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

Anawat Rittirong*, LIS and CANDLES collaborations

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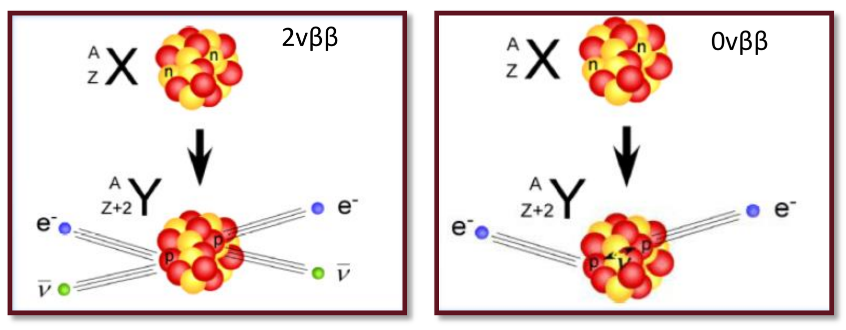
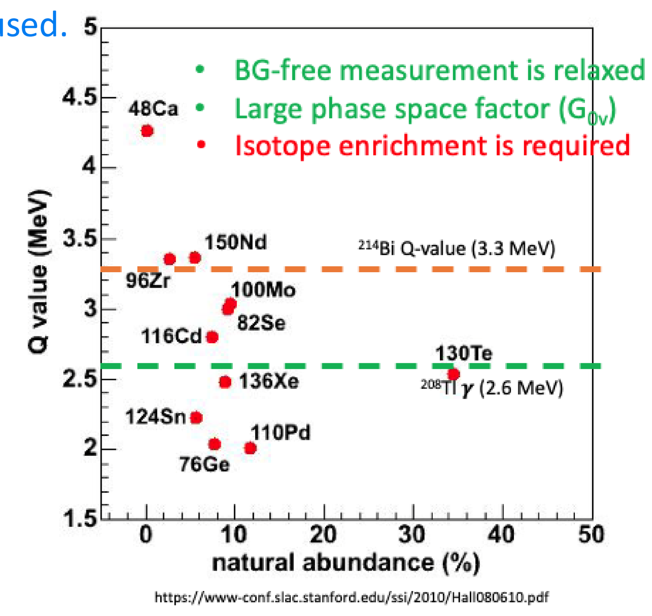
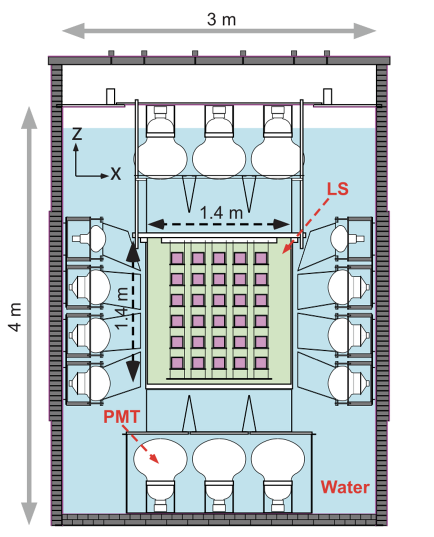
CANDLES

- **Purpose:** To verify the Majorana nature of neutrino via neutrinoless double beta decay (0νββ).
- ^{48}Ca (Q-value =4.27 MeV, highest energy for 2ββ decay) is used.



0νββ is beyond the standard model!!

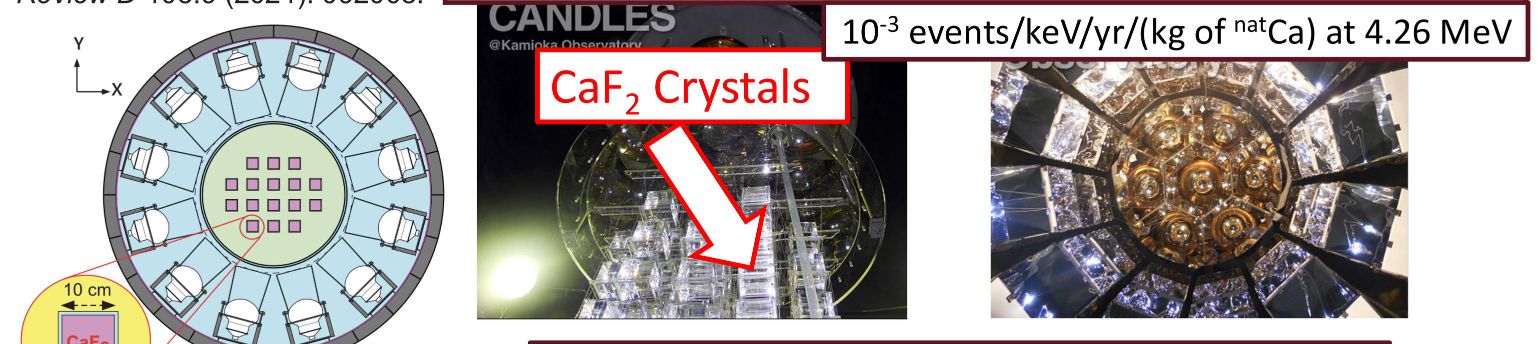




S. Ajimura, et al. Physical Review D 103.9 (2021): 092008.

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) at 4.26 MeV



Large amount of ^{48}Ca is required (tons scale)

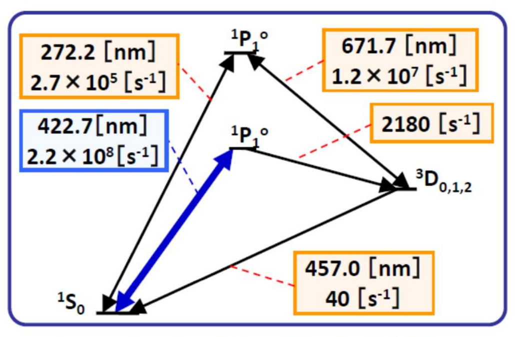
- Regrettably, the gas centrifuge or gas diffusion techniques are not viable for the isolation of calcium due to the unavailability of the gaseous compound.
  Expensive [~1,000,000 $/g] via an electromagnetic separator

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

Ca Isotope	^{40}Ca	^{42}Ca	^{43}Ca	^{44}Ca	^{46}Ca	^{48}Ca
% abundance	96.94	0.65	0.135	2.09	0.004	0.187

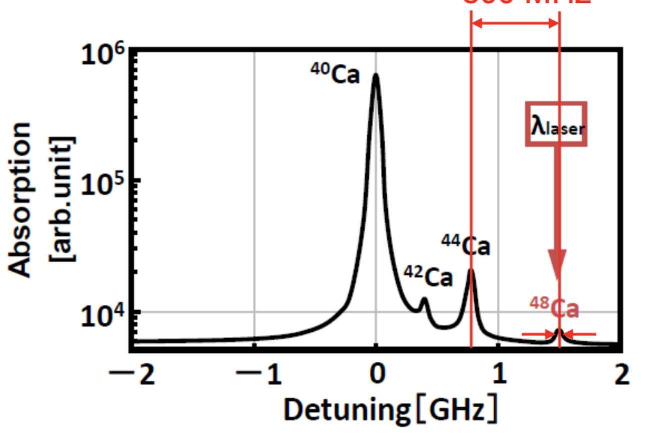
Laser Isotope Separation (LIS) for ^{48}Ca

Transition : $^1P_1$ - $^1S_0$
λ = 422.7 nm
τ = 4.6 ns
$\Delta\nu_{nat.} = \frac{1}{2\pi\tau} = 34$MHz (HWHM)
I_s = 59.9 mW/cm²



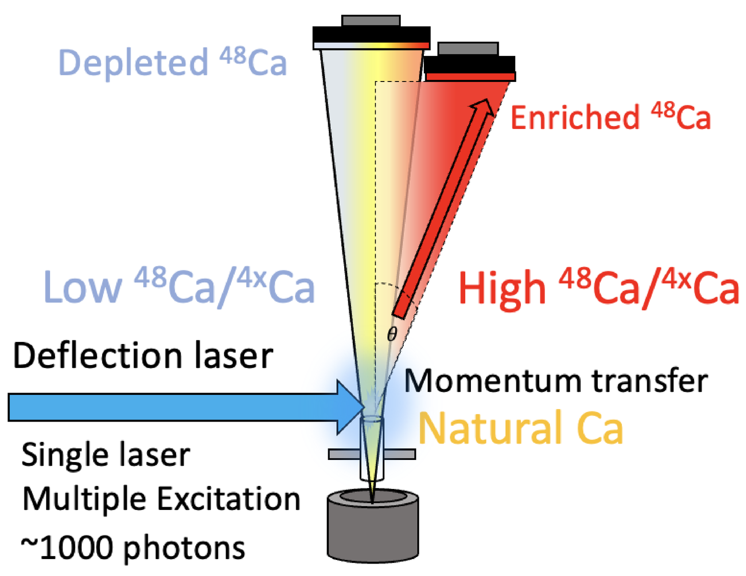
Energy levels and Transition Probabilities

Isotope shift: ~800 MHz

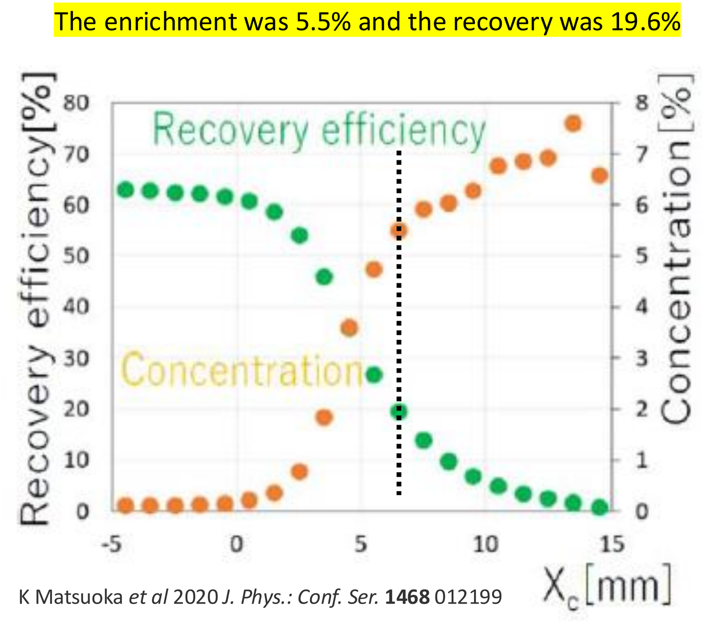


Absorption spectrum of Ca at 423nm

- Deflection method

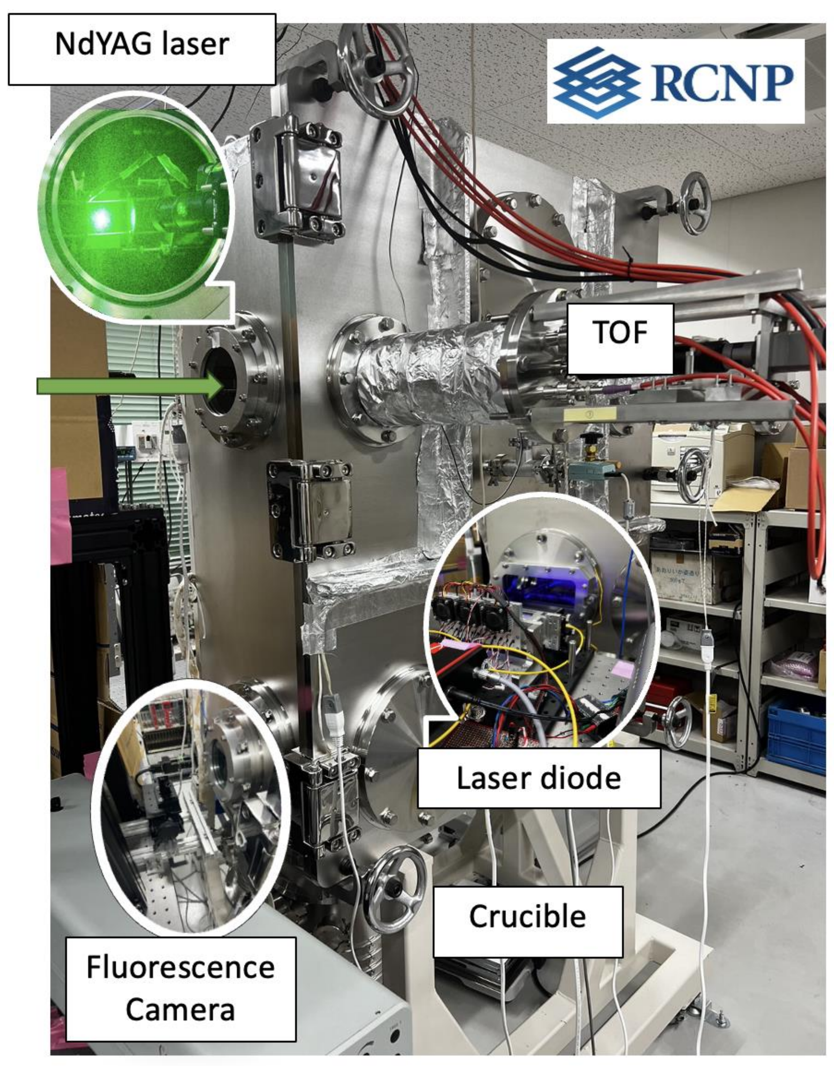


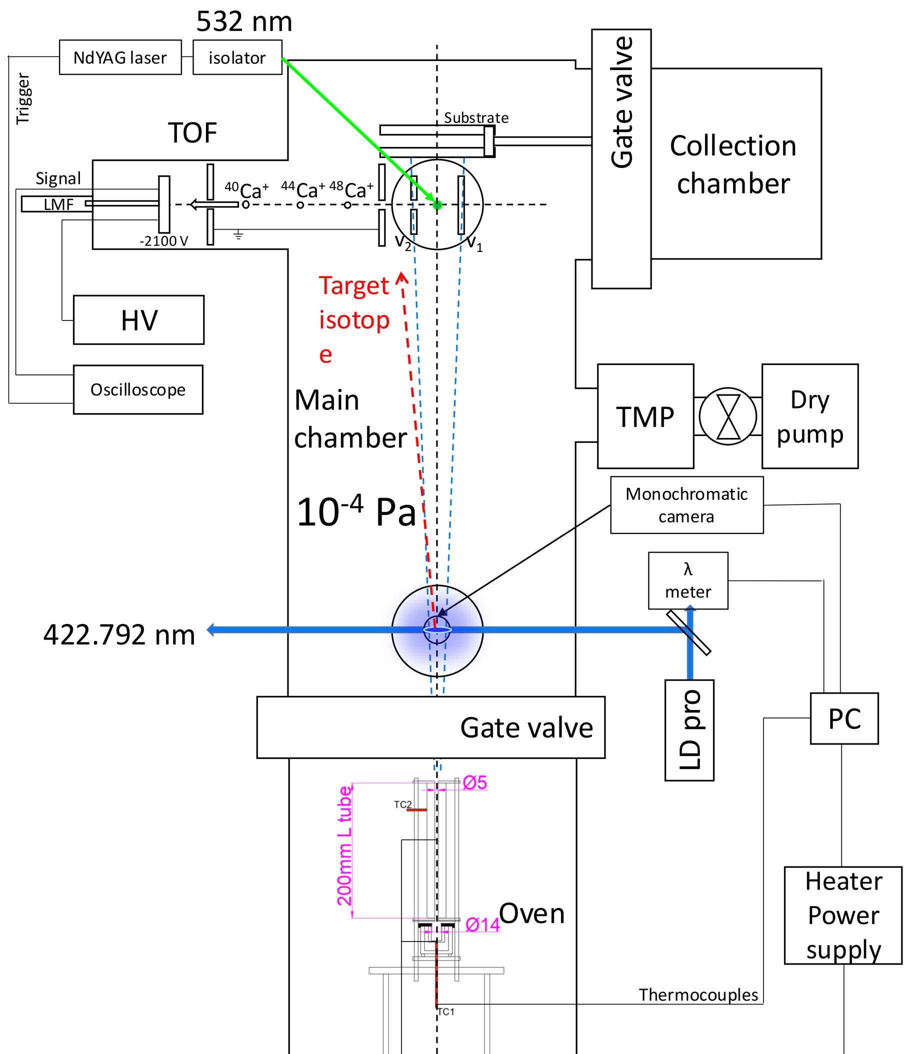
The enrichment was 5.5% and the recovery was 19.6%



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

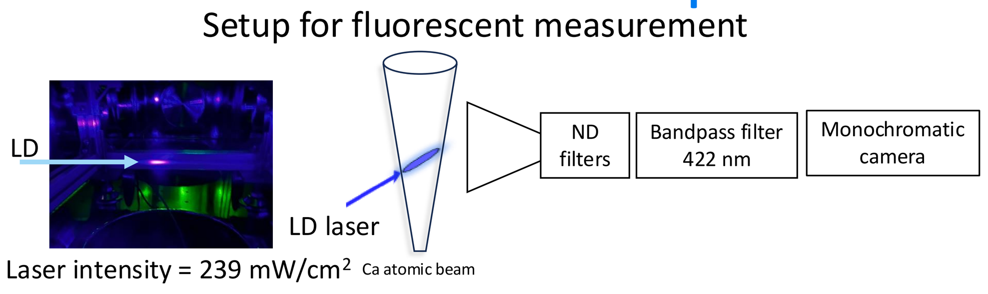
LIS main chamber @The University of Osaka, Toyonaka campus





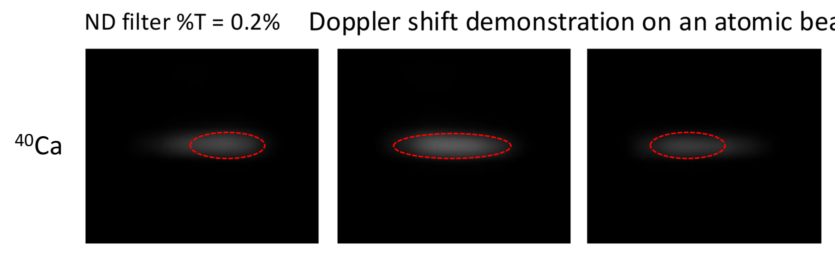
Isotope shift measurement

Setup for fluorescent measurement



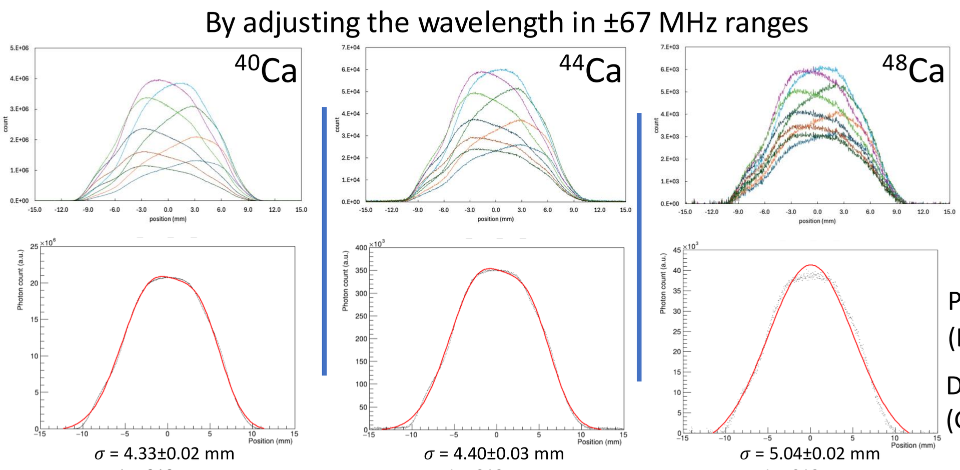
Laser intensity = 239 mW/cm²

ND filter %T = 0.2% Doppler shift demonstration on an atomic beam



422.79168 nm 422.79170 nm 422.79172 nm

By adjusting the wavelength in ±67 MHz ranges



Intensity ratio vs Natural abundance ratio

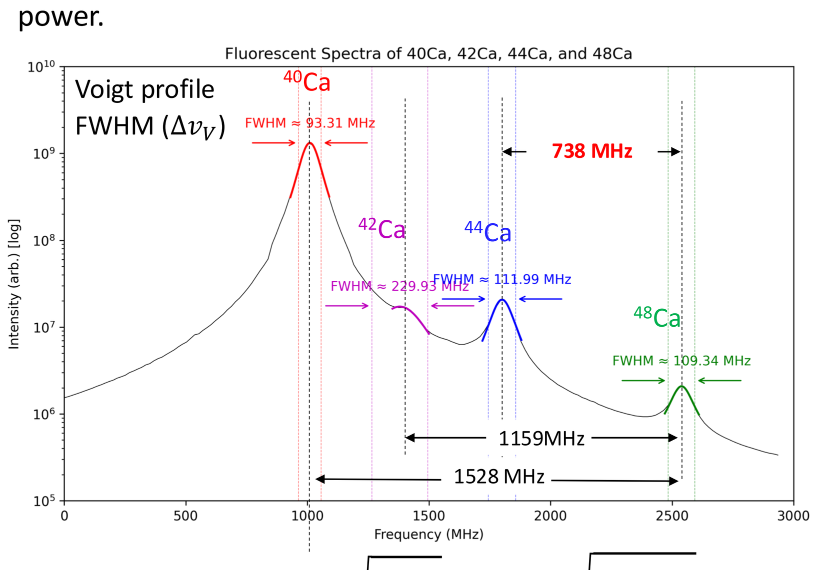
$I_{48/40} = 0.001857$ $I_{44/40} = 0.017406$

$N.A._{48/40} = 0.001929$ $N.A._{44/40} = 0.021518$

−3.90% −19.11%

Fluorescent measurement

1. Ensure the presence of a calcium atomic beam.
2. Identify the isotope shift at the 422.792 nm transition.
3. Measure the angular distribution (θ) of the atomic beam
4. Identify the Doppler broadening at the operating temperature (500 °C), and implement appropriate techniques to minimize its impact and optimize the utilization of laser power.



Power broadening (Lorentzian profile) $\Delta\nu_L = \Delta\nu_N \times \sqrt{1+\frac{I}{I_S}} = 34.7 \times \sqrt{1+\frac{239}{59.9}} = 77.51\ MHz$

Doppler broadening (Gaussian profile) $\Delta\nu_V \approx 0.5346\Delta\nu_L + \sqrt{0.2166\Delta\nu_L^2 + \Delta\nu_G^2}$

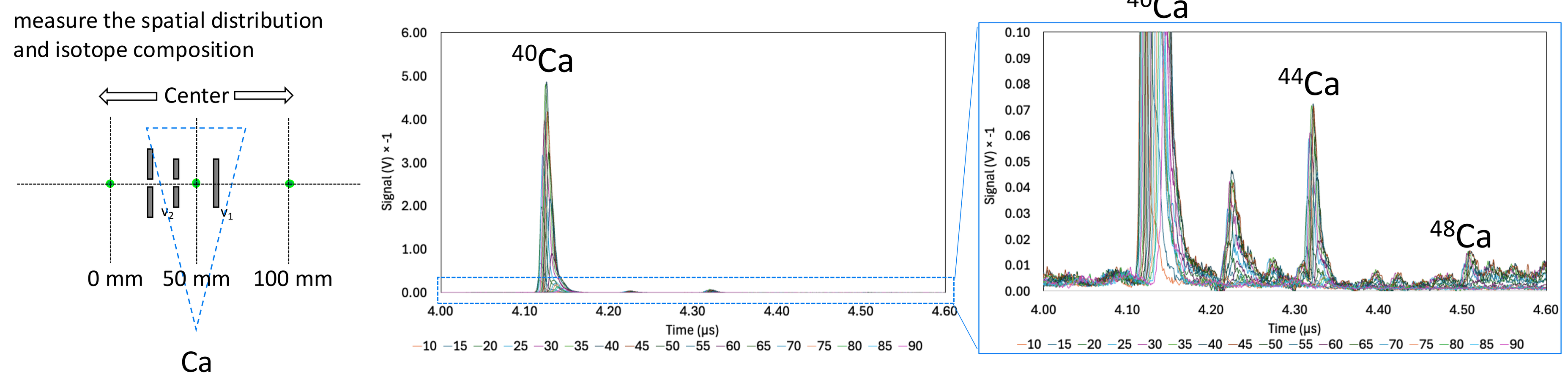
$93.31 \approx 0.5346(77.51) + \sqrt{0.2166(77.51^2) + \Delta\nu_G^2}$

$\Delta\nu_G \approx 37.28\ MHz$

- The isotope shift of ^{48}Ca at the 422.792 nm transition is 738 MHz.
- The Doppler broadening ranged from 93 MHz to 112 MHz, except for ^{42}Ca.
- ^{40}Ca tail interfered with other isotopes. Power broadening can be improved by reducing the laser intensity.

TOF measurement

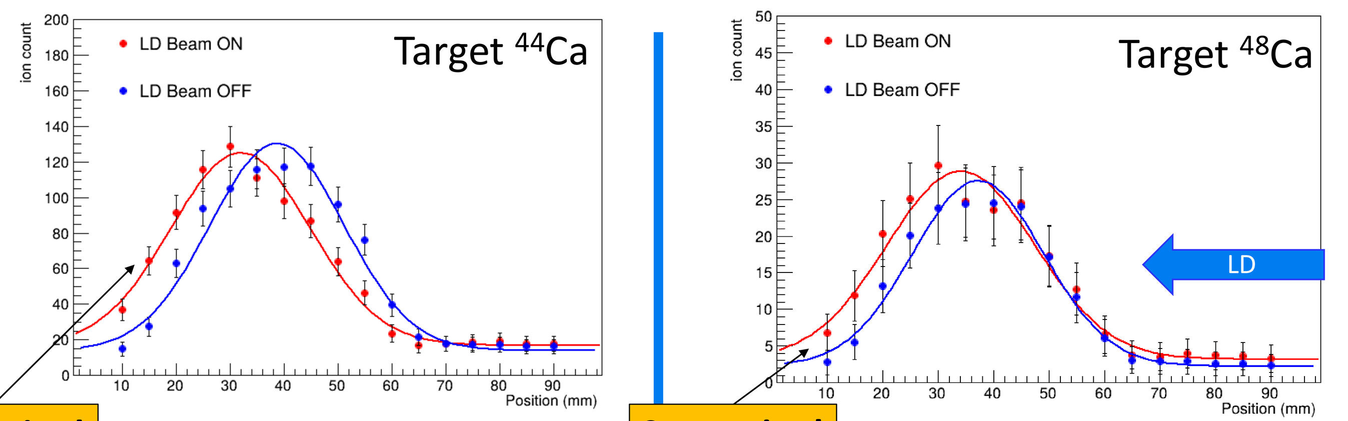
The TOF system was moved over the calcium atomic beam to measure the spatial distribution and isotope composition



Displacement of the target calcium isotope when the beam is on and off

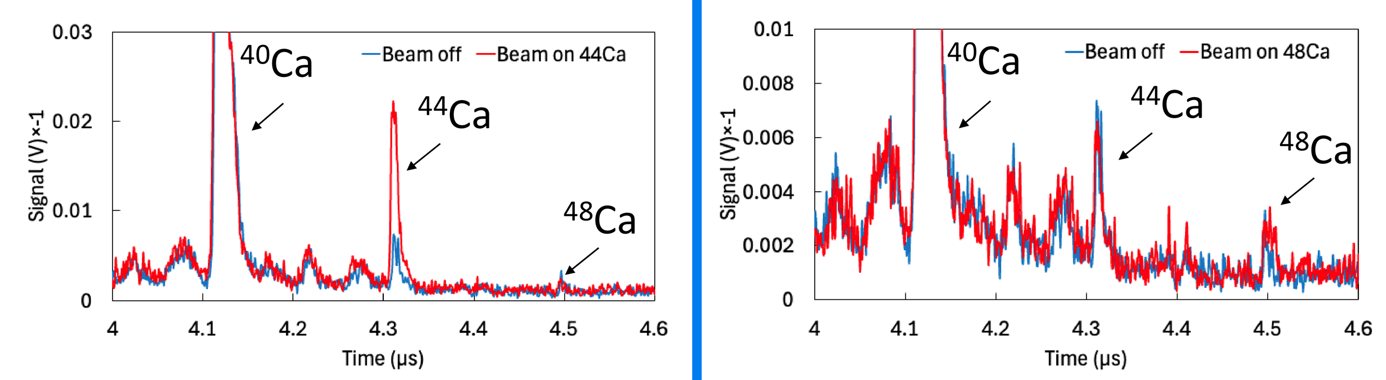
Q (ion) $= \frac{\Sigma V \Delta t}{qGz} \pm \sqrt{Q}$

ΣV = voltage signal
Δt = time difference (s)
q = ion charge
G = MCP gain
Z = MCP resistance (50 Ω)



Separation! Displacement = 6.77 ± 0.89 mm **Separation!** Displacement = 3.10 ± 1.79 mm

TOF spectra at the separation position (P = 10 mm)



Success separation of ^{44}Ca by laser deflection Improve the sensitivity of ^{48}Ca

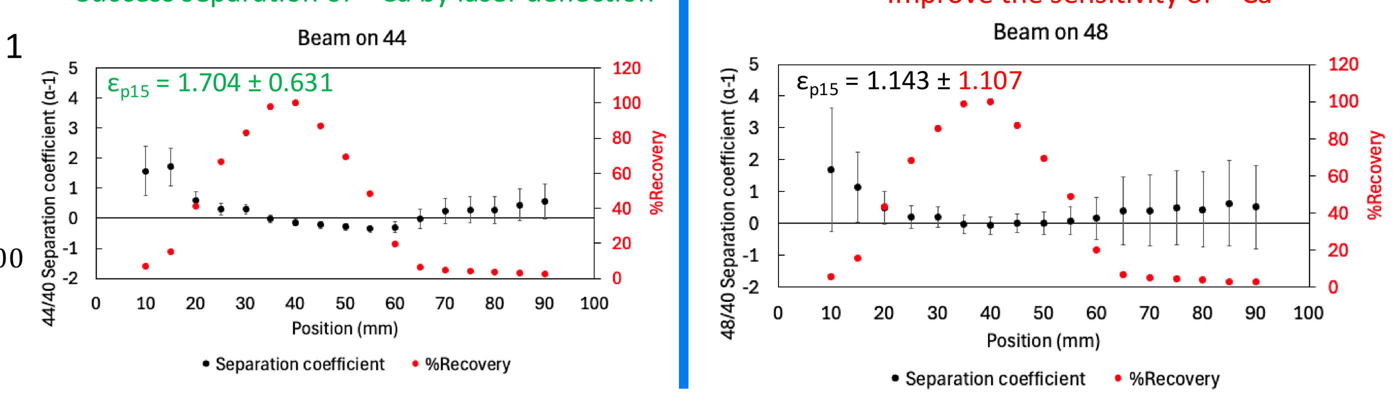
Separation coefficient (ε) = α – 1

$\alpha = {}^{4x/40}IR_{Beamon} / {}^{4x/40}IR_{Beamoff}$

$IR = {}^{4x}Q/{}^{40}Q$

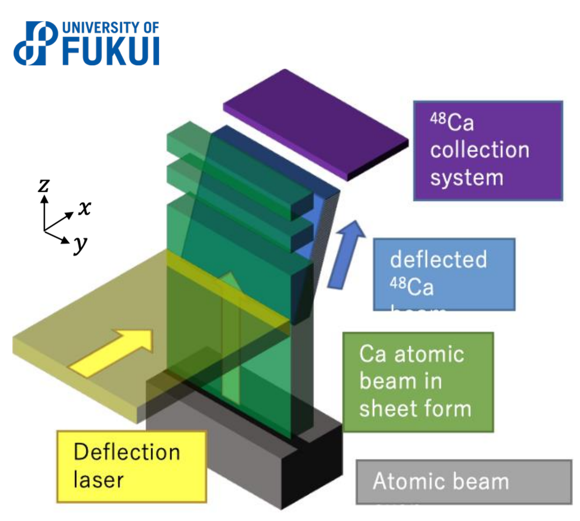
$\%Recovery\ of\ calcium_x = \frac{Total\ calcium\ at\ x\ position}{Total\ calcium\ at\ peak\ position} \times 100$

At p15 %recovery = 15.5 ± 0.5 %

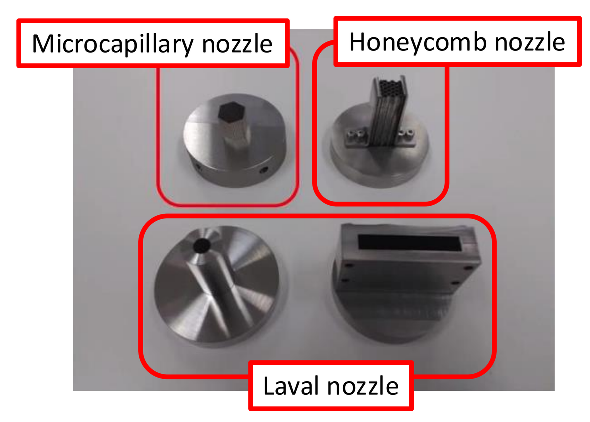


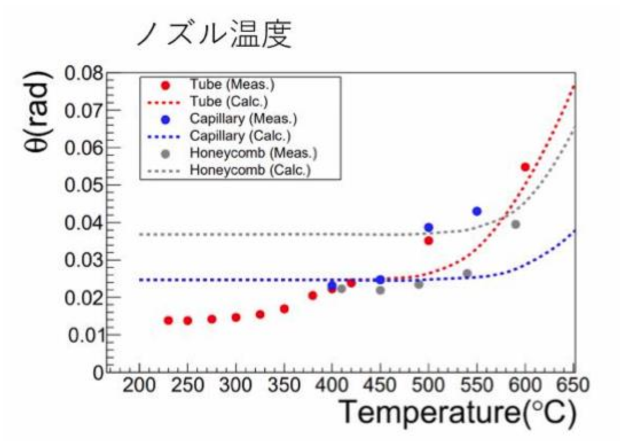
Future developments

Nozzle development

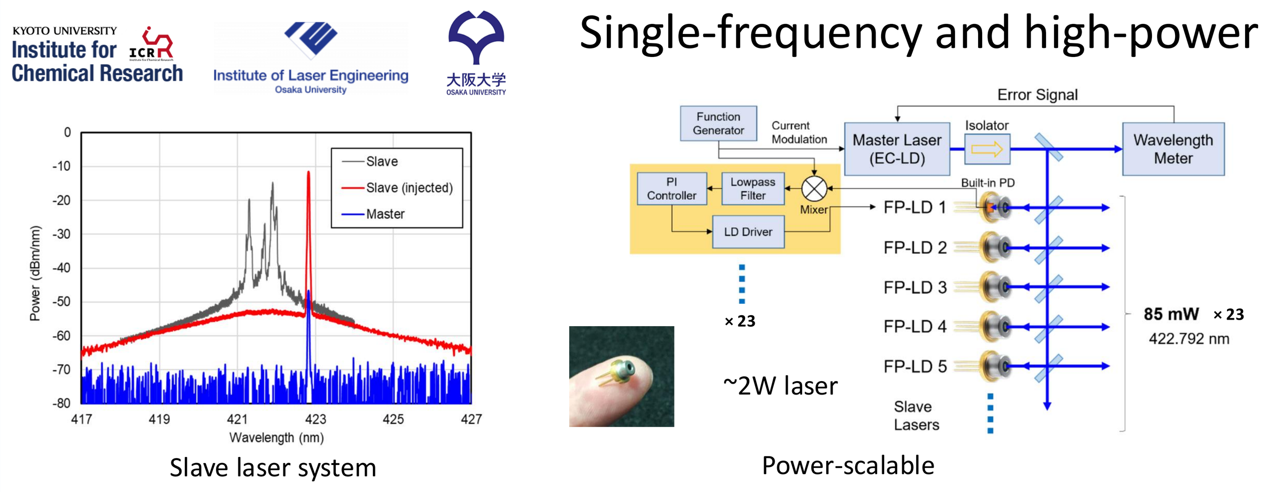


Target: A sheet-like atomic beam.



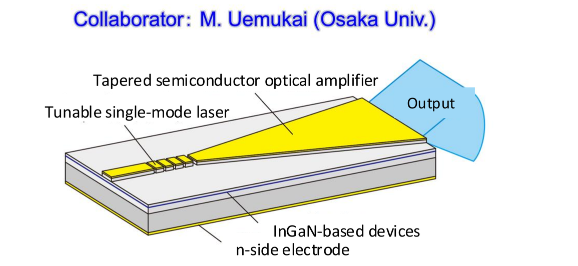


Single-frequency and high-power laser



Development of tapered semiconductor optical amplifier (SOA)

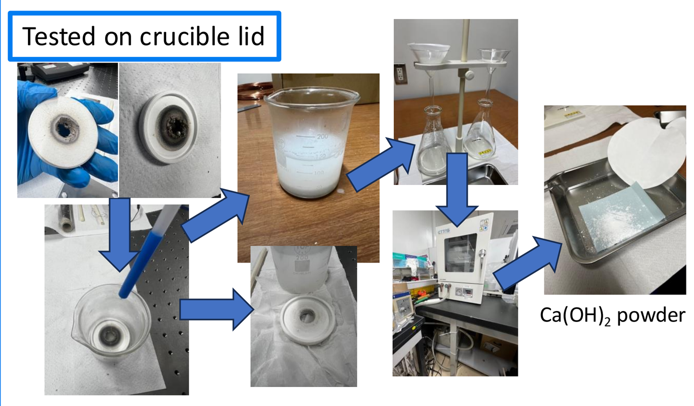
Collaborator: M. Uemukai (Osaka Univ.)



Integrated single-frequency master laser and power amplifier system.

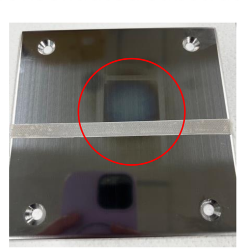
Calcium recovery method : chemical method

Tested on crucible lid



$Ca + 2H_2O \rightarrow 2Ca(OH)_2 + H_2$
Expose to air: $2Ca + O_2 \rightarrow 2CaO$
$CaO + CO_2 \rightarrow 2CaCO_3$
Reacte with DDW: $CaO + H_2O \rightarrow Ca(OH)_2$
Sampling for ICP-MS measurement:
$Ca(OH)_2$+ 2HCl → $CaCl_2 + 2H_2O$
$CaCO_3$ + 2HCl → $CaCl_2 + CO_2 + H_2O$

Consolidated substrate



Expose to air to form CaO

Soak with water in an ultrasonic cleaner to get $Ca(OH)_2$

Settling and filtration of the powder and drying it in an oven heater

Sampling and dissolving it in HCl to get $CaCl_2$ (aq) and measure the isotope ratio by ICP-MS

Full operation of 6 ports for 2 mols/year