PROCEEDINGS OF SCIENCE

The Status of ^{48}Ca Production by Laser Isotope Separation for the Study of Neutrinoless Double Beta Decay

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CANDLES used ^{48}Ca for the search of neutrinoless double beta decay ($0\nu\beta\beta$). However, a large amount of ^{48}Ca is required to sense this ultra-rare event. Laser isotope separation (LIS) provides feasibility for separating the target isotope by selectively exciting the target isotope with a high-power and single-frequency laser. The new design of the irradiation unit was introduced, and the first step is to achieve a stable production system from one of the six atomic beam generators by 2W laser power at 422.792 nm oscillation wavelength. The proper design of the upscale production system was developed, including a stable atomic beam generator, power scalability of the laser system, a collection system, and a monitor and control system. The first milestone is achieving a 2 mol/year production rate.

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1. Double Beta Decay Experiment at CANDLES

Neutrinoless double beta decay (0v$\beta\beta$) is a powerful method for investigating the mysteries of the universe, such as the Majorana nature of neutrino, the matter-dominated universe, and lepton number non-conservation. In this context, the CANDLES (CAlium fluoride for studies of Neutrino and Dark matters by Low Energy Spectrometer) experiment focuses on studying this exceptionally rare event utilizing the isotope calcium-48 (^{48}Ca), which possesses the highest Q-value (4.3 MeV) among double beta decay nuclides. In a recent study conducted by the CANDLES III, 96 high-purity calcium fluoride ($^{nat}CaF_2$) crystals (305 kg) were employed, corresponding to a total mass of 0.35 kg of ^{48}Ca. These crystals were surrounded by a liquid scintillator, and the experimental data were analyzed alongside Monte Carlo (MC) simulations. The analysis revealed a lower limit for 0v$\beta\beta$ half-life based on 21 selected high-purity crystals, resulting in a value of 5.6×10^{22} years. Additionally, the effective Majorana neutrino mass $\langle m_\nu \rangle$ was determined to be $\leq$ 2.9 to 16 eV (90% C.L.) [1].

2. Laser Isotope Separation (LIS) for the enrichment of ^{48}Ca

Natural calcium contains only 0.187% of ^{48}Ca. The enrichment of ^{48}Ca is required to improve the detector sensitivity in the search of 0v$\beta\beta$. To explore the effective neutrino mass in the inverse hierarchy region (0.03 eV), approximately 0.36 - 13 kmol of ^{48}Ca is required, as estimated by the nuclear matrix elements [2] and based on the assumption of background-free conditions, a total detection efficiency of 0.8, and a measurement time of 10 years. However, calcium has no gaseous compound, and industrial-scale isotope separation methods such as gas diffusion and gas centrifuge are inapplicable. The only available method is the electromagnetic separator, which has a low annual production yield and leads to a costly material ($\approx$ 1,000,000 USD/g). Other methods for the enrichment of calcium are proposed, including chemical isotope exchange using macrocyclic polyether [3], ion exchange chromatography [4], electrophoresis [5], and laser isotope separation (LIS) [6, 7]. Laser isotope separation (LIS) is based on the isotope shift of ^{48}Ca. The proof of principle experiment revealed that the blue-light laser diode at the oscillation wavelength of 422.792 nm, could deflect ^{48}Ca from the atomic beam. The enrichment of ^{48}Ca and the recovery were up to 5.5% and 19.6% at the deflection angle of 12.5 mrad, respectively.[6] Our development goals for the production system include a stable atomic beam generator, single-frequency and power-scalable laser diode, large-scale irradiation unit, collection and recovery system, and a monitor and control system. These efforts aim to optimize performance, improve efficiency, and ensure long-term stability.

We are developing a stable atomic beam generator, focusing on generating a sheet-like atomic beam, as reported in Ref.[7]. To achieve this improvement, we employ the crucible equipped with three collimator tubes, which play a crucial role in collimating and directing the atomic beam. To further enhance the performance of our sheet-like atomic beam generator, we are considering the implementation of microchannel capillary tubes. This technique reduces inter-atomic scattering and maintains high beam intensity with minimal divergence angle, as explained in Ref.[8]. We are also developing a simulation code to analyze the interaction between calcium atoms, laser photons, collimators, and other structural components. This simulation plays a crucial role in understanding

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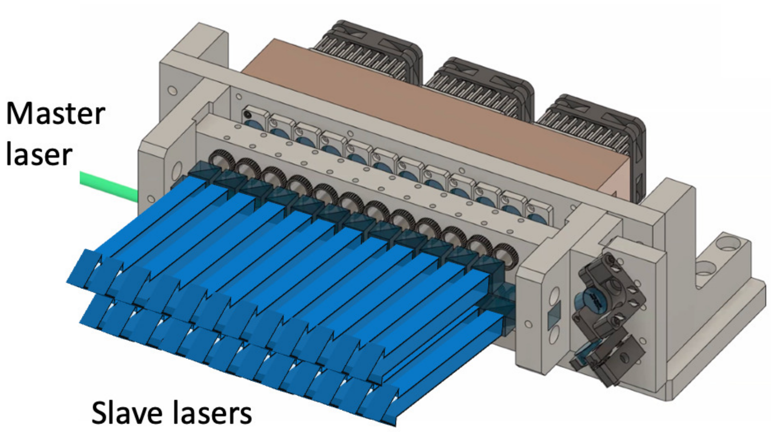

Figure 1: A beam splitter device for multiple FP-LDs to produce laser power up to 2W, with controlled wavelength via injection-locking technique.

the dynamics of our atomic beam generator and helps optimize its design and performance. To achieve the large-scale production of ^{48}Ca for $0\nu\beta\beta$ study, the requirement of laser components consists of a stable and single frequency (< 2 MHz rms), power scalability (from 10 to 1000 W), long lifetime, and cost-effectiveness. GaN laser diode as a light source could achieve both stable oscillation frequency and power scalability with semiconductor optical amplifiers (SOAs) increasing the power per element. However, the high cost is inevitable. Alternatively, we used multiple slaves laser (Fabry-Perot laser diodes: FP-LDs) whose wavelength could be controlled by a single master laser (external cavity laser diode: EC-LD), called the injection-locking technique. The relative frequency deviation of the slave laser, which is stabilized by the Pound-Drever-Hall (PDH) signal, was measured to be 0.6 MHz rms over 3 hours. This value is significantly smaller than the natural broadening observed in the Ca atomic beam. Figure 1 shows the device that increases the number of FP-LD lasers. As a result, 24 FP-LDs could achieve around 2 W of laser power by a single master laser. Approximately 200 W of laser power can be realized using ~2500 slave lasers and one master laser. In the research and development of the collection and recovery system, investigations are underway to identify the most effective materials for calcium collection. A small chamber equipped with a commercial crucible is being utilized in this research. In the evaluation process, the thickness of the calcium films is measured using a calcium thickness meter employing the stylus method. Multiple candidate materials are being explored and assessed to determine their coating efficiency. This investigation aims to optimize the performance of the collection and recovery system by selecting the most suitable material for this crucial task.

An essential aspect of mass production involves the development of a large-scale irradiation unit, which holds an important role in the production process. Figure 2 presents a schematic drawing of the irradiation unit comprising six atomic beam generators. The deflection laser is irradiated perpendicular to the vertically generated atomic beam. ^{48}Ca was separated and collected at the center of the vacuum chamber by the collection plate. Moreover, the isotope composition was measured by the time of flight (TOF) with the NdYAG ionization laser as a trigger. At the same time, a large amount of depleted ^{48}Ca is carefully removed using the conveyor belt system. Managing the depleted ^{48}Ca stream is important since each 21.4 kg of calcium yields approximately 1 mol

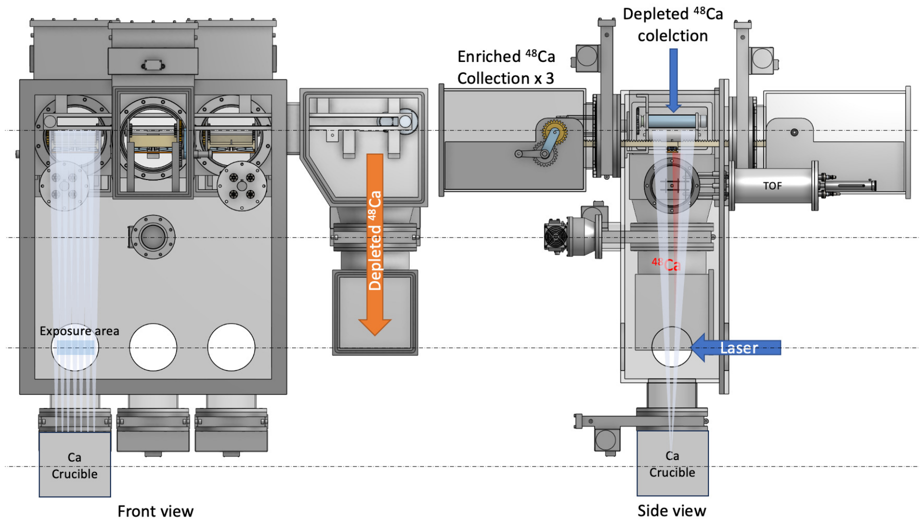

Figure 2: Schematic diagram of the irradiation unit for a large-scale production system (2 mol/year). (Left) Front view, (Right) Side view

of ^{48}Ca. Hence, efficient handling and removal of the depleted stream is crucial for the smooth operation and optimization of the mass production process. This irradiation unit is expected to produce up to 2 mol/year of ^{48}Ca. The first step is to achieve the stable operation of the first port with 2 W of laser power, resulting in approximately 10 grams of production rate per year.

3. Summary

Laser isotope separation (LIS) provided feasibility for the enrichment of ^{48}Ca in a large-scale manner toward the study of neutrinoless double beta decay of CANDLES. The development of a comprehensive large-scale production system with various components, including the atomic beam generator, single frequency and power-scalable laser, automated collection system, and irradiation unit. An atomic beam generator was developed toward the sheet-like atomic beam with a well-collimated (small divergence angle) and high-intensity beam. Twenty-four slave lasers wavelength were controlled by one master laser through an injection-locking technique along with the PDH method, providing up to 2 W of laser power. Research on appropriate materials for consolidating calcium is currently underway. The new irradiation unit was introduced. The first step is focusing on achieving stable operation from the first port with 2 W laser power, which has the potential to produce up to 10 grams of enriched ^{48}Ca per year. By scaling up the system to incorporate all six ports, production is projected to reach up to 2 mol of enriched ^{48}Ca per year. The ultimate goal is to achieve a production rate in the kmol scale by increasing laser power per unit and expanding the irradiation chamber, thus paving the way for ton-scale production.

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