Development of the laser isotope separation method to study for the neutrino-less double beta decay of ^{48}Ca

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Abstract. Search for the ultra-rare process, neutrino-less double beta decay, is a powerful tool to test the Majorana nature of neutrinos. Among the potential double beta decay nuclei, ^{48}Ca has the largest Q-value, hence we can expect the measurement with least background. On the other hand, due to its low natural abundance, isotope enrichment is essential to achieve the sensitivity in the region of interest (inverted/normal mass hierarchy of neutrinos).

We have been developing a laser isotope separation method using a tunable semiconductor laser that can excite only a specific isotope (^{48}Ca). The laser is irradiated perpendicular to the collimated calcium vapor beam. Only ^{48}Ca atoms are separated by deflecting them from the original atomic beam by momentum transfer due to multiple absorption and emission of laser photons. The isotope separation is confirmed by irradiating an ionization laser and time-of-flight measurement.

Following the success of the proof-of-principle experiment, we are now developing the mass production system which include efficient generation of atomic beams, frequency-stabilized high-power laser, efficient laser irradiation method, and collection method of deflected isotope beams. In this paper, we report on the status and prospects of these developments.
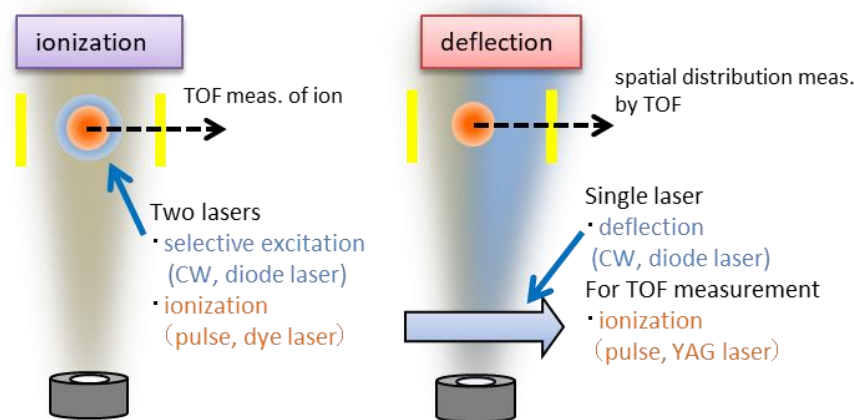
1. Introduction

As an experimental study to verify the Majorana nature of neutrinos, double beta decay experiments without neutrino emission ($0\nu\beta\beta$ decay) are being conducted all over the world [1] . Since $0\nu\beta\beta$ decay is a very rare event (experimental lower limit for its half-life:$T_{1/2}^{\text{limit}} \sim 10^{26}$ yr [1]), it is important to prepare a large number of target nuclei for the experiment in an extremely low background environment. There are many nuclei that can undergo double beta decay, among which ^{48}Ca has the largest Q-value (4.27 MeV) which is higher than those of natural radio-activities (2.6/3.3 MeV for γ/β - rays), hence we can expect the measurement with least background. In the CANDLES experiment, the background level of approximately 10^{-3} events/keV/yr/(kg of ^{nat}Ca), was achieved using about 305 kg of ^{nat}CaF$_2$ scintillator (containing ~7.3 mol of ^{48}Ca) as the main detector [2]. To further improve the sensitivity of experiment, since its natural abundance is small (0.187%), isotope enrichment is essential, along with efforts to reduce the background. For the half-life of 10^{26} years, a

signal-to-background ratio larger than 1 can be expected with one order of magnitude further reduction of the background and isotope enrichment to 50%. Unfortunately, since a gaseous compound of calcium at room temperature is not known, traditional methods such as centrifugation and gas diffusion cannot be used for the enrichment of ^{48}Ca, and some efforts are being made to develop chemical [3] and physical [4] methods.

We have been developed the laser isotope separation (LIS) methods to enrich ^{48}Ca [5]. The wavelength of a certain transition of calcium from the ground state is $\lambda = 422.7$ nm, where a commercially available blue-violet diode laser (DL) can be used to excite. The linewidth of external cavity DL is narrow enough compared to the isotope shift in this transition which is about several hundred MHz.

There are two different separation methods in LIS: ionization and deflection methods shown in figure 1. In the ionization method, a specific isotope is selectively excited by a cw DL, then ionized by a pulsed dye laser, and collected by an electric field. In the deflection method, a well-collimated atomic beam is irradiated perpendicularly with a DL, and the multiple excitation and de-excitation by absorbing and emitting the photons lead to the momentum transfer from the laser photons to the atoms, which are then deflected from the original atomic beam to be collected. We have conducted proof-of-principle (POP) experiments on both methods. The atomic beam was generated by heating a crucible containing metallic calcium to 600°C, using an aperture with a diameter of 2.5 mm and an aperture with a diameter of 1.5 mm placed 20 cm downstream. For the ionization method, we succeeded in enriching ^{48}Ca to 90% with a small duty cycle. For the deflection method we enriched ^{48}Ca to 5.5% with a collection efficiency of 20% [5], where the collection efficiency is the ratio of collected ^{48}Ca to the evaporated one. For the mass production system dedicated to the double beta decay experiments, we have decided to employ the deflection method.

Figure 1. Conceptual diagram of the two types of collection methods for separated atoms in LIS.

2. Production equipment by deflection method

A schematic drawing of production equipment by deflection method is shown in figure 2. A deflection laser is irradiated perpendicularly (x-axis) to the atomic beam (z-axis), and the target isotope is deflected by gaining momentum in the x direction through multiple (N_{ph}) excitations and deexcitations. To prevent contamination of the separated atoms with the original atomic beam, the atomic beam should have almost zero momentum in the x and y directions, and the thickness (d) in the x direction should be small enough to allow separation and collection of enriched atoms. For mass production, it is necessary to generate a sheet-like (wide dimension in y-axis) atomic beam. The wavelength of the laser is stabilized to match the transition of the enriched isotope, and the power is adjusted to be sufficient for the atoms on the opposite side of the irradiated surface in the x-direction
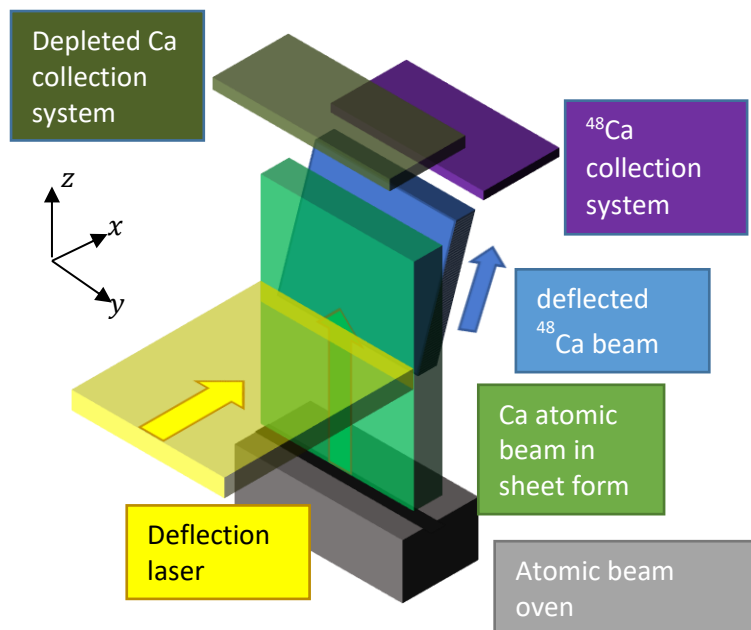
Figure 2. A Schematic drawing of mass production system by deflection method.

of the atomic beam to absorb N_{ph} photons. The collected enriched calcium should be extracted at appropriate time intervals. In addition, a collection system for the depleted calcium is necessary to prevent it from adhering to the inner walls of the equipment. The entire system is monitored and controlled to operate stably over a long period of time.

The mass production system using the deflection method consists of the following parts: 1) atomic beam generator, 2) deflection laser system, 3) laser irradiation unit, 4) collection system for enriched and depleted calcium, and 5) monitor and control system. In order to produce m moles of ^{48}Ca, natural metallic calcium of mass $M = (21.4m/\epsilon_{\text{coll}}) \times 10^3$ g is needed as a raw material, where ϵ_{coll} is the collection efficiency of ^{48}Ca. To achieve the production rate of n (mol/yr), we estimated the performance required for 1) and 2). For 1), the required evaporation rate of natural calcium obtained by dividing M by the actual working time is

$$v_{ev}(\text{g/sec}) = 0.27 \left(\frac{n}{100 \text{ mol/yr}}\right) \left(\frac{\epsilon_{\text{coll}}}{0.5}\right)^{-1} \left(\frac{\tau}{0.5}\right)^{-1},$$

where $\tau(0 < \tau < 1)$ is duty factor of the system. To achieve this value of $v_{ev} = 0.27$ g/sec, required surface area S of the calcium evaporation source in a furnace is $S = 0.27$ m^2, for the furnace temperature of $T = 870$ K. If each atom absorbs and emits $N_{ph} = 1000$ laser photons, transferred momentum in x-ais is $p_x = hN_{ph}/\lambda = 1.6 \times 10^{-24}$kg m/sec, which leads the deflection angle of $\theta \sim$ 30 mrad. The m moles of ^{48}Ca absorb the energy of $\varepsilon = N_{ph} \times hc/\lambda \times N_A = 2.83 \times 10^3$ J in this process. For 2), the required laser power P (kW) is given by

$$P \text{ (kW)} = 3.52 \left(\frac{n}{100 \text{ mol/yr}}\right) \left(\frac{\epsilon_{\text{las}}}{1.0}\right)^{-1} \left(\frac{\epsilon_{\text{coll}}}{0.5}\right)^{-1} \left(\frac{\tau}{0.5}\right)^{-1},$$

where ϵ_{las} is the laser utilization efficiency.

2.1. Atomic beam generator

The concept of the atomic beam generation is the same as that of the POP experiment, i.e., the furnace containing the natural metallic calcium is heated by a heater in order to ensure stable long-term operation and easy maintenance for mass production. The capacity of the furnace was increased to about 50 times that of the POP experiment. The atomic beam flux can be stabilized by controlling the temperature of the furnace and is monitored by a film thickness gauge during operation. For beam collimation, we decided to use a combination of nozzles and tubes instead the slits used in the POP experiment to avoid wasting raw materials.
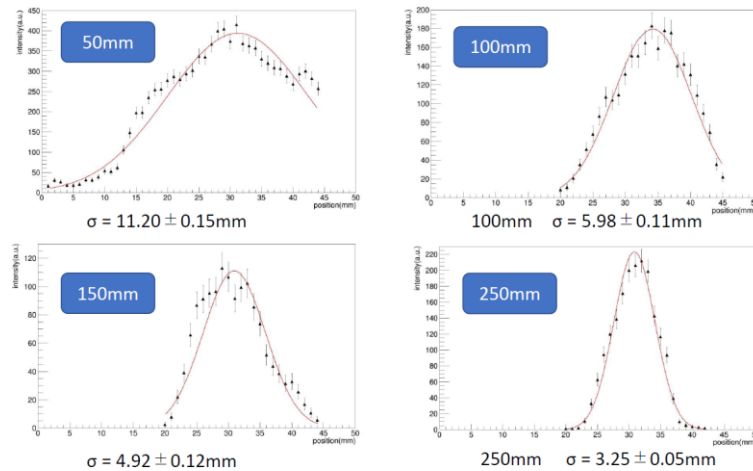
Figure 3. Measurement of collimation effect by circular tubes with 5 mm inner diameter and different lengths (50, 100, 150 and 250 mm). The spatial distribution of the atomic beam was measured while moving the ionization laser (YAG) along the y-axis. The distance from the furnace to the ionization position was kept constant.

We studied the effect of the length of a single circular tube without a nozzle on the atomic beam spread. The spatial distribution of the atomic beam was measured by irradiating an ionization laser (pulsed YAG laser with a beam diameter of ~1 mm) perpendicular to the atomic beam and shifting the irradiation position (see right figure in figure1). The total charge of the ions was measured by applying a guide electric field to the irradiated area, leading to the microchannel plate detector (MCP). At the same time, the time of flight (TOF) of the ions was measured from the pulsed laser trigger to the ion signal of the MCP, to be confirmed that the ions were calcium. Figure 3 shows an example of the measurements using circular tubes with an inner diameter of 5 mm. By increasing the length of the tube, the spatial spread is reduced. We will investigate the optimum inner diameter, length, and cross-sectional shape of the tubes to create a sheet-like atomic beam by arranging them in the y direction.

2.2. Deflection laser system concept of blue-violet high-power diode laser system
A kilowatt-level high-power continuous-wave laser source with a wavelength of 422.7 nm are required to realize a production facility for enrichment of ^{48}Ca. Gallium nitride (GaN) laser diode (LD) is a most promising laser source for the isotope separation due to its advantages such as high efficiency, compact, long life time, high reliability, and low cost [6]. However, the cost-effective technology to achieve both high-power and single-frequency in laser diodes has not been developed yet. For example, to obtain a total optical power of 1 kW, 100 to 10,000 of LD emitters are required. The wavelength of all emitters should be stabilized to match the absorption line of ^{48}Ca isotope. The absolute accuracy of the wavelength stabilization should be less than a few MHz, because the Doppler broadening of Ca atomic beam is typically ~60 MHz full width at half maximum.
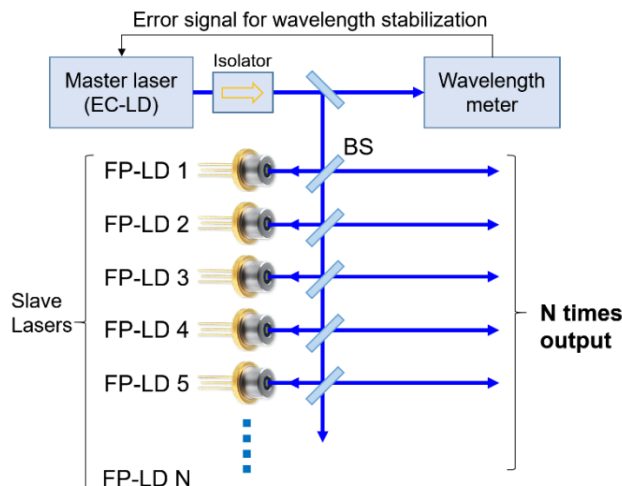

Figure 4. Conceptual diagram of injection-locked multiple Fabry-Perot lasers. EC-LD: External cavity laser diode, FP-LD: Fabry-Perot laser diode, BS: Beam splitter.
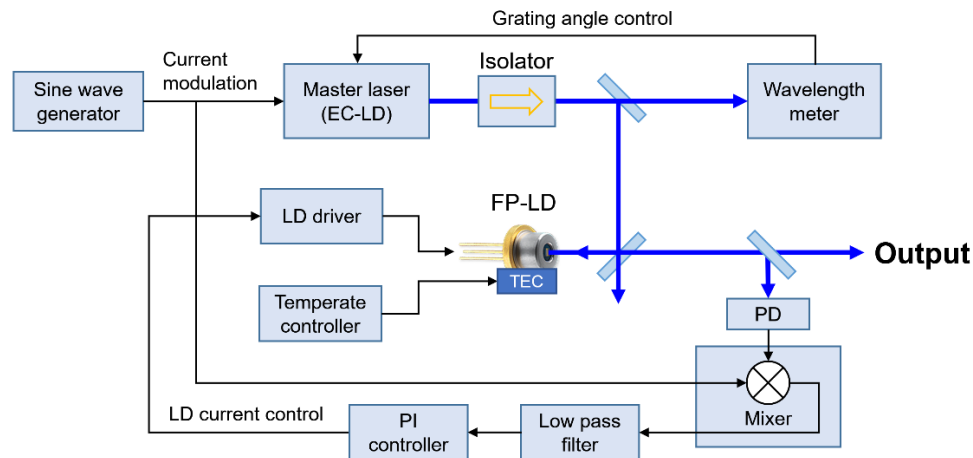
Figure 5. Experimental setup of injection-locked laser system with Pound-Drever-Hall technique. EC-LD: External cavity laser diode, FP-LD: Fabry-Perot laser diode, PD: Photo diode, TEC: Thermoelectric cooler, PI controller: proportional-integral controller.

Semiconductor optical amplifier (SOA) is one of solution to realize such a high-power system [7,8]. Master oscillator power amplifier (MOPA) laser system using multiple SOAs would enable to achieve both high-power and single-frequency. However, since SOA devices based on GaN do not exist yet in general, a high cost is inevitable. Alternatively, injection locking technique would be useful to develop a power-scalable laser system with a low cost [9,10]. In laser injection locking, a weak monochromatic signal from a master laser oscillator is injected into the resonant cavity of a second self-sustained slave laser oscillator. The injected signal from the master can then capture or "lock" the subsequent oscillatory behavior of the free-running slave laser, so that in the end the slave's output is controlled by the injected signal. If the power of the master laser is high enough, multiple slave lasers can be locked by the injected signal dirided from a master laser. In this way, power scaling is possible by increasing the number of slave lasers. Figure 4 shows the conceptual diagram of injection-locked multiple Fabry-Perot lasers. The master laser is an external cavity laser diode (EC-LD) which is wavelength-stabilized into the absorption line of ^{48}Ca by the high-accuracy laser wavelength meter. The output beam from the master laser is divided by beam splitters and injected into multiple Fabry-Perot laser diodes (FP-LDs). Total output power of this laser system is the single device power multiplied by the number of that.

For the injection locking of FP-LDs, the laser frequency of a longitudinal mode should be precisely matched to that of the master laser. The frequency of a longitudinal mode is varied by the temperature and the electric current applied to the laser diode. To lock the frequency of a FP-LD to that of the master laser, the Pound-Drever-Hall (PDH) technique [11] is commonly used. As a basic demonstration, we have performed injection locking and frequency stabilization of a GaN FP-LD. Figure 5 shows the experimental setup of the injection-locked laser system with PDH technique. The master laser is an external cavity laser diode (Toptica, DL pro). The wavelength of the master laser is stabilized to 422.7918 nm (in vacuum) by using a wavelength meter (HighFinesse, WS-7-60) with an absolute accuracy of 60 MHz. The output beam of the master laser is injected into the slave laser which is a Fabry-Perot laser diode with a maximum output power of 120 mW (Nichia, NVA4A16). When a portion of the master laser output (~2 mW) is injected into the slave laser, injection locking is observed. Figure 6 shows the output spectra of the master and slave lasers. The slave laser shows multi-mode oscillation at free-running condition but shows single-frequency oscillation at injection-locking condition. The amplified spontaneous emission (ASE) suppression ratio is higher than 40 dB at the injection-locking condition. It was confirmed that the spectral purity of the slave laser is sufficiently high. The PDH stabilization system for the slave laser consist of a silicon photo diode, a synchronous detection circuit, and a proportional-integral (PI) controller. The sidebands generated by
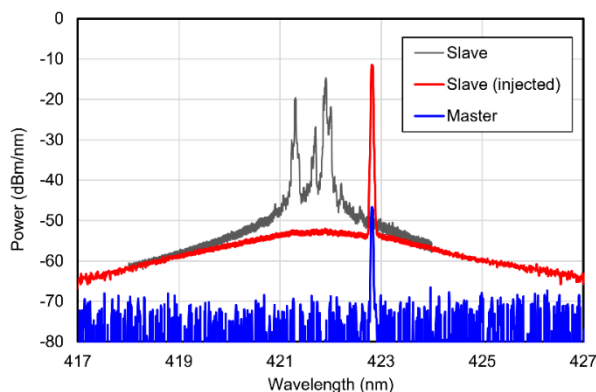
Figure 6. Typical output spectra of master and slave lasers measured by an optical spectrum analyzer.
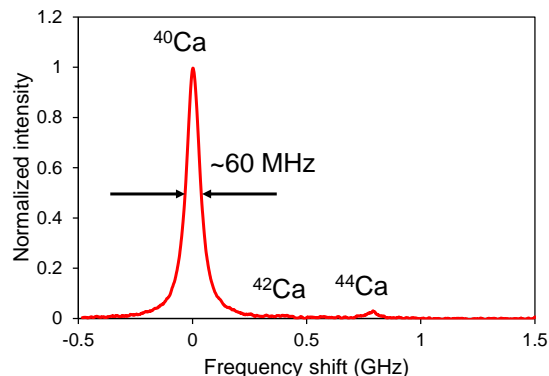
Figure 7. The absorption spectrum of calcium measured with a combination of the injection synchronous laser and the atomic beam generator.

phase modulation of the master laser produce a small amplitude modulation of the slave laser output when the slave laser cavity is detuned. A PDH error signal is generated by detecting the amplitude modulation. The standard deviation of relative frequency variation of the slave laser stabilized by the PDH signal is measured to be 1.2 MHz in 1 hour, which is much smaller than the natural broadening of Ca atomic beam. Thus, it was demonstrated that the narrow linewidth laser system introduced here has sufficient wavelength stability for Ca isotope separation. As a next step, we will increase the number of slave lasers to demonstrate power scalability of the injection-locked laser system.

2.3. Absorption spectrum measurement
To demonstrate the present performance of our system, we measured the absorption spectrum of Ca isotopes using the atomic beam generation system described in section 2.1 and the deflection laser system in section 2.2. The slave laser, synchronized with the master laser whose frequency can be controlled externally, is irradiated onto an atomic beam generated from a furnace heated to about 600°C. A preliminary result is shown in the figure 7 with the laser frequency on the horizontal axis and the fluorescence intensity on the vertical axis. This result shows that it is possible to selectively excite specific isotopes with our laser system.

3. Summary
We have been developing the mass production system for enriched ^{48}Ca using a laser deflection method to study the neutrino-less double beta decay. The development of each part of the system is in progress. In the atomic beam system, beam collimation with tubes is used to reduce the contamination of the separated atoms with the original atomic beam and increase the efficiency of use of metallic calcium. In the laser system, we use multiple LDs, which are relatively inexpensive and available in the market, and achieve high power by the injection-locking method. In order to confirm the performance of the system at this stage, we measured the absorption spectrum with these two systems. We will make further improvements to the atomic beam generator to enable high-efficiency, high-volume beam generation, and the laser system with higher intensity with longer-term stable oscillation. In addition, we will conduct research and development on laser irradiation unit, collection system for enriched and depleted calcium, and monitor-and-control system to finally establish a mass-production system for ^{48}Ca.

References
[1]   Dolinski M.J., Poon A.W.P. and Rodejohann W., 2019 *Ann. Rev. Nucl. and Part. Phys.* **69** 219.
[2]   Ajimura S, Chan W. M., Ichimura K. et al., 2021 *Phys. Rev.* D **103** 092008
[3]   Rittirong A, Noithong P et al. *in this volume*

[4] Kishimoto T, Matsuoka K, Fukumoto T and Umehara S 2015 *PTEP* 2015 033D03
[5] Matsuoka K, Niki H, Ogawa I, Shinki Y, Kawashima Y, Matsumura K 2020 *J. Phys.: Conf. Ser.* 1468 012199
[6] Nakamura S 2000 *JSAP International (The Japan Society of Applied Physics)* **1** 5
[7] Kono S, Koda R, Kawanishi H and Narui H 2017 *Opt. Express* **25** 14926
[8] Stanczyk S, Kafar A, Grzanka S, Sarzynski M, Mroczynski R, Najda S, Suski T and Perlin P 2018 *Opt. Express* **26** 7351
[9] Pagett C J H, Moriya P H, Celistrino Teixeira R, Shiozaki R F, Hemmerling M and Courteille P W 2016 *Rev. Sci. Instrum.* **87** 053105
[10] Hosoya T, Miranda M, Inoue R and Kozuma M 2015 *Rev. Sci. Instrum.* **86** 073110
[11] Drever R W P, Hall J L, Kowalski F V, Hough J, Ford G M, Munley A J and Ward H 1983 *Appl. Phys. B* **31** 97