



# Polarized Lanthanum Target for the T-violation Search in Slow Neutron Transmission

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The Nd<sup>3+</sup> doped LaAlO<sub>3</sub> crystal is a good candidate of a polarized <sup>139</sup>La target for the search of the P-odd and T-odd interaction in the low-energy neutron absorption of <sup>139</sup>La. As the first step for realizing the polarized <sup>139</sup>La target, we grow the undoped LaAlO<sub>3</sub> crystal and the La<sub>1-x</sub>Nd<sub>x</sub>AlO<sub>3</sub> ( $x = 5 \times 10^{-4}$ ) crystal by floating-zone method with four halogen lamps. The ESR measurements of Nd<sup>3+</sup> in the latter crystal indicated that the linewidth was about 6 gauss, whose value is the same as the crystal in which 50 % <sup>139</sup>La polarization was achieved in the previous DNP experiment. As the result, the crystals grown by this method can be applicable to the target study for the T-violation search.

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### 1. Introduction

The ratio of amount of matter to anti-matter in the universe can be predicted from the Cabibbo-Kobayashi-Maskawa(CKM) model, but there is still a large discrepancy from the observational cosmology [1]. One possible explanation is the existence of undiscovered CP violation in the fundamental interactions [2].

In the <sup>139</sup>La, the P-violating asymmetry in the polarized low-energy neutron absorption is about six orders of magnitude larger than that in the nucleon-nucleon scattering [3, 4]. A theory also predicts the large enhancement of the P-odd and T-odd asymmetry in the reaction [5]. According to the CPT theorem, the measurement of T-violating effects is applicable to search the unknown CP violating interaction. In the above system, an effect of the P-odd and T-odd appears as a contribution of  $\vec{\sigma} \cdot (\vec{I} \times \vec{k})$  in the forward scattering amplitude, where  $\vec{\sigma}, \vec{I}$ , and  $\vec{k}$  are a neutron spin, a nuclear spin of the target, and a neutron momentum, respectively. In experimental configuration sensitive to the P-odd and T-odd asymmetry, a spin rotation of the incident neutron is unavoidable because of a pseudomagnetic field induced by the polarized nuclei [6]. The field strength in the LaAlO<sub>3</sub> crystal is estimated to be roughly less than 0.2 T [7, 8]. If we apply the external magnetic field as low as the psudomagnetic field, it is expected to cancel out the effect of the psudomagnetic rotation.

The NOPTREX (Neutron Optics for Parity and Time Reversal EXperiment) collaboration is planning to make the precise measurement of the T-violating effects in the absorption of the low energy neutrons. Our previous investigations with a detection of emitted gamma rays have revealed that the <sup>139</sup>La nucleus is one of candidates for the target nuclei [9]. Moreover, about 50% <sup>139</sup>La polarization in the single crystal of LaAlO<sub>3</sub> doped with Nd ions has been achieved by the conventional DNP in PSI [10]. These are reasons why we have chosen the <sup>139</sup>La for the first experimental exploration.

The LaAlO<sub>3</sub> crystals have preferable characteristics as the target material. First, other kinds of nuclei in the crystal, <sup>16</sup>O and <sup>27</sup>Al have very low absorption compared to that of the <sup>139</sup>La nuclei. Secondary, the symmetrical axises of the electrical field gradient around each nucleus are aligned to the  $C_3$  axis. This feature allows us to diagonalize the quadruple coupling along the  $C_3$  axis. Therefore, if the magnetic field is parallel to the  $C_3$  axis, the reduction of the magnetic field is in principle possible with keeping the high polarization.

In general, the DNP needs paramagnetic centers as a seed of highly electron polarization and a narrow linewidth of ESR spectrum is preferable for an efficient polarization transfer. Since the  $Nd^{3+}$ : LaAlO<sub>3</sub> crystal has one magnetically equivalent site for La nuclei and Nd ions, the ESR linewidth is very narrow. This feature has a great advantage in performing the DNP [11].

Toward the realization of the polarized <sup>139</sup>La target for a practical use, we have some issues on the Nd<sup>3+</sup> : LaAlO<sub>3</sub> crystals. Inhomogeneity of local magnetic field at the La site must be suppressed so that the ESR line width is sufficiently narrow relative to the nuclear Zeeman splitting and well-resolved solid effect can be expected. The twin domain, which easily appears due to the structural phase transition from cubic to trigonal at 813 K for LaAlO<sub>3</sub> [12], should be removed as much as possible. Additionally, the density of the paramagnetic center should be optimized by comparing the efficiency of the polarization buildup in the DNP and the spin-lattice relaxation rate. These requirements should be satisfied in sufficiently large crystal for the T-violation experiment,

Kohei Ishizaki

in which the preferred crystal size is 4 cm x 4 cm x 5 cm.

As the beginning stage on the preparation of the crystals, we have studied the crystal growth for a small size of the  $Nd^{3+}$ : LaAlO<sub>3</sub> crystals and evaluated the grown crystals through the ESR spectrum.

#### 2. Crystal growth of LaAlO<sub>3</sub>



**Figure 1:** (a) : Illustration of floating-zone method. The details are written in the main text. (b) : Floating-zone furnace with four elliptical mirrors, which was used in the crystal growth.

We tried to make the crystal growth of  $LaAlO_3$  by floating zone method with halogen-lamp (Figure 1). In the method, the material of hanged rod is molten by intensively irradiating the infrared light with elliptical mirrors. Molten material is crystallized when the melting material is cooled down by traveling it upward from the zone in which the infrared light is collected with the mirrors. This method has a merit that a dopant concentration is easily controlled. In the Czochralski method, for example, a segregation easily appears, although it is useful for growing large crystals. For the purpose of growing small crystals, the floating zone method is widely used for researchers, especially, in the field of solid state physics.

We used the floating-zone furnace made by Crystal Systems Corporation with four halogen lamps. Since the melting point of LaAlO<sub>3</sub> is about 2100 °C, almost all growth of LaAlO<sub>3</sub> by floating-zone method was conducted with high-power xenon lamps. Therefore, the furnace was used with the maximum output power (4 kW) to melt the LaAlO<sub>3</sub> rod. First, we synthesized the powder of LaAlO<sub>3</sub> by calcining raw materials, 4N La(OH)<sub>3</sub> and Al<sub>2</sub>O<sub>3</sub>, at 1400 °C for 8 hours. After that, the material rod was prepared by the hydrostatic molding of the calcined LaAlO<sub>3</sub> powder and the sintering at 1400 °C for 8 hours more. Additionally, the rod was sintered using the floatingzone furnace with a traveling speed of 5 mm/h before the growing process.

We grow the crystals with the speed of 10 mm/h in the atmosphere. As the results, we obtained the following LaAlO<sub>3</sub> crystals, undoped(#1), 0.05% Nd doped (#2), and undoped and grown along the  $C_3$  axis (#3) (Figure 2, Table 1). The laue diffraction confirmed that these were single crystals.

The undoped crystal (#1) was cut into two pieces, and one of them was annealed at 1200  $^{\circ}$ C for 60 hours in the atmosphere. This process changed the color of the crystal from dark brown to

Kohei Ishizaki

transparent brown (Figure 3). It may be caused by changing the valence of impurity ions in the crystal.



**Figure 2:** Undoped LaAlO<sub>3</sub> crystal (5 mm in diameter and 20 mm in length). The growth was implemented under the condition : the growth direction was  $<111>_{cubic}$  and the growth speed was 10 mm/h.

sample number	diameter	dopant	remarks
#1 a	5 mm	none	No annealing
#1 b	5 mm	none	annealing for 60 hours at 1200 °C in the atmosphere
#2	5 mm	Nd 0.05%	No annealing
#3	5 mm	none	growing along the $C_3$ axis.

Table 1: Summary of the crystals grown by the floating zone method

## 3. ESR measurement of Nd doped LaAlO<sub>3</sub>

The ESR spectra of the #2 crystal,  $La_{1-x}Nd_xAlO_3$  ( $x = 5 \times 10^{-4}$ ), were measured by the CW-ESR apparatus to check the possibility of the DNP. The temperature of the crystal was kept to 15 K and the microwave frequency was 9.493 GHz. We also measured the angular dependence of the ESR spectrum.

Figure 4 shows a typical ESR spectrum in the set of measurements. There is a large peak of Nd<sup>3+</sup> around 0.3165 T. The angular dependence of its resonance frequency leads to the g-factors of  $g_{\parallel} = 2.14$  and  $g_{\perp} = 2.66$  and its linewidth is about 6 gauss. These values are consistent with the ones in the reference [11]. Particularly, the ESR width and the Nd concentration are almost the same as that of the crystal used in the past DNP, which shows the achievement of about 50% <sup>139</sup>La polarization [10]. Therefore, the grown crystal (#2) is expected to indicate the large enhancement by the DNP. Incidentally, the linewidth is explained by the superhyperfine between the Nd<sup>3+</sup> ion and the eight neighboring <sup>27</sup>Al nuclei [11].

In contrast, some small peaks can be seen in the low field region. It can be explained that these peaks are originated from the alternative domains, because the resonance frequency is consistent with the estimation from the obtained g-factors and the orientation of  $C_3$  axis of the other domains.



**Figure 3:** Annealing of the undoped LaAlO<sub>3</sub> crystal. (1) : Before annealing, (2) : The crystal #1 b had been annealed for 60 hours at 1200  $^{\circ}$ C in the atmosphere.



Figure 4: One of the ESR spectrums of the sample # 2 in the Table 1. In this measurement, the  $C_3$  axis was aligned to the magnetic field.

#### 4. Summary and prospects

The NOPTREX collaboration is planning to make the precise measurement of the T-violation in the polarized neutron absorption of the polarized <sup>139</sup>La nucleus. We are proceeding to the development of the <sup>139</sup>La polarized target. One of significant issues is to prepare the target material for the DNP test, which is the single crystal of LaAlO<sub>3</sub> doped with Nd<sup>3+</sup> ions. The small crystals were grown by the floating zone method under the various conditions and the ESR measurements were performed with one of the grown crystals, La<sub>1-x</sub>Nd<sub>x</sub>AlO<sub>3</sub> ( $x = 5 \times 10^{-4}$ ). The results have shown that the ESR linewidth as well as the Nd concentration is almost same as that of the crystal examined in the past DNP. Therefore, the crystal grown by the floating zone method is promising for obtaining the large enhancement by the DNP, although it has twin domains. The study for reducing the twinning is progressing with the grown crystals.

The preparation for the DNP tests is in progress at Research Center for Nuclear Physics, Osaka University. Achieving the sufficiently high polarization to satisfy the practical use, we are going to study the DNP and investigate the dependence of the spin-lattice relaxation on the concentration of  $Nd^{3+}$  with the grown crystals. The T-violation experiment also requires the large crystals, whose growth may be feasible by using the other growth methods with the crucible [13, 14]. We will grow the large crystal in the Institute for Materials Research, Tohoku University.

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