

Status for the Enrichment of ^{48}Ca by Laser Isotope Separation (LIS) at RCNP

A. Rittirong

Research Center for Nuclear Physics (RCNP), Osaka University, Ibaraki, Osaka 567-0047, Japan

RCNP is now developing the production system for the enrichment of ^{48}Ca by laser Isotope separation (LIS) toward the study of neutrinoless double beta decay by the CANDLES project.

Double Beta Decay Experiment at CANDLES

One of the most powerful methods for verifying the mysteries of the universe, such as the matter-dominated universe, and lepton number non-conservation, is neutrinoless double beta decay ($0\nu\beta\beta$). CANDLES (CALium fluoride for studies of Neutrino and Dark matters by Low Energy Spectrometer) aims to investigate this rare event using ^{48}Ca , which has the highest Q-value (4.3 MeV) among the double beta decay nuclides. In the recent study by the CANDLES III system, 96 of high-purity $^{nat}\text{CaF}_2$ crystal surrounded by the liquid scintillator (0.35 kg of ^{48}Ca) and Monte Carlo (MC) simulation, revealed the lower limit of the half-life of $0\nu\beta\beta$ obtained by 21 selected high-purity crystals was 5.6×10^{22} year (90% C.L.) [1].

Laser Isotope Separation for ^{48}Ca

As mentioned in the previous section, CANDLES III used natural calcium, which contains only 0.187% of ^{48}Ca . The enrichment of ^{48}Ca is required to improve the detector sensitivity. The signal-to-background ratio at 4.3 MeV is expected to be greater than one when total isotope enrichment exceeds 50%. However, calcium has no gaseous compound, and industrial-scale isotope separation methods such as gas diffusion and gas centrifuge are inapplicable. The only currently available method is the electromagnetic separator, which has a low annual production yield and is a costly material (1,000,000 \$/g). Other methods for the enrichment of calcium are proposed, including chemical isotope exchange using macrocyclic polyether [2, 3], ion exchange chromatography [4, 5], electrophoresis [6], and laser isotope separation (LIS) [7, 8].

In this paper, we focus on the development of laser isotope separation. K. Matsuoka and I. Ogawa [7, 8] conducted the proof-of-principle experiment. The deflection laser was irradiated perpendicular to the calcium atomic beam. The target calcium isotope absorbed the momentum of the incoming photon, which has a stable (less than 2 MHz rms) continuous wave oscillation at a calcium absorption wavelength of 422.792 nm and was deflected from the original atomic beam. The separation of ^{48}Ca and the calcium recovery were up to 5.5% and 19.6% at the deflection angle of 12.5 mrad, respectively. Figure 1a shows the time variation of the oscillation wavelength of injection-locked laser system was measured over time, with the EC-LD being stabilized by PDH method [9]. The multiple slave system was capable of power scalability of laser system, as shown in Figure 1b.

The requirements for mass production consist of the development of an atomic beam generator, irradiation unit, collection and recovery system, and monitor and control system. The atomic beam generator is being developed by the University of Fukui. The effect of the length of the single-tube collimator on the atomic beam

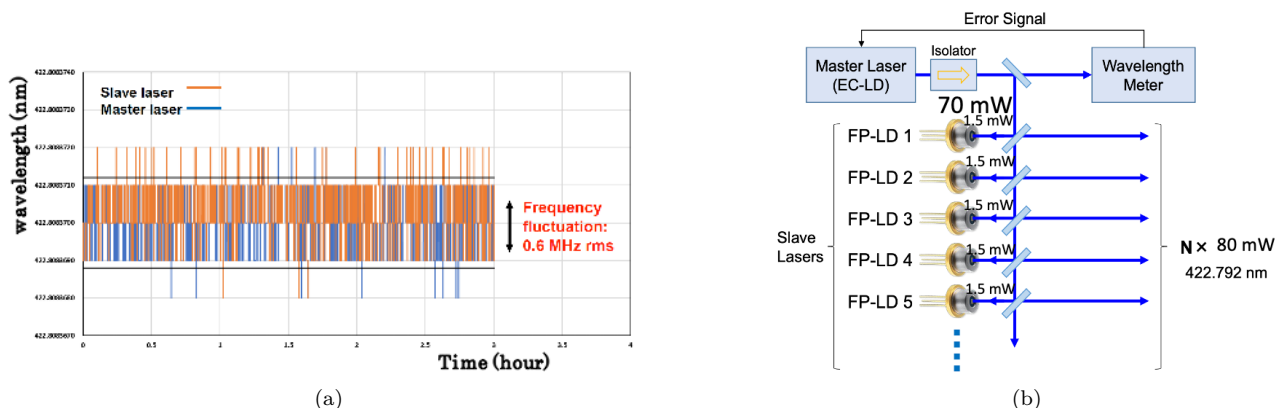


Figure 1: Deflection laser system for the separation of ^{48}Ca , the time variation of the oscillation wavelength of the injection-locking laser measured by a wavelength meter (a), and the power scalability of the laser system to achieve high power output using multiple slave lasers(b).

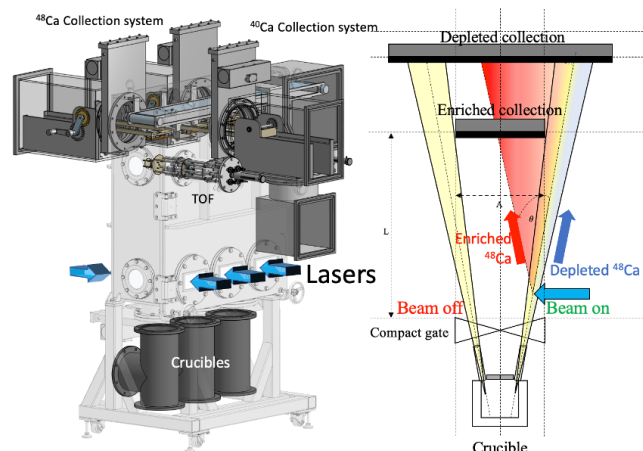


Figure 2: The schematic diagram of the mass production system of laser isotope separation for ^{48}Ca , including crucible, collection system for depleted and enriched stream, laser flanges, and TOF tunnel

shape was reported in Ref. [8]. In addition, the capillary tube collimator is now developing to reduce the spatial distribution and maintain the atomic beam intensity. On the other hand, the recovery system is now being developed to find the most applicable material to be used as a collection plate. The small chamber was introduced to expose the material to the calcium atomic beam. The calcium layer thickness measurement was conducted by optical measurement and the stylus method.

For future development, we introduced the new irradiation chamber consisting of 6 irradiation ports aiming to produce up to 2 mol/year of ^{48}Ca by the full operation of six ports, as shown in Figure 2. The first step is to get stable operation from one of the six ports with the 2 W laser power. The atomic beam intensity and isotope fractionation were measured by the time of flight (TOF) system. The collection system is designed to be automated for 24-hour operation via the conveyor belt and the collection plate at the center of the chamber.

References

- [1] S. Ajimura, W. Chan, K. Ichimura, T. Ishikawa, K. Kanagawa, B. Khai, T. Kishimoto, H. Kino, T. Maeda, K. Matsuoka, *et al.*, “Low background measurement in candles-iii for studying the neutrinoless double beta decay of ca 48,” *Physical Review D*, vol. 103, no. 9, p. 092008, 2021.
- [2] A. Rittirong, T. Yoshimoto, R. Hazama, T. Kishimoto, T. Fujii, Y. Sakuma, S. Fukutani, Y. Shibahara, and A. Sunaga, “Isotope separation by dc18c6 crown-ether for neutrinoless double beta decay of 48ca,” in *Journal of Physics: Conference Series*, vol. 2147, p. 012015, IOP Publishing, 2022.
- [3] B. Jepsen and R. DeWitt, “Separation of calcium isotopes with macrocyclic polyether calcium complexes,” *Journal of Inorganic and Nuclear Chemistry*, vol. 38, no. 6, pp. 1175–1177, 1976.
- [4] S. Umehara, T. Kishimoto, H. Kakubata, M. Nomura, T. Kaneshiki, T. Suzuki, Y. Fujii, and S. Nemoto, “A basic study on the production of enriched isotope 48ca by using crown-ether resin,” *Progress of Theoretical and Experimental Physics*, vol. 2015, no. 5, 2015.
- [5] S. Nemoto, K. Suga, Y. Fukuda, M. Nomura, T. Suzuki, and T. Oi, “Calcium isotope fractionation in liquid chromatography with crown ether resins,” *Journal of nuclear science and technology*, vol. 49, no. 4, pp. 425–437, 2012.
- [6] T. Kishimoto, K. Matsuoka, T. Fukumoto, and S. Umehara, “Calcium isotope enrichment by means of multi-channel counter-current electrophoresis for the study of particle and nuclear physics,” *Progress of Theoretical and Experimental Physics*, vol. 2015, no. 3, 2015.
- [7] K. Matsuoka, H. Niki, I. Ogawa, Y. Shinki, Y. Kawashima, and K. Matsumura, “The laser isotope separation (lis) methods for the enrichment of 48ca,” in *Journal of Physics: Conference Series*, vol. 1468, p. 012199, IOP Publishing, 2020.
- [8] I. Ogawa, Y. Kawashima, T. Hiraiwa, M. Tozawa, H. Niki, S. Tokita, B. Han, H. Okuda, N. Miyana, S. Umehara, *et al.*, “Development of the laser isotope separation method to study for the neutrino-less double beta decay of 48ca,” in *Journal of Physics: Conference Series*, vol. 2147, p. 012012, IOP Publishing, 2022.
- [9] R. W. Drever, J. L. Hall, F. V. Kowalski, J. Hough, G. Ford, A. Munley, and H. Ward, “Laser phase and frequency stabilization using an optical resonator,” *Applied Physics B*, vol. 31, pp. 97–105, 1983.