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# Direct Observation of Spontaneous Polarization in Freestanding GaN Substrate Through Nuclear Magnetic Resonance

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**Abstract**

**We have directly observed spontaneous polarization in freestanding *c*- and *m*-GaN crystal through Ga nuclear magnetic resonance (NMR). We also clarified that the polarization lies in the *c*-direction (in the in-plane direction) for the *c*-GaN (*m*-GaN).**

## 1. Introduction

In GaN, it has been widely believed that, due to strong ionic bonding, there should exist spontaneous polarization. To the best of our knowledge, however, no direct observation has been reported.

Nuclear magnetic resonance (NMR) is a powerful method to observe internal electronic states of various materials, since the nuclei that constitute the material are used to probe the surrounding electronic states in atomic-scale. Indeed, NMR has been contributing a great deal to clarify the microscopic electronic states in various materials such as superconductors [1] and magnetic materials. Mainly due to technological difficulties, however, NMR has not been fully employed to the study of semiconductors [2].

Here, using home-built and customized NMR spectrometer, we have succeeded in direct observation of spontaneous polarization in *c*-GaN and *m*-GaN through Ga-NMR. We also found that the polarization lies in the c-direction (in-plane direction) for the *c*-GaN (*m*-GaN). The present result is well reproduced by the first-principle calculations.

## 2. Experiments, Results and Discussions

For this study, we used two types of freestanding GaN substrate. One is HPVE *c*-GaN and the other is *m*-GaN.

If spontaneous polarization exists, it should be observed as splitting of NMR frequency spectra through the interaction between Ga nuclei and the electric field gradient (EFG) at the Ga-sites.

**(a) *c*-GaN** : This is exactly observed in Figure 1 where $\theta$ (the angle between the c-axis and the magnetic field) is zero. As is seen in Fig. 1, the splitting between the right-hand satellite and the left one is equally divided by the center peak with the interval of 1.36 MHz. From other experiments, we have confirmed that the interval is completely attributed to the EFG at the Ga-sites.

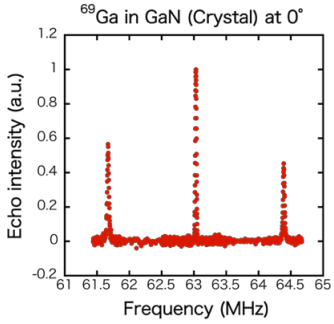


Figure 1. Ga-NMR spectrum for *c*-GaN with $\theta$ =0 under the magnetic field of 6.16 tesla.

Moreover, we found that the interval depends on $\theta$ as $(3\cos^2\theta - 1)$ [3], which is clearly seen in Fig. 2.

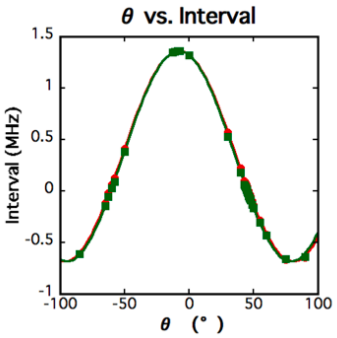


Figure 2. $\theta$ -dependence of the interval of Ga-NMR spectra for *c*-GaN. The shift of the peak angle from 0° is confirmed to come from canted setting of the *c*-GaN sample.

These results indicate that there exists surely spontaneous polarization in *c*-GaN and the direction is parallel to *c*-axis.

**(b) *m*-GaN**

For *m*-GaN, we also observed splitting of the Ga-NMR spectra which are confirmed to come from the EFG at the Ga-sites. Compared to the results of *c*-GaN, however, the angular dependence is totally different. The upper panel of Fig. 3 shows the Ga-NMR spectrum in the case that the m-surface is parallel to the magnetic field. In contrast, the lower panel is the spectrum when the m-surface is placed perpendicular to the magnetic field. These results can be understood only when the polarization lies in the in-plane direction for *m*-GaN, as expected.

## 3. Conclusions

Through Ga-NMR spectra, we have directly observed the spontaneous polarization in both *c*-GaN and *m*-GaN. From the angular dependence, the polarization in *c*-GaN is perpendicular to the *c*-surface, whereas the polarization in *m*-GaN lies in the *m*-surface.

## References


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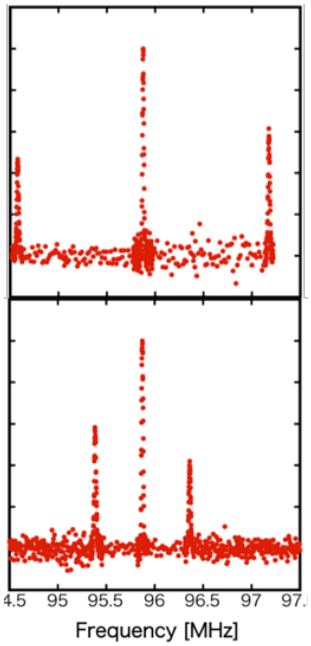


Figure 3. Ga-NMR spectra for *m*-GaN under the magnetic field of 9.4 tesla. Upper: The *m*-surface is placed parallel to the magnetic field. Lower: The *m*-surface is placed perpendicular to the magnetic field. These results indicate that, for *m*-GaN, the spontaneous polarization lies in the *m*-surface.