Application of ²¹O β -NMR spectroscopy to study the microscopic properties of CuO single crystal

A. Gladkov,^{*1} H. Yamazaki,^{*1} Y. Ichikawa,^{*1} A. Takamine,^{*1} H. Nishibata,^{*2} K. Asahi,^{*1} K. Kawata,^{*1,*3} K. Imamura,^{*1} M. Tajima,^{*1} K. Tsubura,^{*1} S. Go,^{*1} H. Endo,^{*1} R. Mitsuyasu,^{*1} K. Kikuchi,^{*1} S. Takahashi,^{*1} S. Akimoto,^{*1} Y. Fukuzawa,^{*1} M. Ito,^{*1} M. Mukai,^{*1} and H. Ueno^{*1}

Since the discovery of high- T_c superconductivity in cuprates, copper ions coordinated by oxygen have been extensively studied. The multiferroic phase of CuO exists between 213 K and 230 K, a much higher temperature range than the multiferroic phase of other known compounds. When the temperature of the CuO sample is decreased from T > 230 K (the paramagnetic phase) to 230 K > T > 213 K (the multiferroic phase), it is expected that the electric field gradient (EFG) at oxygen sites will be changed due to the multiferroic transition. By substituting the O sites in CuO with polarized ²¹O ions, this hypothesized change in EFG can be detected by the β -detected nuclear quadrupole resonance $(\beta$ -NQR) method, providing the microscopic information on the "improper" ferroelectric phase. The advantages of using ²¹O relative to other oxygen isotopes have been discussed in a previous report.¹⁾

Previously, we have successfully measured the ground-state magnetic dipole and electric quadrupole moments of $^{21}O^{2,3)}$ by means of β -detected nuclear magnetic resonance (β -NMR) spectroscopy. Thus, in the present study, the microscopic properties of CuO are investigated by using the ^{21}O isotope as a spectroscopic probe.

The experiment was carried out using the RIPS separator at the RIBF facility. The experimental settings for the production of spin-polarized ²¹O secondary fragments were the same as the optimized settings for the previously conducted measurement of the quadrupole moment in TiO₂ single crystal.²⁾ A ²¹O beam was produced in the projectile fragmentation reaction of a ²²Ne beam at 70 MeV/nucleon on a 185 mg/cm² Be target. The momentum window and emission angle of the secondary fragments were selected to be $p_{\rm F} = p_0 \times (0.97 \pm 0.03)$ and $\theta_{\rm F} > 1.5^{\circ}$, respectively.

The secondary beam of ²¹O was then purified by the RIPS separator and implanted into the single-crystal CuO sample located in the cryostat in the center of the β -NMR apparatus. The well-established method of β -NMR/NQR spectroscopy⁴⁾ in combination with the adiabatic fast passage technique⁵⁾ was used to measure the anticipated change in the EFG around the 210–230 K temperature region.

In order to record a successful β -NQR measurement, it is important to know the EFG orientation through-



Fig. 1. An image of the sample of single-crystal CuO used in the present experiment.

out the sample. For this purpose, the present sample of the single-crystal CuO has been studied using scanning electron microscope imaging, before being placed in the experimental setup so that the main component of the EFG is parallel to the static magnetic field. The β -NQR measurements of the EFG have been obtained at different temperatures in the multiferroic phase region, as well as at 100 K and at room temperature.

Unlike the ²¹O β -NMR measurements for the TiO₂ crystal, where a prominent resonant spectrum was directly observed, no resonance could be distinguished from the measurements of the single-crystal CuO. The obtained β -NMR spectra require further comprehensive analysis to be able to determine the microscopic properties of CuO. This work is in progress and the results will be reported in due course.

References

- 1) T. Kimura et al., Nat. Mater. 7, 291 (2008).
- A. Gladkov *et al.*, RIKEN Accel. Prog. Rep. **54**, 28 (2021).
- 3) Y. Ishibashi et al., Phys. Rev. C 107, 024306 (2023).
- D. Nagae *et al.*, Nucl. Instrum. Methods Phys. Res. B 266, 4612 (2008).
- 5) A. Abragam, *The Principles of Nuclear Magnetism* (Clarendon, Oxford, 1961).

^{*&}lt;sup>1</sup> RIKEN Nishina Center

 $^{^{\}ast 2}$ $\,$ Department of Physics, Kyushu University

^{*&}lt;sup>3</sup> Center for Nuclear Study, University of Tokyo