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Current status of the polarized La target development for T-violation search with slow neutron

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We are addressing the development of a polarized lanthanum (^{139}La) target by applying a standard dynamic nuclear polarization (DNP) technique to a single crystal of lanthanum aluminate ($LaAlO_3$) doped with neodymium ions (Nd^{3+}), which is very attractive to search for violating effects of the time-reversal invariance with neutrons. The use of the $LaAlO_3$ crystals as target material is very crucial to extend the spin-lattice relaxation time (T_1) through the optimization of the Nd^{3+} concentration. We have almost established a method for precise control of the Nd^{3+} amount and this method has allowed us to grow the crystal with 0.01 mol% concentration differently from the 0.05 mol% reached before. Simple DNP experiments using a single-shot dewar at 1.3 K and 2.3 T allowed the evaluation of the T_1 and the maximum polarization of these crystals. By that we have found that the 0.01 mol% concentration is the best one among the crystals tested in the past. Both, T_1 and the maximum polarization of the 0.01 mol% crystal, have not been exactly obtained because of the long relaxation time, Thus, the preparation of a new DNP system is in progress toward DNP experiments for long-term measurement.

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1. Introduction

In compound nuclear resonance of unpolarized ^{139}La nuclei with epithermal neutrons at 0.75 eV, an observable effect of the parity-nonconservation (PNC) becomes 10^6 times larger compared to nucleon-nucleon scattering [1, 2]. A model explaining such an enhancement has suggested that violating effects of time-reversal symmetry are expected to be also enhanced similar to the PNC effect [3, 4]. The NOPTREX collaboration (Neutron OPtics Time Reversal EXperiment) is planning to search for a time-reversal asymmetry in the compound nuclear state produced by a polarized target and a polarized neutron. Hence, a polarized ^{139}La target is an attractive device to search for the enhancement-assisted T-violation effect.

A single crystal of lanthanum aluminate ($LaAlO_3$) doped with neodymium ions (Nd^{3+}) allows us to polarize the nuclear spin of ^{139}La by a standard dynamic nuclear polarization (DNP) technique [5]. A few past DNP experiments have shown that a crystal of approximately 0.03 mol% Nd^{3+} concentration is the best and achieved a saturated polarization of about 20% at 1.5 K and 2.3 T [6] and about 50% at a temperature less than 0.3 K and 2.3 T [7]. Using the $LaAlO_3$ crystal as practical target material, one of the biggest issues is to maintain the polarization at about 0.1 T. This comes from the reason that a highly polarized nuclear spin induces a neutron spin rotation due to the spin-dependent nuclear potential, that is called pseudomagnetic effect[8]. Applying an external field of 0.1 T is likely to cancel or reduce systematic effects caused by the systematic neutron spin rotation, a theoretical study has shown that the pseudomagnetic effect in the 50% polarized $LaAlO_3$ crystal corresponds to about 0.1 T [9].

In order to study the spin-lattice relaxation time (T_1) of the 0.03 mol% crystals in such a magnetic field, we have measured T_1 under various conditions and estimated that T_1 0.1 K and 0.1 T is longer than 1 hour [10]. This result leads to the necessity of making T_1 longer at about 0.1 T because the length of T_1 the feasibility of the T-violation search with the $LaAlO_3$ crystal. A possible solution to solve this problem is to find an optimal concentration of the Nd^{3+} ions because T_1 is theoretically expected to be inversely proportional to the square of the Nd^{3+} concentration due to the relaxation process through the electronic Spin-Spin Reservoir(SSR) [6, 11]. In this scenario, it becomes necessary to optimize the Nd^{3+} concentration around 0.03 mol% and to additionally study T_1 at about 0.1 T by using highly crystalline samples of the optimal concentration.

Thus, we are proceeding in terms of three specific issues as follows:

- Improvement of crystal growth methods
- Simple DNP tests for optimizing the Nd^{3+} concentration
- Preparation of a new DNP system for long-term measurements

The method for growing small crystals with rough control of the Nd^{3+} amount has been already established in IMR, Tohoku University [12]. However, this is not enough because it is also desired to precisely control the added amount of the Nd^{3+} at 0.001 mol% level to search for the optimal concentration in the region less than 0.03 mol%. Such precise control by ourselves makes it possible to prepare new crystals quickly and flexibly for iteration of simple DNP tests. In the future, a technique for growing 4 cm^3 crystals with the optimal concentration is necessary, which is the most suitable size for T-violation experiments. In the DNP tests only for the Nd^{3+} optimization, direct measurements of T_1 and the maximum polarization are not always necessary. Instead, we are preparing the new DNP system based on a continuously operating refrigerator at the Research Center of Nuclear Physics, Osaka University (RCNP), and planning to perform the DNP experiments using the optimal crystals in long-term measurements only. In this report, we present the current status in terms of the above three issues and the future prospect.

2. Improvement of crystal growth methods

We have adopted the floating-zone (FZ) technique as our fundamental method [12]. Although this technique is relatively suited to the growth of small-sized crystals, the control of unexpected impurities and the Nd^{3+} concentration is relatively easy compared to other methods because a crucible is unnecessary as shown in Figure 1(a), which is typically responsible for the contamination of paramagnetic impurities including Ir, W, and Mo. In the FZ method, a portion of raw material rod hung from the top is melted by exposing to infrared light focused by ellipsoid mirrors. By moving the ellipsoid mirrors upward slowly, the melting zone is also moving upward and the melted material is crystallized after cooling down. We used the FZ furnace with halogen lamps in terms of low cost, so the possible diameter of the rod is limited to about 5 mm.

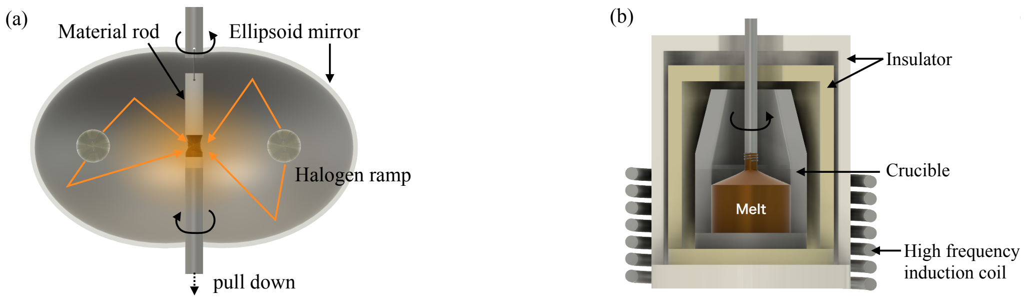

Figure 1: (a) Floating Zone system: In the FZ method, the whole of the material rods are sequentially crystallized from the bottom by cooling the melted part through moving the mirrors upward slowly. (b) Czochralski system: The Cz method grows large bulk crystals by pulling up the melted raw material in the crucible under a temperature gradient.

In the process of preparing the material rod, the Nd^{3+} amount is handled as follows. Raw materials for growing the $LaAlO_3$ crystals are $La(OH)_3$(4N), Al_2O_3(4N) and Nd_2O_3, where the 4N indicates a guaranteed value of the purity of material more than 99.99 %. The powder of $LaAlO_3$ with 0.1 mol% Nd^{3+} concentration as well as pure $LaAlO_3$ powder, which corresponds to the doping amount of 0.1%, can be also easily produced by mixing above raw materials and calcining at 1400 C° for over 8 hours. The mixing two kinds of powders, pure $LaAlO_3$ and $LaAlO_3$ with 0.1 mol% Nd^{3+} concentration, at appropriate mass ratio allows us to prepare the powder of $LaAlO_3$ with a precise concentration in less 0.1 mol%. As the result, this technique eventually allows us to grow the crystal of 0.01 mol% concentration, which has not been tested in the past.

In the future, the 4 cm^3 crystals with the optimal Nd^{3+} concentration are necessary. For this purpose, we are considering the Kyropoulos (Ky) method as a growth method of large-sized crystals. As the first step, we have started fundamental studies on the Czochralski (Cz) method, which is technically similar to the Ky method, as shown in Figure 1(b) due to the easy operation of instruments at IMR.

3. Simple DNP tests for optimizing the Nd concentration

In order to observe the dependence on the Nd^{3+} concentration, simple DNP tests of the grown crystals have been carried out by using the DNP system at Yamagata University, which provides the environment of 1.3 K and 2.3 T. The sample was fixed inside of the sample box mounted in the glass dewar as shown in Figure 2, and cooled down to 1.3 K by pumping liquid ^{4}He with a booster pump. The cooling system is single-shot type and has a capability of continuously running during about 5 hours, which is limited by a storage capacity of liquid ^{4}He. In using the crystals with lower concentration, the direct measurements of the T_1 and the maximum polarization are likely to be more difficult because the time length of the buildup and the relaxation becomes longer, but the comparison on various Nd^{3+} concentrations is possible. The enhanced NMR signals of both, ^{139}La and ^{27}Al nuclei, were obtained via sweeping the NMR frequency in the range of 0.1 times as large as the width of Q value. We have actually tested two kinds of crystals with different concentrations: one is 0.05 mol% and the other is 0.01 mol%. Both crystals have a cylindrical shape with a diameter of 5 mm. The length of 0.05 mol% crystal is about 10 mm and that of 0.01 mol% crystal is about 5 mm. Figure 2 shows the 0.01 mol% crystal used practically.

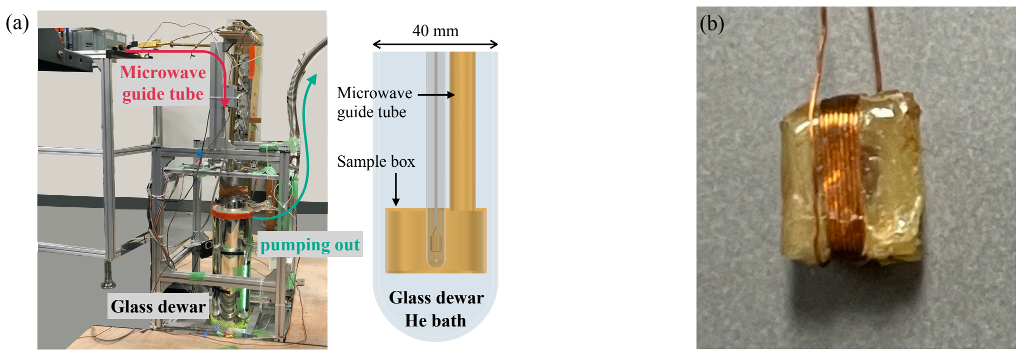

Figure 2: (a) Main DNP system at Yamagata University: It consists of a single-shot cooling system, a microwave system with a frequency of about 70 GHz and an input power of 200 mW, a vacuum system, and a magnet system. (b) Crystal with 0.01 mol% concentration: The crystal has a cylindrical shape with a diameter of 5 mm and a height of 5 mm. The NMR pickup coil was produced by turning wire around the crystal and fixed with epoxy resin (produced by Nichiban Co., Ltd. [13]) without losing shape.

We have successfully observed the enhancement of the NMR peaks for both of the 0.05 mol% and the 0.01 mol% crystals grown by ourselves. The results of the 0.05 mol% crystal have shown the maximum polarization of 0.2 % and a T_1 of 15 min from the measurements of enhanced NMR peaks of the transition between $-1/2$ and $+1/2$ sublevels. In the 0.01 mol% crystal, although the polarization has not been saturated in the limited experiment time, the unsaturated polarization of about 26 % (preliminary) has been obtained at an elapsed time of 150 min after the irradiation of the microwave pulse. Figure 3 shows the results of the NMR peaks at three different elapsed times, 10 min, 60 min, and 120 min. Although T_1 has not been directly measured due to the limitation of experiment time, we have obtained a buildup time of 120 min from the analysis of the increasing amplitude of the NMR peaks in the transition between −1/2 and +1/2 sublevels. Taking into account the relation between T_1 and the buildup time in the DNP process, T_1 is always longer than the buildup time. Therefore, the result leads to the conclusion of $T_1 \geq 120$ min.

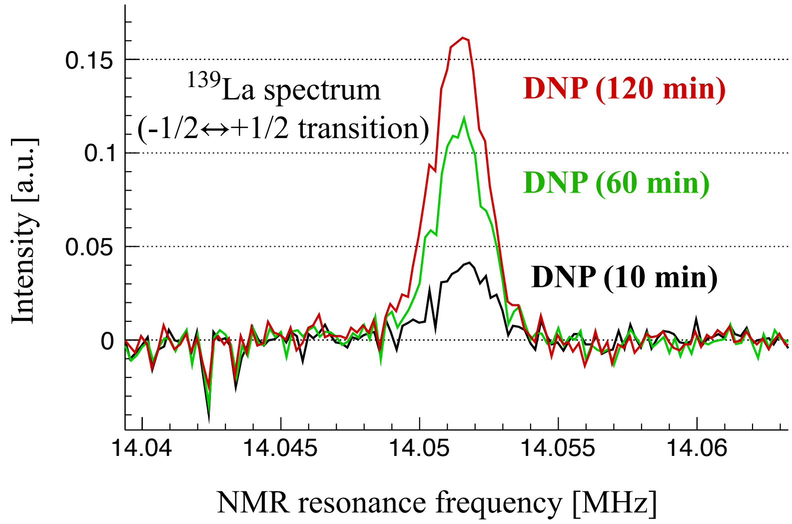

Figure 3: Enhancement of the ^{139}La NMR peak (−1/2 ↔ +1/2 transition) at 1.3 K and 2.3 T using the sample of 0.01 mol% Nd concentration. The NMR peaks are shown at elapsed times of 10 min (black), 60 min (green), and 120 min (red) after the microwave irradiation.

Table 1 shows the summary of the experimental results including the past one on the Nd^{3+} concentration.

Table 1: Summary of DNP experiments

Nd conc. [mol%]	Condition	^{139}La polarization P [%]	^{139}La Relaxation Time T_1 [min]
0.05	1.3 K, 2.3 T	0.2	15
0.03 [6]	1.5 K, 2.3 T	20	82
0.01	1.3 K, 2.3 T	$P > 26$	$T_1 > 120$

According to Table 1, the 0.01 mol% concentration is the best one in the region more than 0.01 mol%. For more clear confirmation, it is necessary to directly measure T_1 and the maximum polarization and to compare these results with the other ones. Additionally, these results in Table 1 also show that we achieve the precise control to bring a distinguishability between 0.03 mol% and 0.01 mol% on the DNP. In the environment of 0.1 K and 0.1 T also, T_1 of the 0.01 mol% crystal is likely to be longer than the one of the 0.03 mol% crystals. Further studies in the region less than 0.01 mol% are quite significant.

4. Preparation of the new DNP system

The DNP system at Yamagata University cannot be operated for longer time than 5 hours. Since there is no helium recovery system in the institute, the budget for the operation becomes expensive. A complete measurement of the long T_1 needs a new DNP system which can be operated continuously for a few days. We decided to produce this new DNP system at RCNP, Osaka University, because a Helium recovery system exists and liquid helium is stably provided by the low temperature center for a reasonable price.

HD targets polarized by the static method have been developed at RCNP [14], and a dilution refrigerator (DRS2500) produced by Leiden Cryogenics and a superconducting magnet with a maximum field of 17 T produced by JASTEC have been operated. The DRS2500 has a hole called clear-shot where samples are exchanged. We installed an insert into the clear-shot and liquid helium was filled in the top part of the insert. When a needle valve was released, a small amount of liquid helium went down to the bottom part where a sample was placed. After pumping the bottom part by a mechanical booster pump (Edwards EH1200) and a dry pump (Kashiyama NeoDry36E), temperatures around 1.5 K were obtained.

A microwave generator (Klystron) provided by KEK generates microwaves with a frequency of 140 GHz and a power of about 10 W. The appropriate magnetic field for the DNP is about 5 T which is easily provided by the superconducting magnet.

We plan to carry out an experiment to cool a $LaAlO_3$ sample at about 1.5 K continuously for at least a few days. If the continuous operation is successful, the DNP experiments will be carried out for a long time.

5. Summary and prospect

According to the past studies on the development of the polarized ^{139}La target for the T-violation search, a single crystal of $LaAlO_3$ doped with a 0.03 mol% Nd^{3+} concentration was thought as the strongest candidate for the practical target. However, our previous studies on the relaxation time have revealed an insufficient T_1 estimated at 0.1 K and 0.1 T and the necessity of determining the optimal Nd^{3+} concentration for making it longer. Thus, we are proceeding with the optimization using crystals grown with various Nd^{3+} concentrations. To perform long-term DNP experiments using the crystals of the optimal concentration, we are also preparing a new DNP system at RCNP.

On the crystal growth, the method for precisely handling the Nd^{3+} amount has been almost established. Differently from the 0.05 mol% crystal, this technique has provided the growth of crystals with 0.01 mol% Nd^{3+} concentration. The DNP tests with the 0.01 mol% crystal have shown that the maximum polarization is higher than 26% and T_1 is longer than 120 min, which are better than those of the 0.03 mol% crystal in the past experiments. As the result, the 0.01 mol% concentration is likely to be the best one. Moreover, we have also confirmed that we can achieve the precise control to bring a distinguishability a Nd^{3+} concentration between 0.03 mol% and 0.01 mol% on the DNP. Since this technique is potentially valid for controlling the concentration at a 0.001 mol% level, a further study of the optimal concentration in the region less than 0.01 mol% is possible.

We are planning long-term DNP experiments with crystals of the optimal Nd^{3+} concentration by using the new DNP system at RCNP. For growing the large-sized crystals as the first step, the fundamental studies on the Czochralski method, technically similar to the Kyropoulos method, are already underway at IMR, Tohoku University.

PoS(PSTP2022)038

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