The Status of ^{48}Ca enrichment by Laser Isotope Separation (LIS) at RCNP

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The Research Center for Nuclear Physics (RCNP) is currently developing an enrichment process for ^{48}Ca using Laser Isotope Separation (LIS), aimed at the study of neutrinoless double beta decay ($0\nu\beta\beta$) by the CANDLES experiment. This report presents the current status of the isotope separation achieved via LIS and outlines future development prospects for the system.

Double Beta Decay Study by CANDLES

To explore the origin of the matter-dominated universe and investigate lepton number non-conservation, the CANDLES experiment searches for neutrinoless double beta decay ($0\nu\beta\beta$) using ^{48}Ca, which possesses a uniquely high Q-value of 4.3 MeV. Recent results from the CANDLES III system—utilizing 96 high-purity $^{nat}CaF_2$ crystals immersed in a liquid scintillator (0.35 kg of ^{48}Ca)—combined with Monte Carlo (MC) simulations, established a lower limit for the $0\nu\beta\beta$ half-life. Based on 21 selected high-purity crystals, the limit was determined to be 5.6×10^{22} years at a 90% confidence level (C.L.) [1]. However, natural calcium contains only 0.187% of ^{48}Ca. Therefore, significant enrichment is essential for the $0\nu\beta\beta$ search, as a signal-to-background ratio greater than unity at 4.3 MeV is expected only when the isotopic enrichment exceeds 50%. Conventional industrial-scale separation methods, such as gaseous diffusion and centrifugation, are inapplicable to calcium due to its chemical properties. Consequently, ^{48}Ca remains expensive and is mainly available by electromagnetic separation, which yields an annual production rate of only tens of grams. To overcome these limitations, several alternative separation techniques have been proposed, including chemical isotope exchange using macrocyclic polyethers [2, 3], ion-exchange chromatography [4, 5], electrophoresis [6], liquid centrifugation [7], and laser isotope separation (LIS) [8, 9, 10].

Laser Isotope Separation for ^{48}Ca

In this report, we describe an experiment conducted to directly observe the isotope shift of ^{48}Ca and its enrichment via the laser deflection method. This process utilizes the calcium resonance transition ($^1S_0 \rightarrow {}^1P_1$) at a wavelength of 422.7918 nm.

Figure 1 shows a schematic diagram of the experimental chamber. The operating pressure was maintained within the 10^{-4} Pa range. A crucible containing 2 g of metallic calcium (99% purity) is located at the base of the assembly. Under high vacuum, heating the crucible generates a calcium atomic beam, which is subsequently collimated by a series of slits and a tube collimator. The central section of the chamber features six irradiation ports, with a single port currently utilized for operation. A tunable external cavity laser diode (EC-LD; TOPTICA DLC DL pro) serves as the deflection laser, irradiating the atomic beam perpendicularly. The laser wavelength is precisely controlled via feedback from a wavelength meter (HighFinesse WS-7-60).

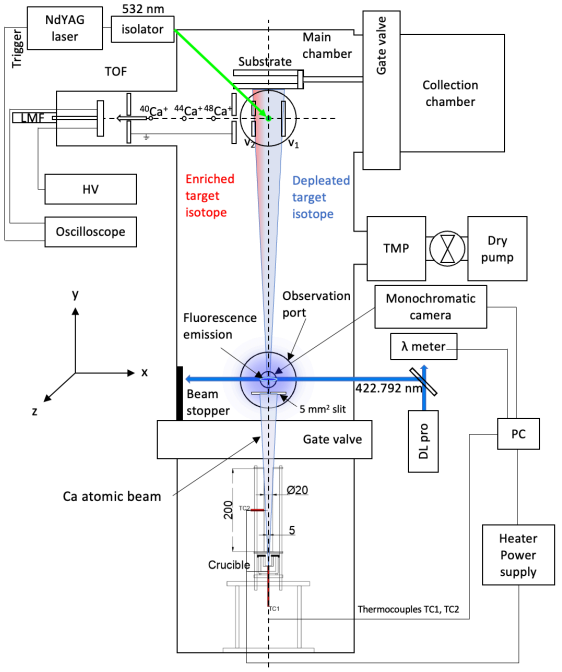


Figure 1: Schematic diagram of the chamber for the production of ^{48}Ca by Laser Isotope Separation (LIS).

For detection, a monochromatic CCD camera is mounted at the observation port to monitor spontaneous emission and identify isotope shifts. The upper section of the chamber is coupled with a time-of-flight (TOF) mass spectrometer, utilizing the second harmonic of an Nd:YAG laser (532 nm) as the ionization source. Both the TOF system and the Nd:YAG laser are movable along the x-axis to enable spatial measurement of the calcium atomic beam and its isotopic components.

Results

A monochromatic CCD camera, equipped with a band-pass filter and a neutral density (ND) filter, was employed to measure the fluorescence from spontaneous emission. The calcium isotope shifts were identified by scanning the laser wavelength around 422.7918 nm in increments of 16.8 MHz (approximately 10 fm). Figure 2 presents the fluorescence intensity (in counts) as a function of frequency, with the ^{40}Ca peak normalized to the center. The experimental data points were fitted using a Voigt profile. At a crucible temperature of 500 °C, and considering the geometry of the collimator and slits, the Doppler broadening characterized by the Gaussian width (σ) was determined to be 20.06 ± 0.01 MHz. Concurrently, the power broadening, represented by the Lorentzian width (γ), was measured at 64.99 ± 0.02 MHz, with laser beam diameters of $w_z = 5$ mm and $w_y = 20.0$ mm. The measured isotope shifts for ^{42}Ca, ^{44}Ca, and ^{48}Ca were 404.21 ± 0.12 MHz, 787.76 ± 0.04 MHz, and 1528.00 ± 0.17 MHz, respectively. These values are in good agreement with the previously reported literature [11, 12].

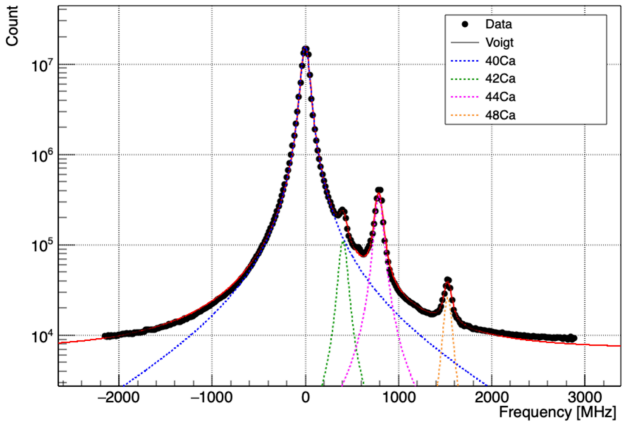


Figure 2: Calcium isotope shift of the $^1S_0 \rightarrow {}^1P_1$ transition obtained by fluorescent measurement.

The effect of the laser diode (tuned to the 422.7918 nm resonance for the ^{48}Ca shift) on the atomic beam was evaluated by comparing the respective TOF spectra. TOF measurements were performed across a spatial range of 0–90 mm, with the beam center defined at $x = 50$ mm. Figure 3 presents the spectra obtained at $x = 25$ mm as a representative case, demonstrating the laser effect on the selective separation of ^{48}Ca.

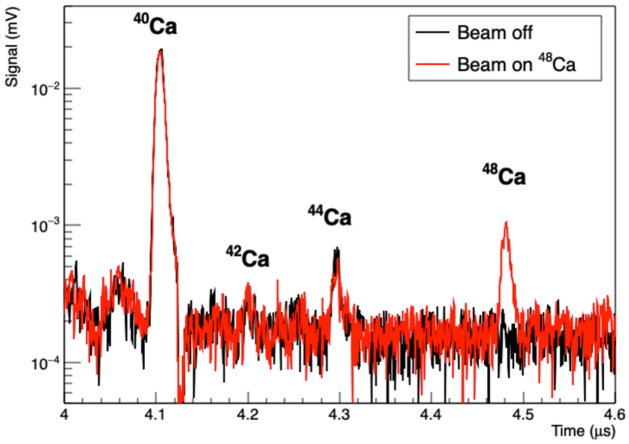
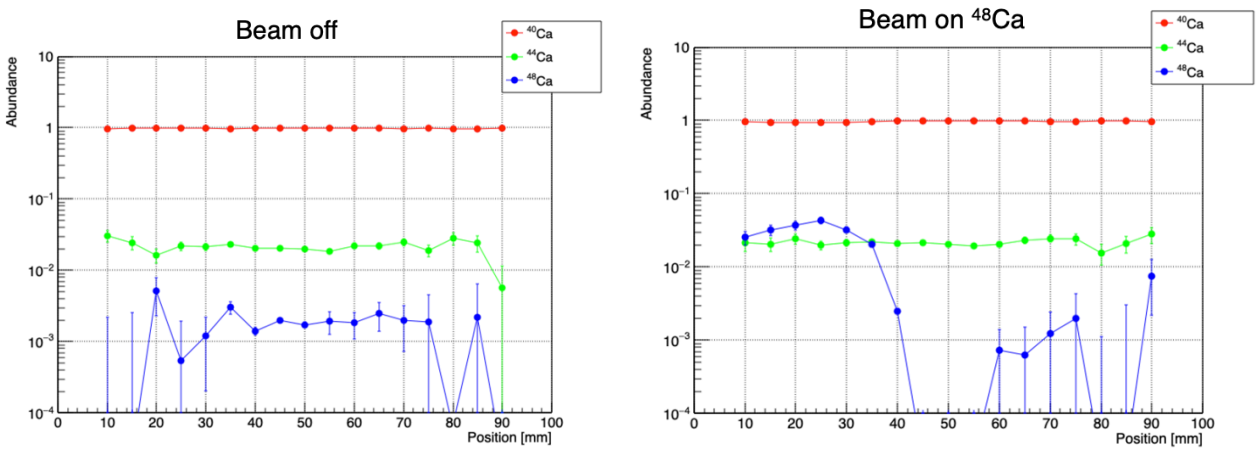


Figure 3: Comparison of time-of-flight (TOF) spectra measured with the laser diode on (tuned to ^{48}Ca shift: Beam on ^{48}Ca) and off (Beam off) at the position of x = 25 nm.



(a) (b)

Figure 4: Isotopic abundance of calcium as a function of spatial position: (a) laser diode off, and (b) laser diode on, with the wavelength tuned to the ^{48}Ca resonance. The laser beam is incident from the $+x$ side, propagating toward the negative x direction.

The TOF spectra acquired at spatial increments of 5 mm were analyzed by integrating the peak areas and applying background corrections. Figure 4 presents the relative isotopic abundances for ^{40}Ca, ^{44}Ca, and ^{48}Ca. Specifically, Figure 4a shows the natural abundances with the laser diode off, while Figure 4b illustrates the distributions with the laser tuned to the ^{48}Ca resonance. When the laser is active, a significant enrichment of ^{48}Ca is observed within the spatial range of 10–40 mm. The maximum abundance of ^{48}Ca was measured at $x = 25$ mm, reaching $4.3 \pm 0.4\%$, which represents an enrichment of approximately 23 times the natural abundance. These results provide clear evidence of the laser deflection of ^{48}Ca from the central beam axis.

The measured isotope ratios were compared with the natural abundance ratio (^{48}Ca/^{40}Ca). Given the natural abundances of 96.941% for ^{40}Ca and 0.187% for ^{48}Ca, the reference ratio is 0.00193. As shown in Figure 5, the isotope ratios (IR) obtained with the laser beam off are in excellent agreement with the natural abundance ratio within the experimental uncertainty.

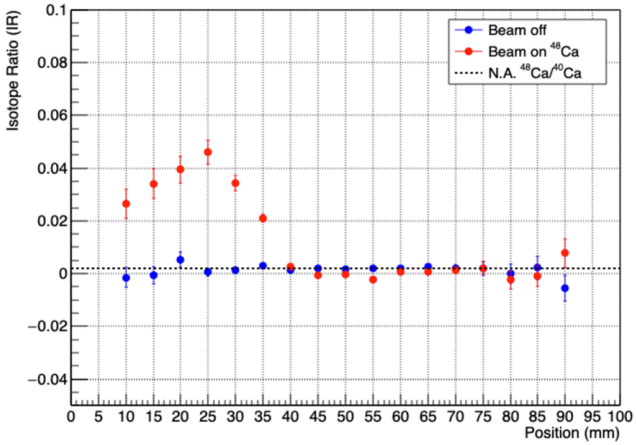


Figure 5: Isotope ratio of ^{48}Ca/^{40}Ca across the calcium atomic beam, measured by TOF

Discussion and prospect for the mass production

The selective separation of ^{48}Ca via the laser deflection method has been successfully demonstrated for the first time. The experimental framework combined isotope shift characterization through fluorescence spectroscopy with direct isotopic composition analysis using TOF mass spectrometry. To further enhance the separation coefficient of ^{48}Ca, current efforts are focused on optimizing the slit and collimator geometries. These improvements aim to deliver a calcium atomic beam with a higher intensity and a narrower angular distribution. As observed in Figure 4, the beam edges still contain residual components of ^{40}Ca and ^{44}Ca; however, the implementation of additional collimation stages is expected to suppress these angular distributions and significantly improve

the ^{48}Ca separation factor. Such enhancements are critical for achieving the mass production of ^{48}Ca targets with at least 50% enrichment, a key requirement for the next-generation CANDLES experiment.

The requirements for large-scale mass production encompass the development of a high-performance atomic beam generator, a high-power laser system, a collection and recovery system, and an integrated monitoring and control framework. Currently, the atomic beam generator is under development at the University of Fukui. The influence of the single-tube collimator length on the resulting beam profile has been previously detailed in refs. [9, 10]. Furthermore, capillary tube collimators are being evaluated as promising candidates to suppress the angular distribution while maintaining high beam intensity. Progress on the single-frequency, high-power laser system has been reported by Kyoto University [13], where a 2 W-class system was fabricated. This system utilizes 23 injection-locked Fabry-Pérot laser diodes (FP-LDs), achieving a relative frequency stability of less than 1 MHz rms. Regarding the recovery process, a system was installed above the TOF assembly to facilitate the deposition of separated isotopes onto a substrate. We have successfully produced the first enriched sample onto the silicon wafer substrate. The enriched ^{48}Ca deposited on the Si substrate was characterized using the stylus method for thickness mapping and TOF-SIMS for surface isotopic ratio analysis. The ongoing experiments aim to identify the substrate materials to maximize collection efficiency. Furthermore, the recovery process will incorporate chemical synthesis methods to convert the collected calcium into $^{48}CaF_2$ crystals, suitable for CANDLES detector. Under full operational capacity with 2 W of laser power, our system is projected to produce up to 2 mol/year of ^{48}Ca.

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