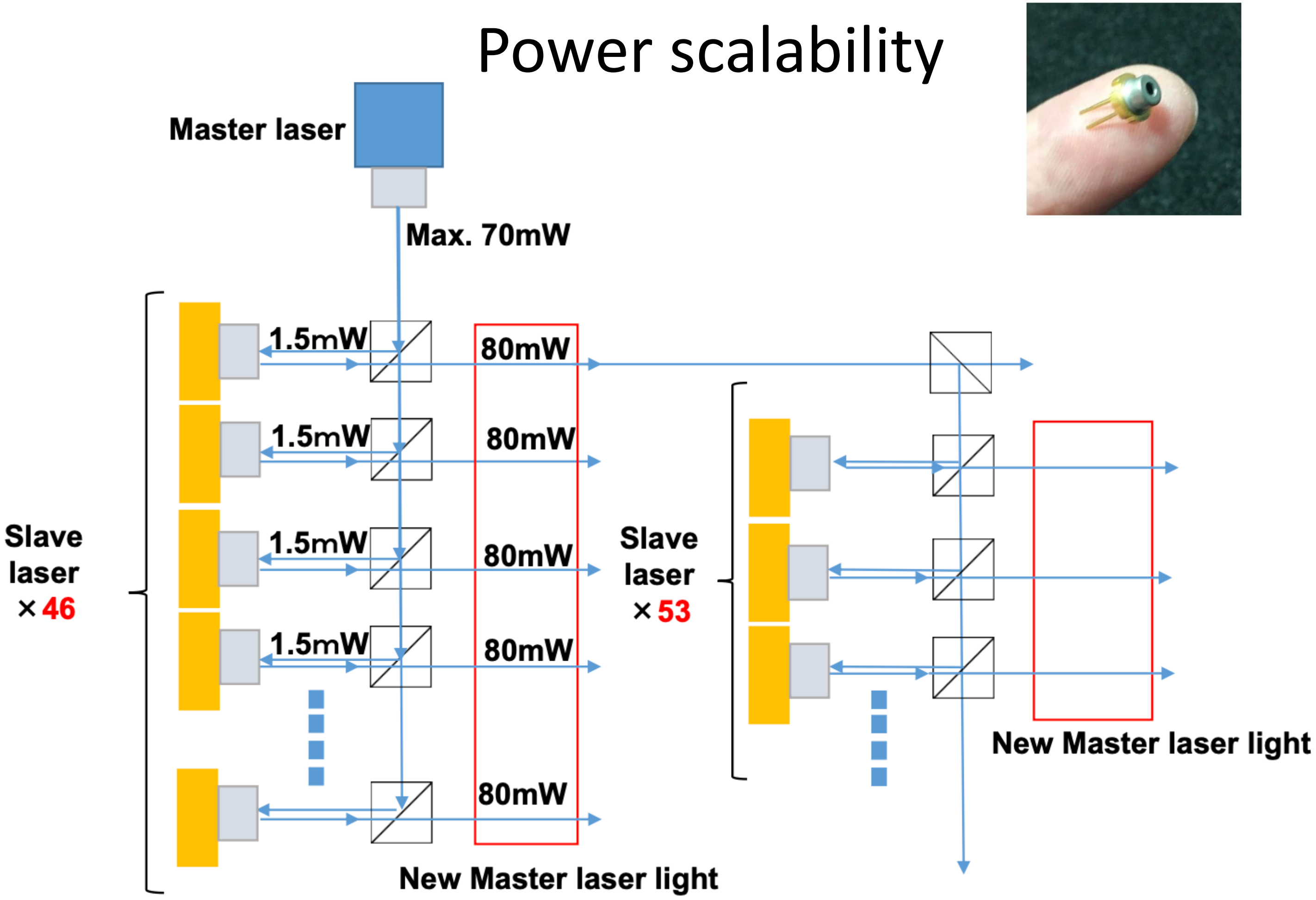
2 MHz rms → 422.792xxxxx ±0.0000006 nm

< 2MHz rms and long term operation was obtained

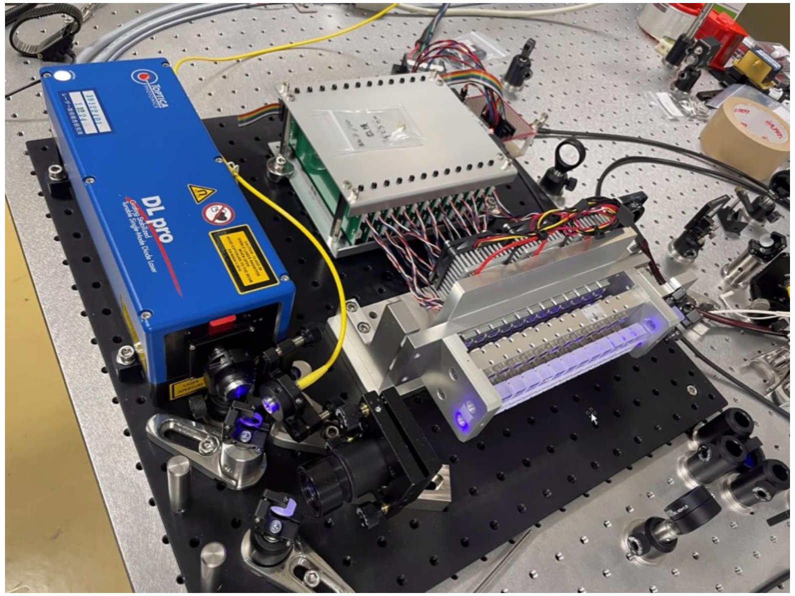
Development of the laser system

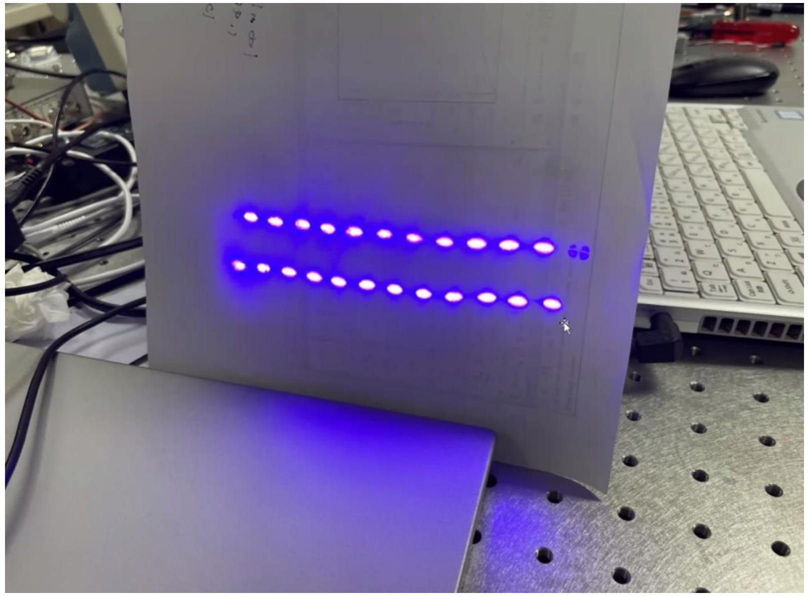
@ICR Kyoto Univ., ILE, and ILT

Power scalability



200 W laser system can be realized using ~2,500 slave lasers and 1 master lasers.

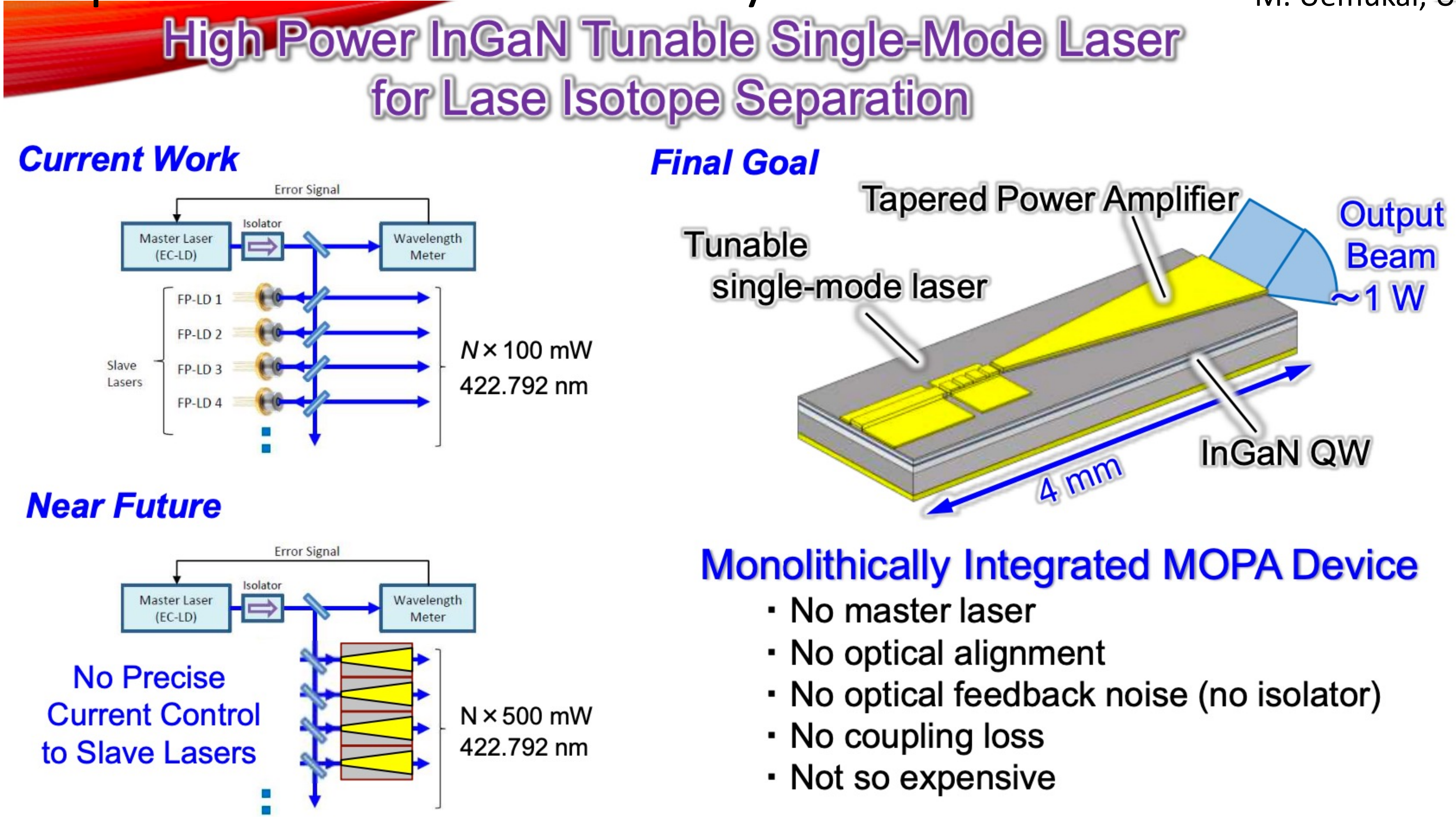




23 slave lasers for 2W laser power

Development of the laser system

M. Uemukai, UGAP 2024



Single-frequency and high power laser could be realized.

Research strategies and requirements

University of Fukui

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

Atomic beam system
- Small scale chamber
- Collimator effect
- Sheet-like atomic beam

ICR, Kyoto Univ., LIE, and LIT

Laser system
- Single frequency laser
- Power-scalable laser
- InGaN tunable + Tapered SOA
- Long-term operation
- Stable laser system

Research Center for Nuclear Physics (RCNP)

Collection system
- Collection plate
- Recovery system

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

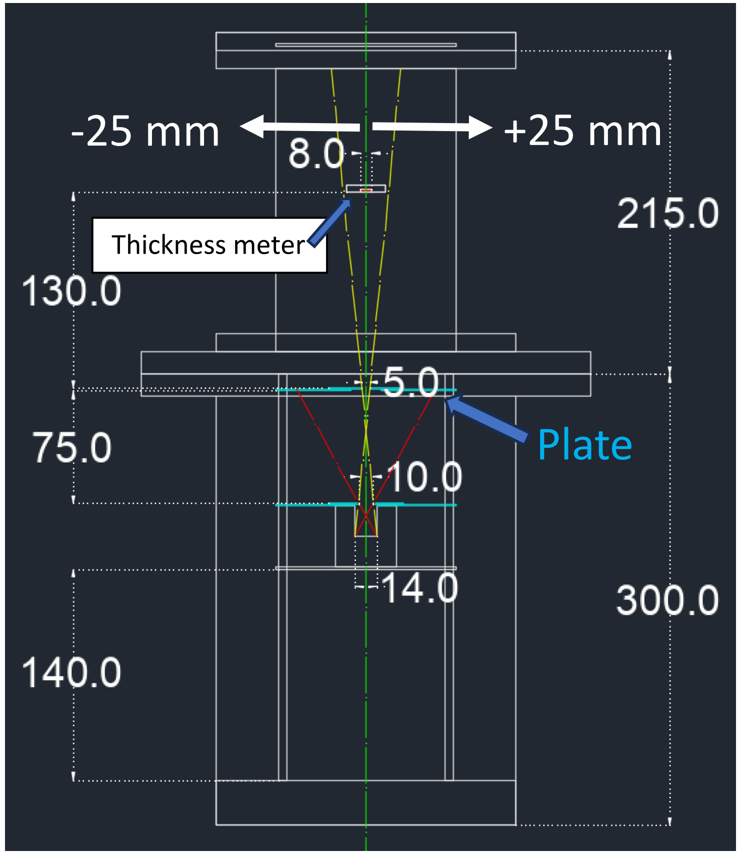
RCNP, Osaka U + ICRR, Tokyo U.

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

Candles

Future development
- Stable operation 1/6 port
- Increase the production rate by multiple 6 port units
- Ton scale production
- LISSE

Development of the collection system



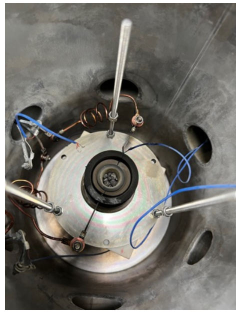
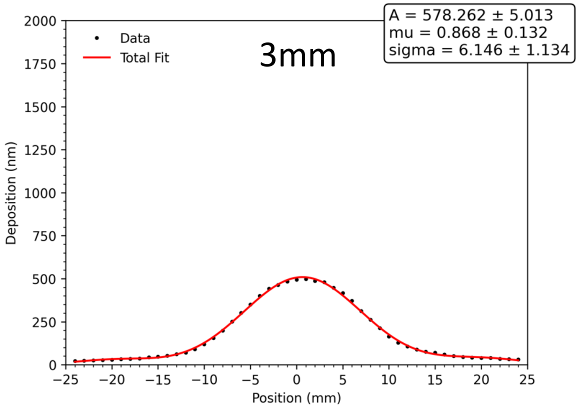
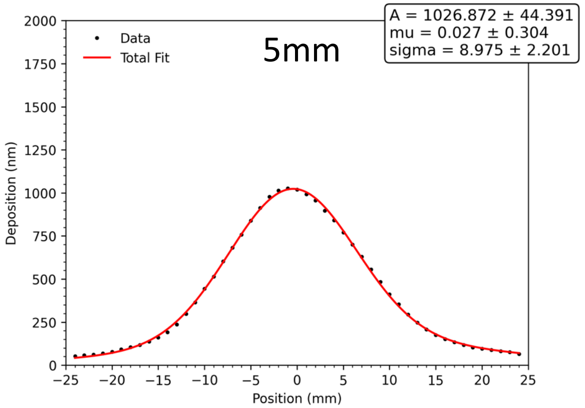

Crucible, 0.7g Ca

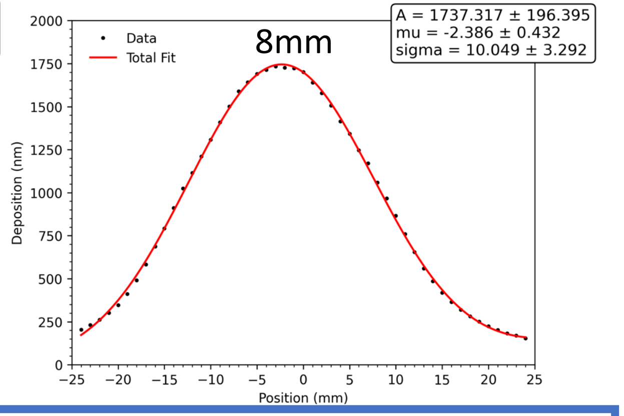

Plate slit

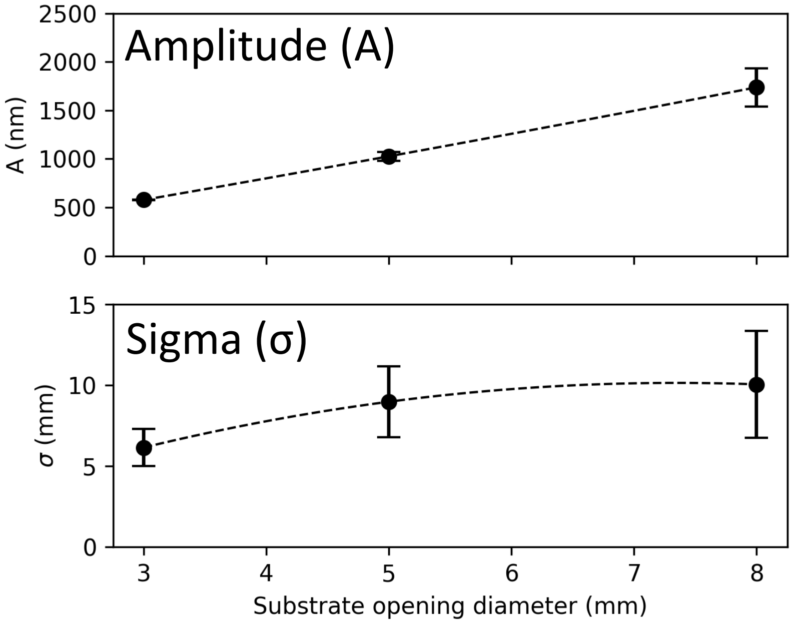

Deposition meter

3 RUNs with different slit collimators

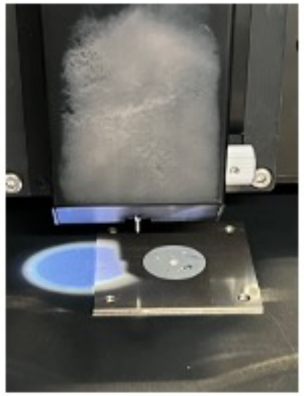


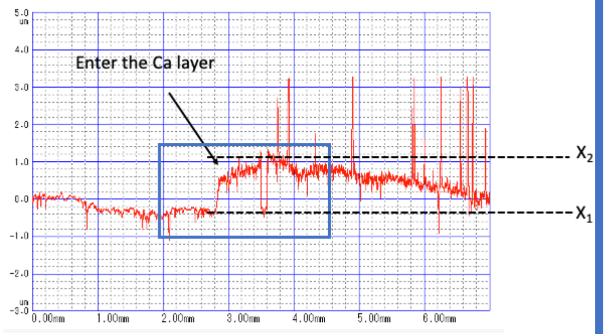






Thickness measurement will be performed by the stylus method.

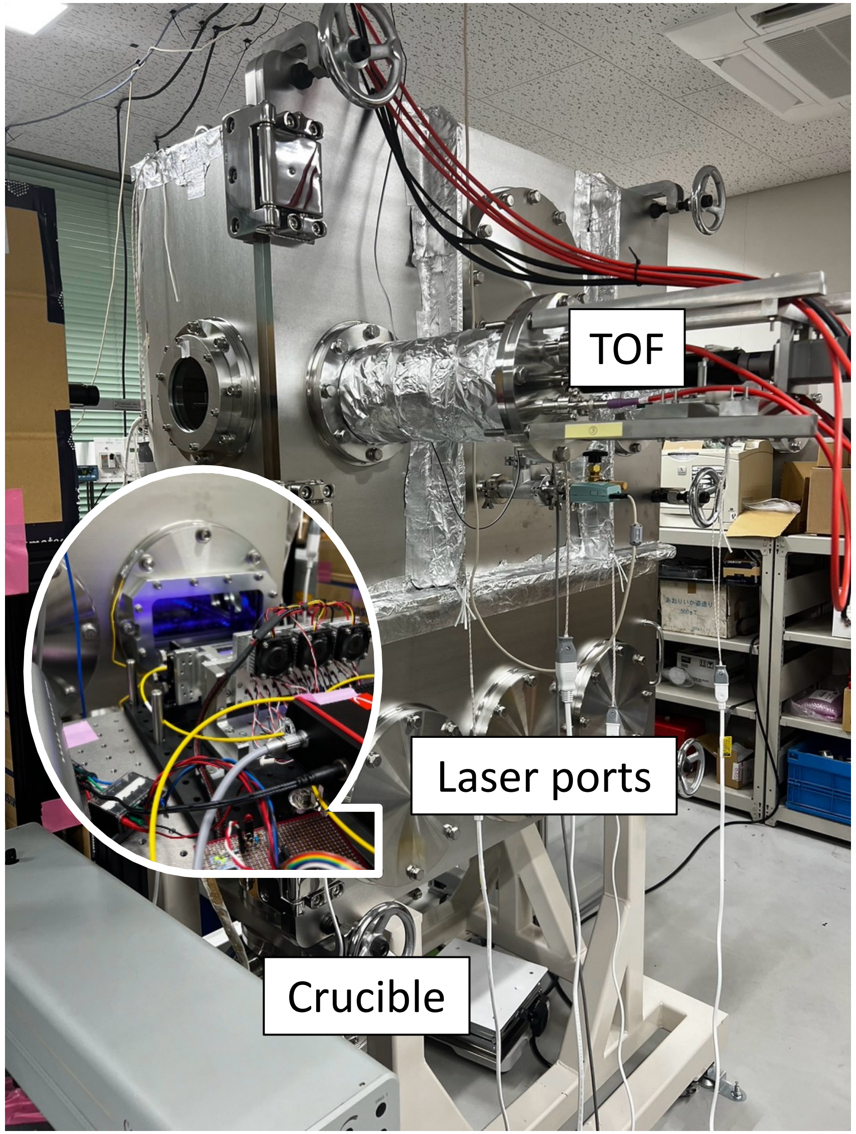




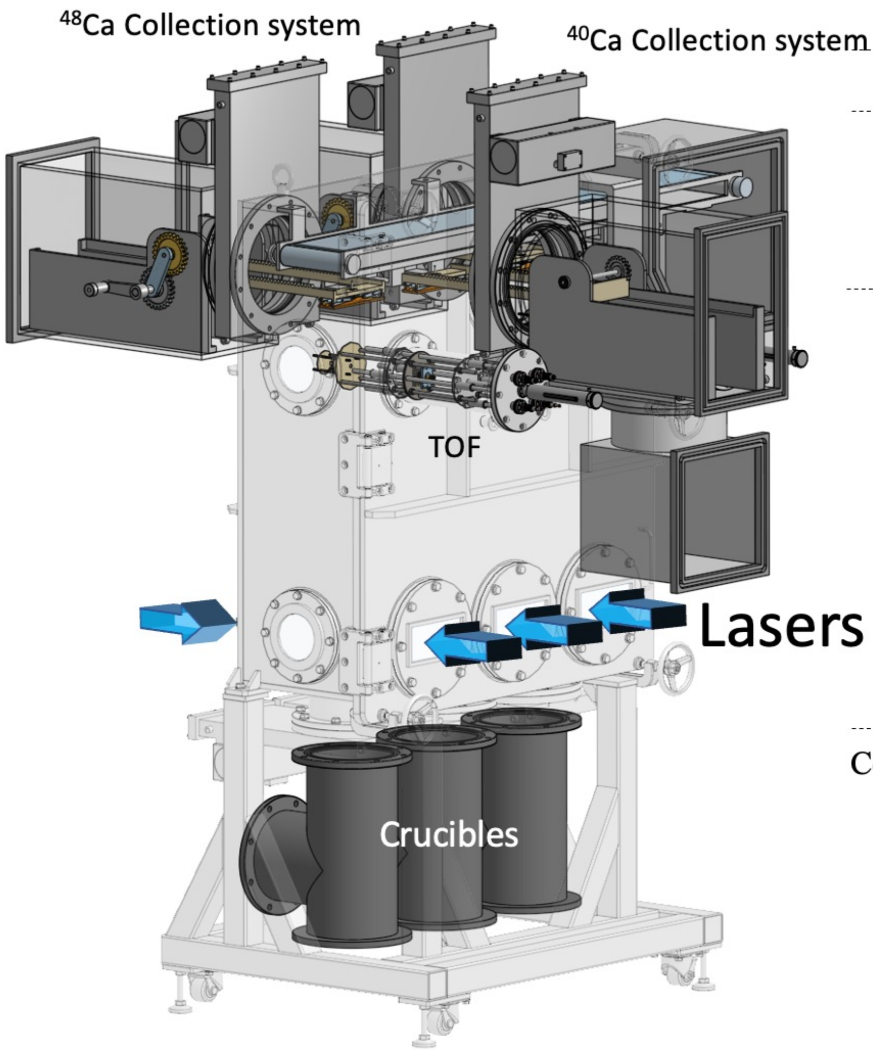
A trial measurement of 1.3 μm thick

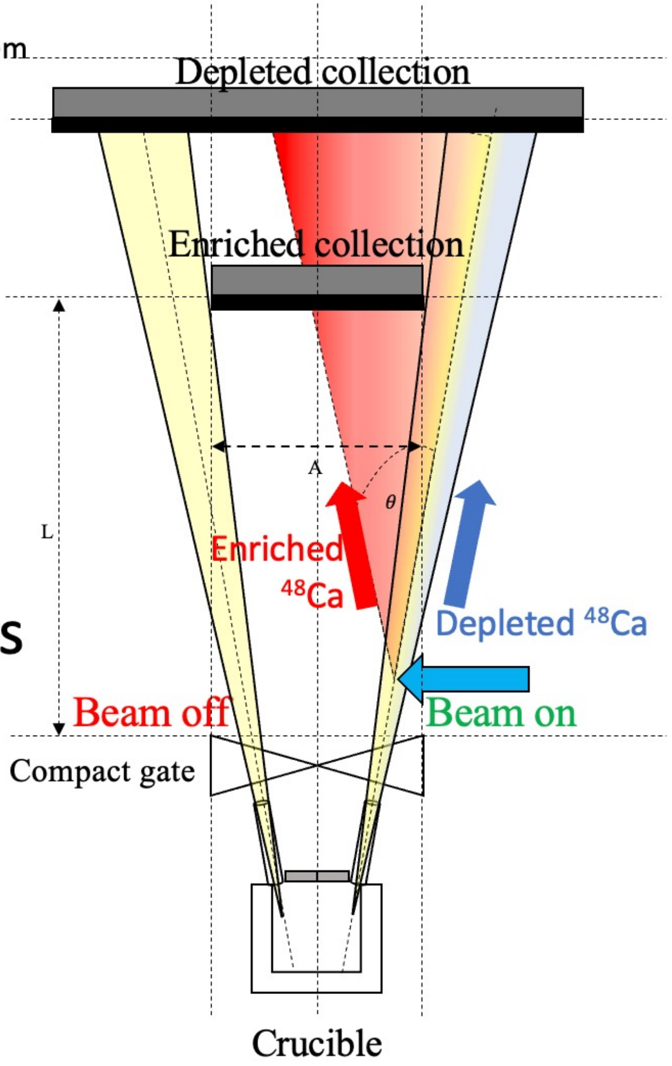
Production chamber

RI Center, Osaka University, Toyonaka, Osaka, Japan

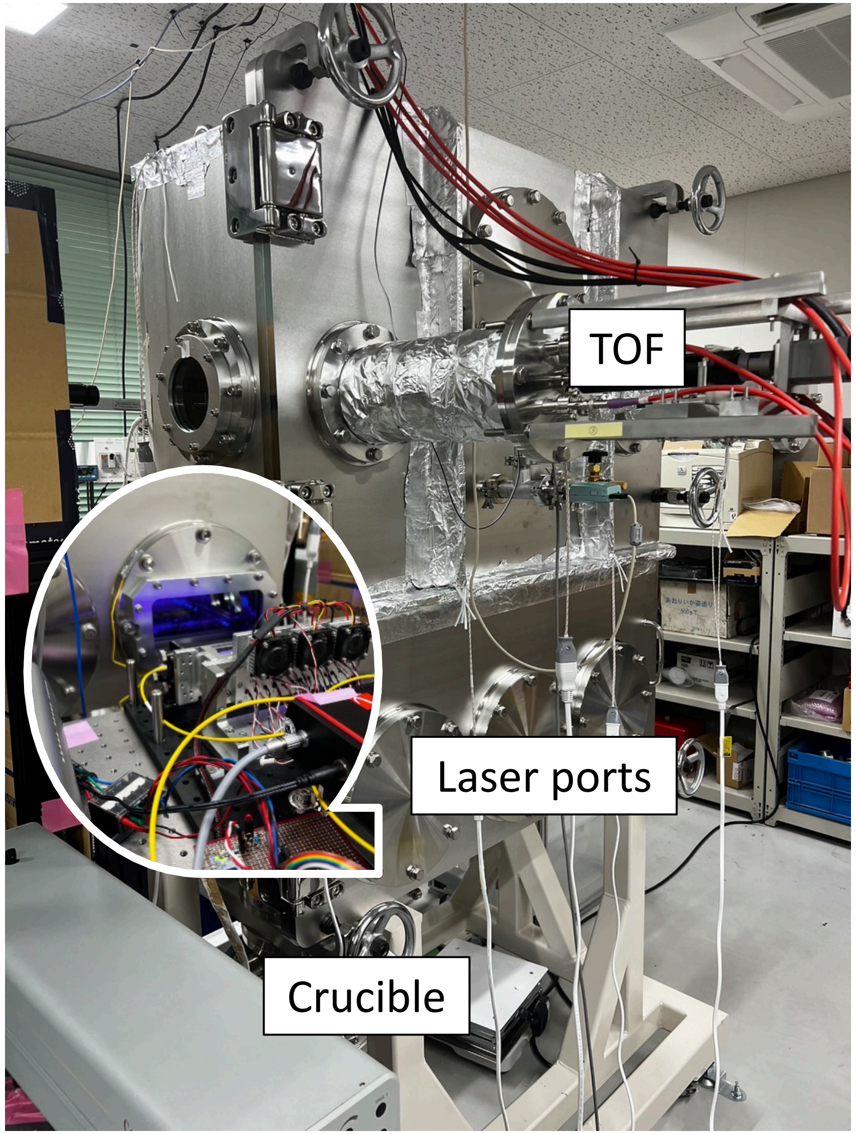


80 mW × 23 lasers = ~ 2W
6 iraadiation ports -> aim for 2 mol/yr



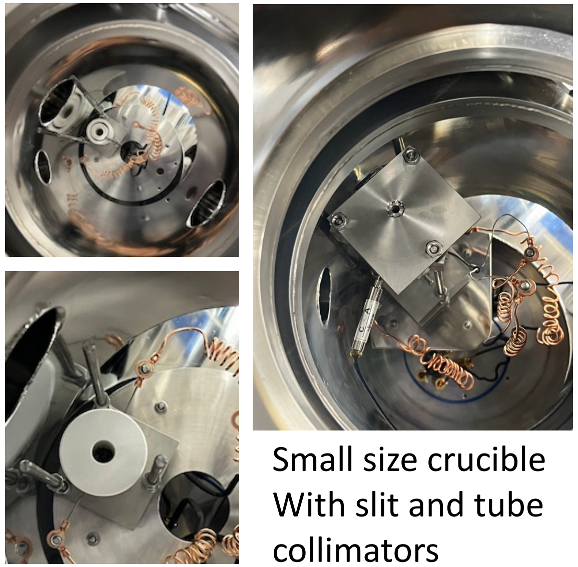


Production chambe

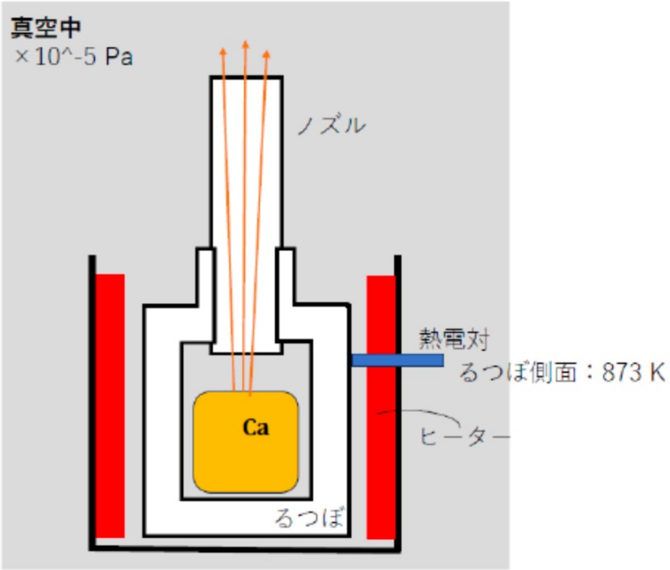


80 mW × 23 lasers = ~ 2W
6 iraadiation ports -> aim for 2 mol/yr

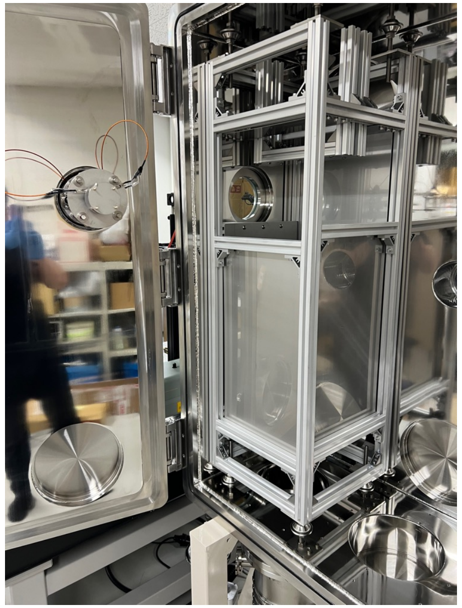
Atomic beam production system

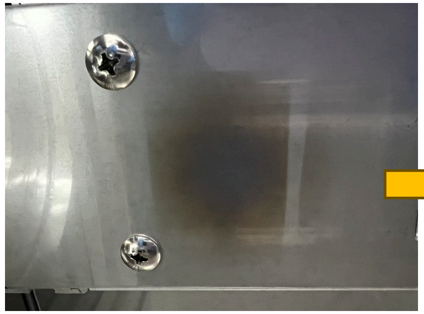


Small size crucible
With slit and tube
collimators



Collection system

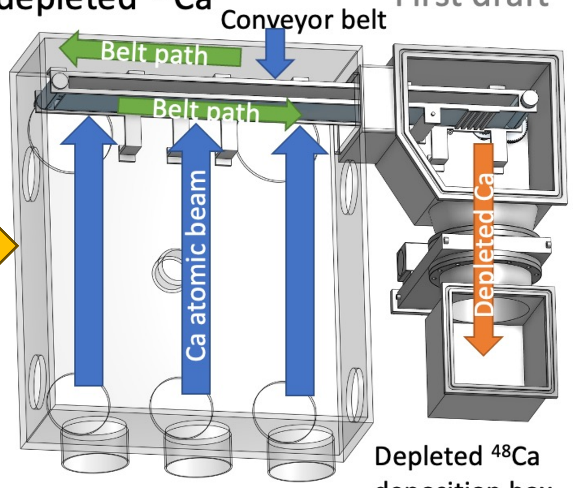




Ca layer is
observed at the
recovery plate

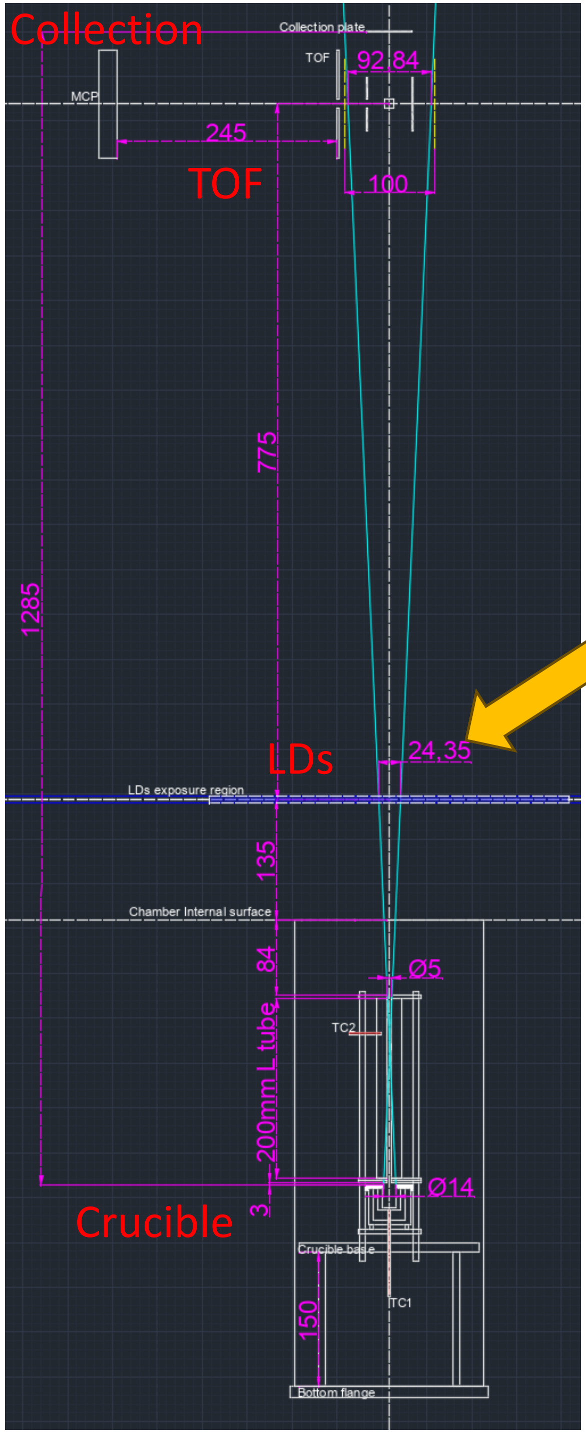
For depleted ^{48}Ca

First draft

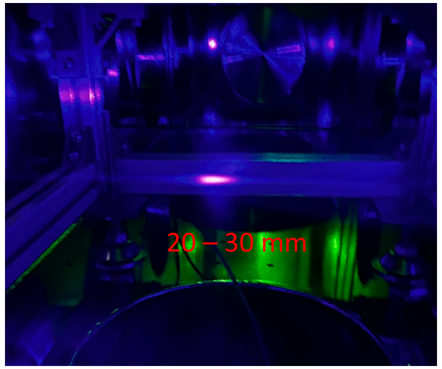


Depleted ^{48}Ca
deposition box

Atomic beam monitoring

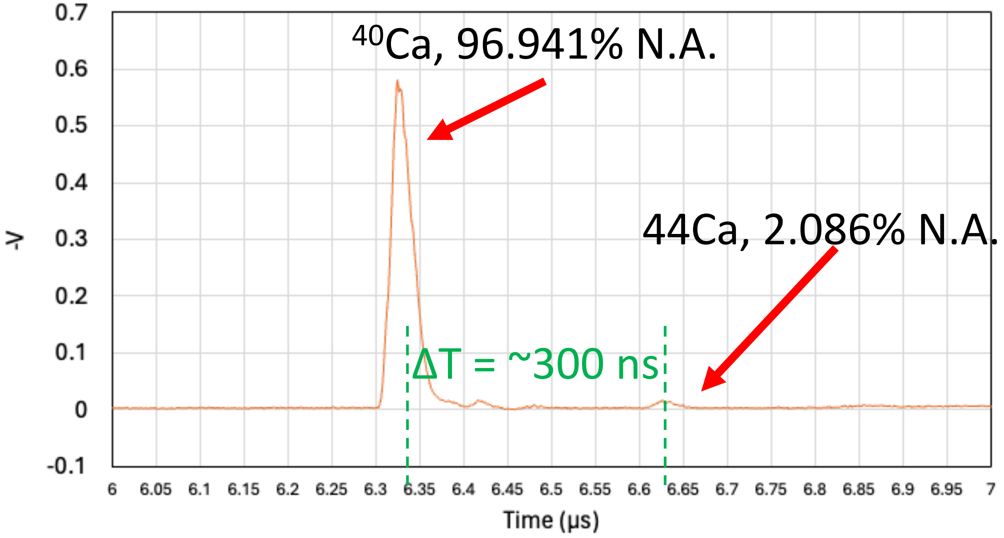


Well-collimated



Fluorescence light by LDs

TOF spectra



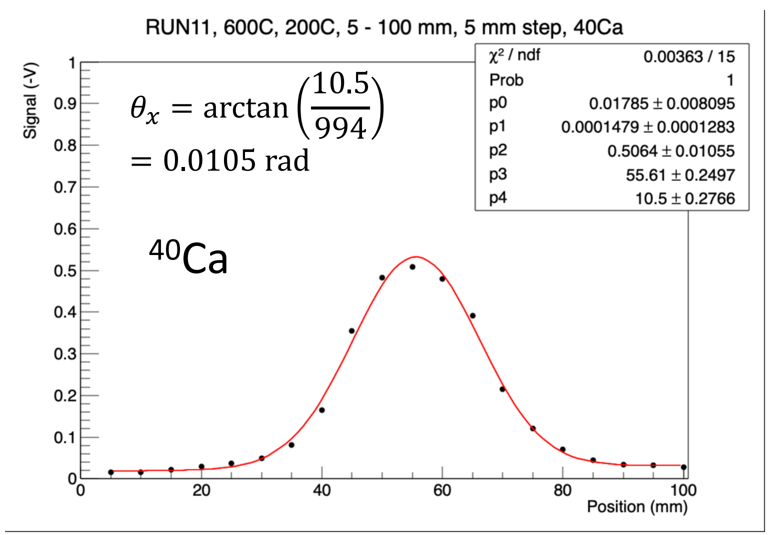
^{40}Ca/^{44}Ca N.A. ratio = 46.5

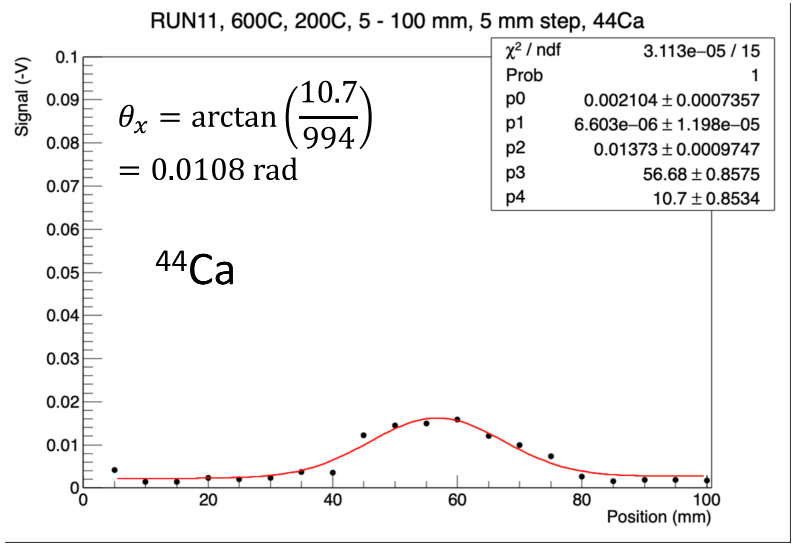
^{40}Ca/^{44}Ca peak ratio = 47.3

Next, verify

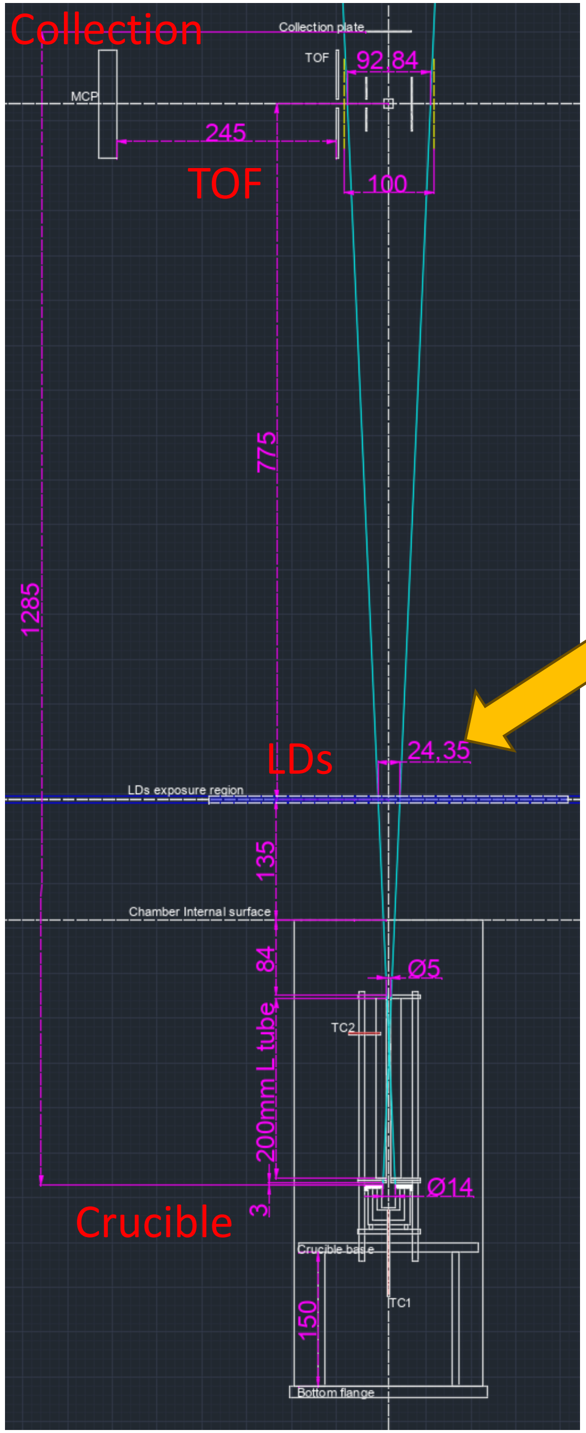
^{48}Ca, 0.187% N.A.

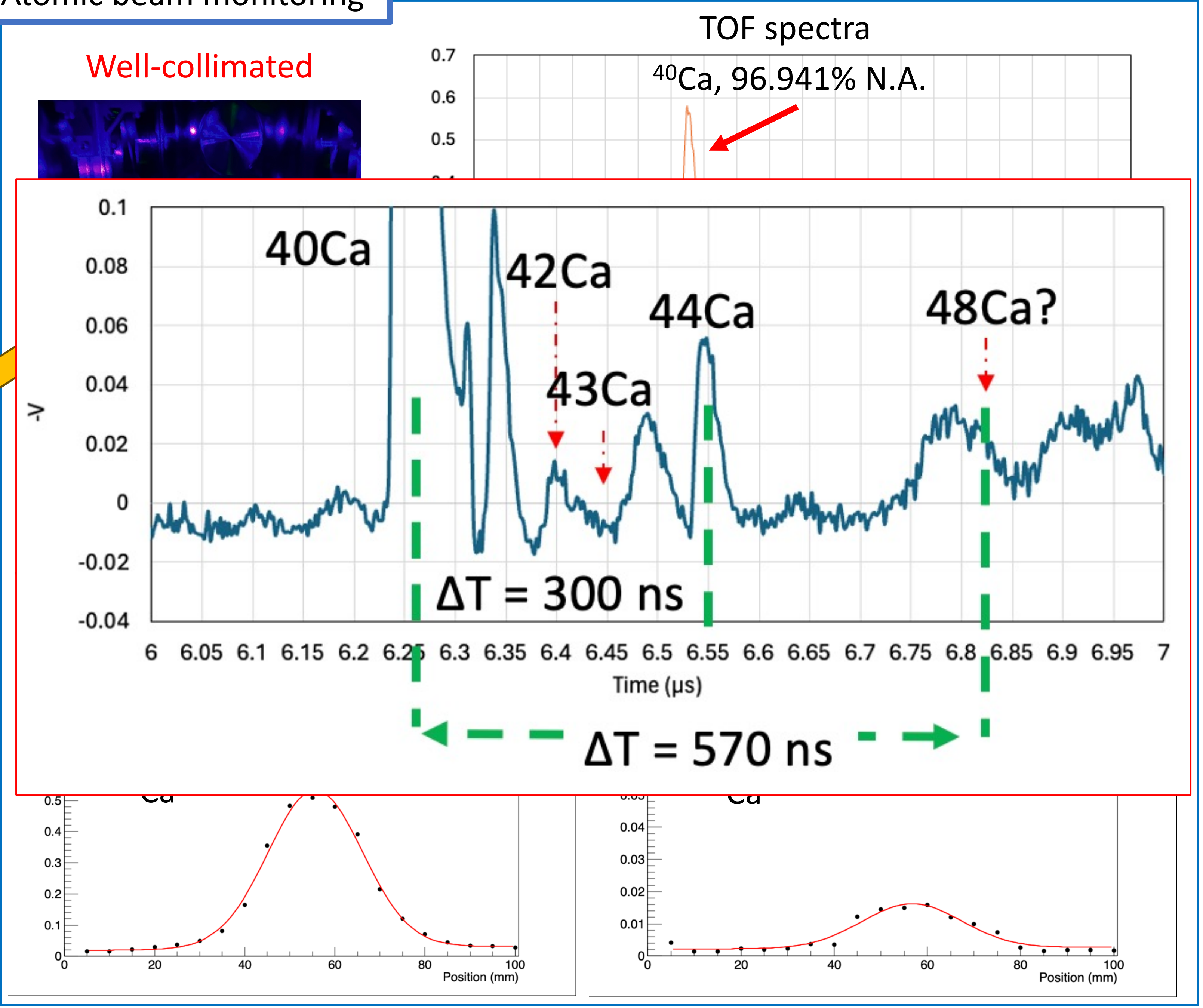
Spatial distribution



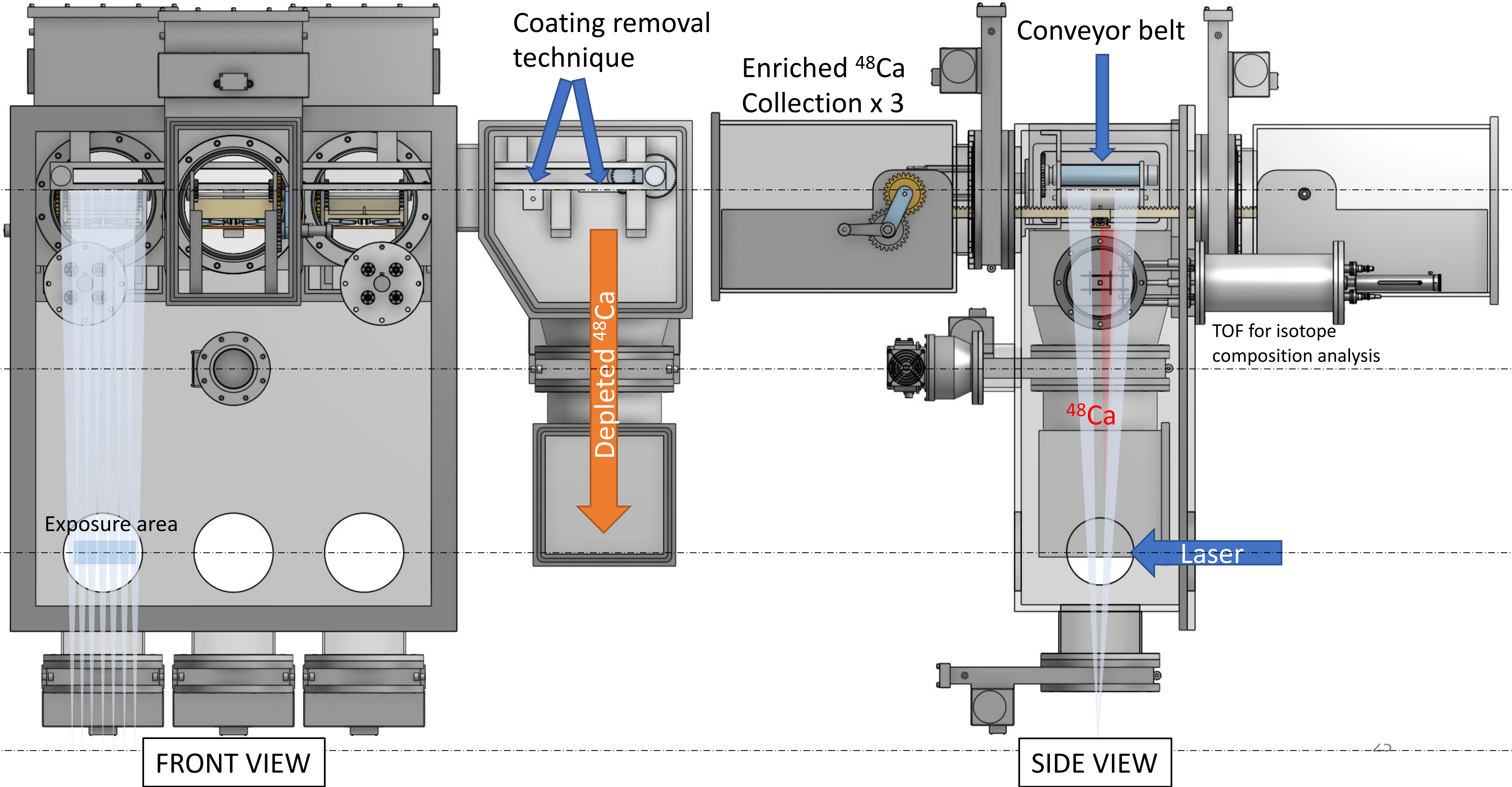


Atomic beam monitoring





Strategies for mass production



Strategies for mass production

What is the required laser power?

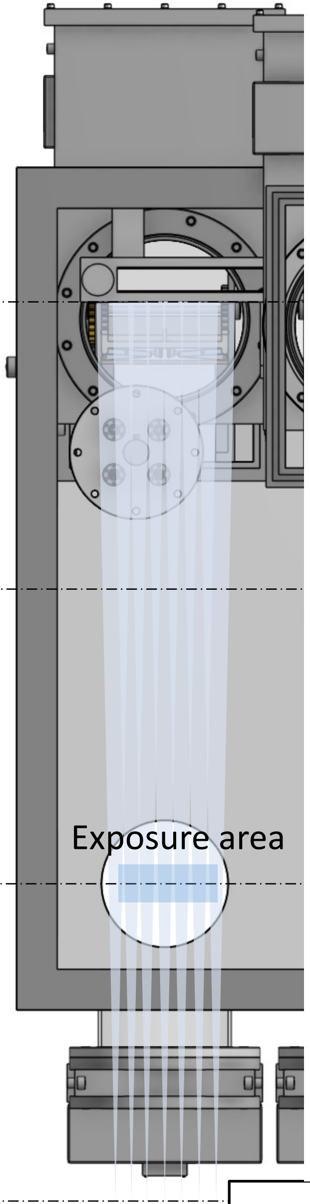
Photon energy: 4.7×10^{-19} J @ 423 nm

Number of photons absorbed by 1 atom: 1,000

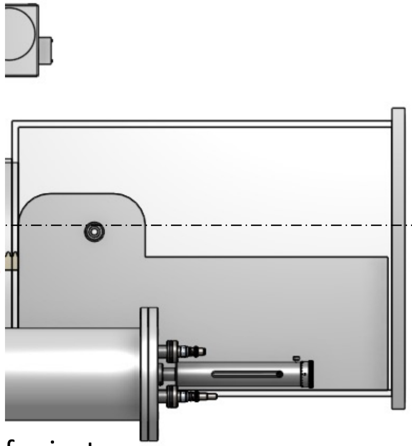
→ Number of ^{48}Ca produced by 1 W laser: 2×10^{15} sec^{-1} → ~5 g/W/year

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

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



FRONT VIEW



for isotope position analysis

SIDE VIEW

Strategies for mass production

FY2023, Installation, and investigation

- Collection system installation ✅
- Crucible installation ✅
- Monitor and control system ✅
- Laser installation (SOON)
- 2W laser power (10 g/year)
- Isotopic composition analysis (TOF, ICP-MS)

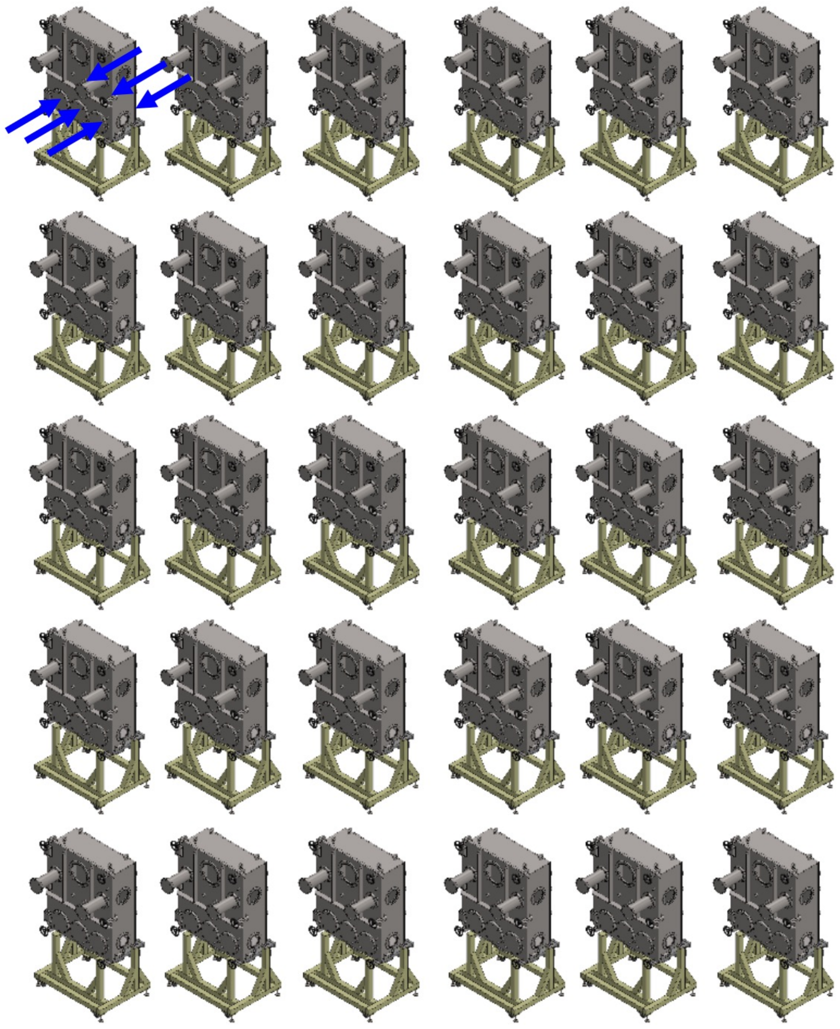
FY2024, Production

- Stable operation of 1st port (10 grams/year)
- Full operation of six ports (2mol/year)

Future

- Automated collection system
- Scale up the mass production ~ 30 units

300 kg/years production plan



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

Power/LD => 1 W
Number of LDs/port = 1700
Optical power => 1.7 kW/unit
Total optical power:
~2 kW/unit -> ~60kW/30 units

Summary

- LIS for ^{48}Ca has been 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 (University of Fukui)
    - Proof-of-principle for LIS
    - Collimator effect and atomic vapor production -> Sheet-like atomic beam
  - Power scalable laser system (ILE, ILT)
    - EC-LD + FP-LD + PD-LDs -> stable and high power laser
    - InGaN tunable + Tapered SOA
  - Collection system (RCNP)
    - Collection material study (SUS, Al, Cu, alloy, etc.)
    - Automated production system
  - Large-scale production system (RCNP)
    - 2mol/years (1st milestone)
    - **LISSE -> Shima-san's talk**
- We will soon launch the trial production for 10 grams/year (2W).

Collaborators

Candles

RCNP

大阪大学
OSAKA UNIVERSITY

UNIVERSITY OF FUKUI

KYOTO UNIVERSITY
Institute for Chemical Research
ICR

筑波大学
University of Tsukuba

Institute of Laser Engineering
Osaka University

大阪産業大学
OSAKA SANGYO UNIVERSITY

徳島大学
Tokushima University

Thank you for your attention

anawat@rcnp.osaka-u.ac.jp