

# Crystal orientation behavior of rare earth substituted $\text{Sr}_2\text{NaNb}_5\text{O}_{15}$ lead-free piezoelectric ceramics under high magnetic field

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## 1. Introduction

Piezoelectric components commonly use PZT ( $\text{Pb}(\text{Zr,Ti})\text{O}_3$ ). However, the use of lead-based piezoelectric materials poses serious health and environmental risks. It can cause pollution and threaten ecosystems. Therefore, searching lead-free alternatives for SDGs is a must.

It was clarified that *c*-axis crystal-oriented  $(\text{Sr,Ca})_2\text{NaNb}_5\text{O}_{15}$  (SCNN) ceramics have good high-power properties such as hard-PZT and outstanding high-power properties than hard-PZT, also was shown the superior performance as ultrasonic motor using the ceramics for the high-power ultrasonic devices in our previous studies<sup>1,2</sup>. However, the *c*-axis crystal-oriented SCNN ceramics can be fabricated by applying high magnetic field that desires to decrease the magnetic field under 1T of permanent magnet at least for practical use.

SCNN is based on  $\text{Sr}_2\text{NaNb}_5\text{O}_{15}$  (SNN) which is a tungsten-bronze-structured ferroelectrics (TBSFs) of  $(\text{A}1)_4(\text{A}2)_2\text{C}_4(\text{B}1)_2(\text{B}2)_8\text{O}_{30}$  with A1, A2, C, and B are the 15-, 12-, 9-, and 6-fold coordinated oxygen octahedra sites in the crystal structure as shown in Fig. 1<sup>3,4</sup>. The A1, A2 sites are occupied by Sr, Na, respectively. C sites are not occupied, instead B1, B2 sites are occupied by Nb<sup>5+</sup>. Therefore, it is considered that the magnetic anisotropy increases substituting the A2 sites to the rare earth elements which have the magnetic susceptibilities are about three orders of magnitude higher than those of  $\text{Sr}^{2+}$ ,  $\text{Na}^{1+}$ ,  $\text{Nb}^{5+}$ ,  $\text{O}^{2-}$  ions<sup>6</sup>. In practice, we experimentally observed a change of crystal orientation under the magnetic field for the rare earth substituted SCNN<sup>7</sup>.

In this study, we focused SNN which is base of SCNN and investigated the crystal orientation behavior of rare earth substituted SNN ceramics under high magnetic field.

## 2. Experimental Procedure

The powders of SNN and Ln substituted SNN (Ln = Nd, Eu, Ho, Yb:  $x = 0.1$  mol) were synthesized by a conventional solid-phase reaction.  $\text{Ln}^{3+}$  ions which is smaller than  $\text{Na}^{+}$  ion occupied in the A2 sites. The SNN and Ln substituted SNN powders

were mixed to prepare a slurry with deionized water and dispersant, respectively.

The crystal orientation of the powders was evaluated under a rotating magnetic field of 10T and a standing magnetic field of 5T. The slurry was dropped on a XRD sample holder (glass). The sample holder was placed in the rotating magnetic field of 10T or the standing magnetic field of 5T. The rotating magnetic field was applied to rotate the sample on a vertical axis under the magnetic field in the horizontal direction. The standing magnetic field was applied a sample in the vertical direction. Those magnetic fields were produced superconducting magnets (Toshiba TM-10VH10, JASTEC JM TD-5T52M). After drying the sample under the magnetic field, its behavior of crystal orientation of the sample was evaluated by XRD. For the comparison, the sample was fabricated without the magnetic field.

## 3. Results and Discussion

SNN and Ln substituted SNN powders were confirmed a single phase belonged to TBSFs. The XRD patterns with the rotating magnetic field were represented that SNN and Ln substituted SNN without Eu were orientated *c*-axis parallel to the rotation axis in the Fig. 2. Eu substituted SNN was orientated *a-b*-axis parallel to the rotation axis while SNN and Ln substituted SNN without Eu were orientated *a-b*-axis parallel to the standing magnetic field and Eu substituted SNN was orientated *c*-axis parallel to the standing magnetic field in the Fig. 3.

As the results, it was clarified that Eu acted to change the magnetic anisotropy of SNN from *a-b*-axis to *c*-axis in the oriented direction parallel to the magnetic field.

## References

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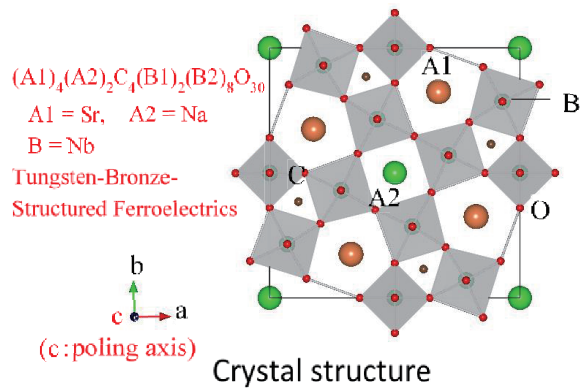


Fig. 1 Schematic of SNN crystal structure.

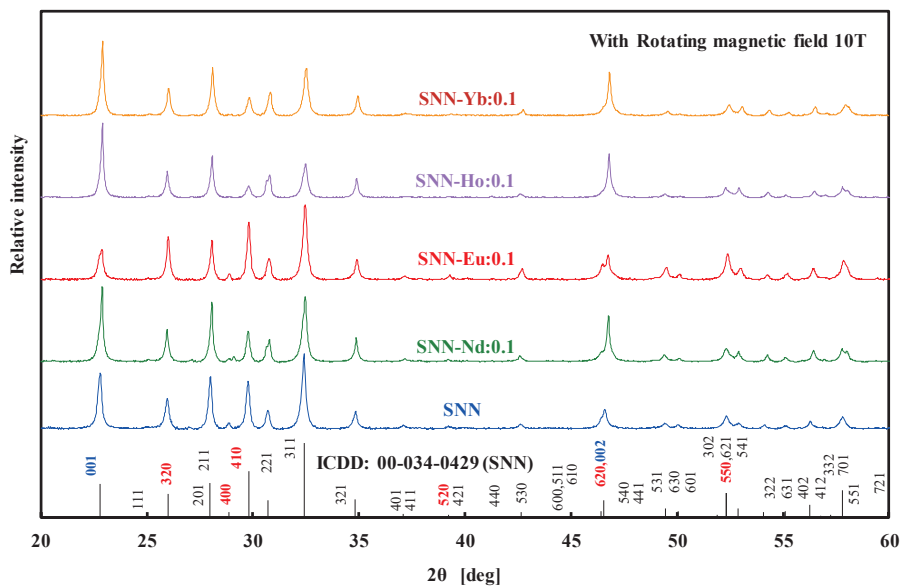


Fig. 2 XRD profiles with rotating magnetic field.

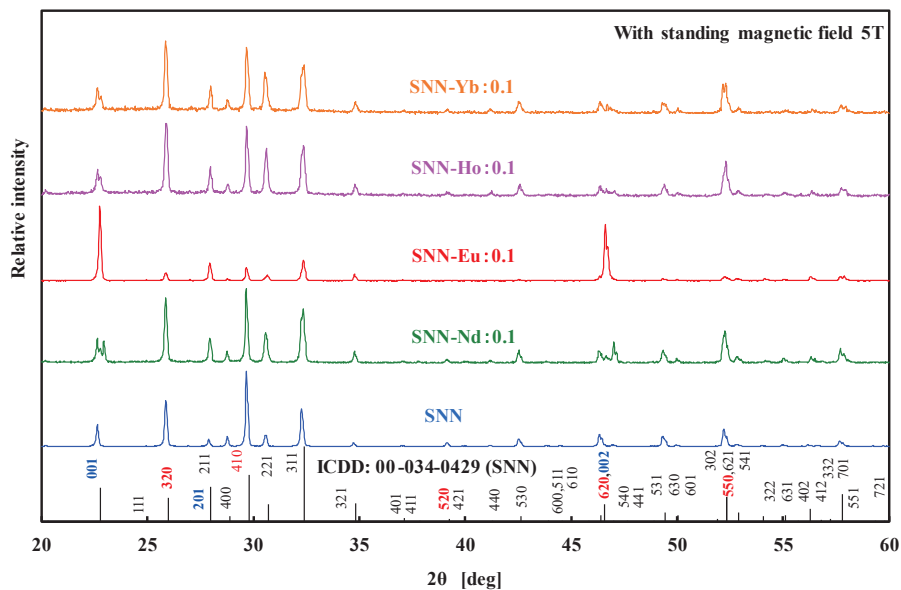


Fig. 3 XRD profiles with standing magnetic field