

# $^{63}\text{Cu}$ NMR Study on $\text{Nd}_{1.85}\text{Ce}_{0.15}\text{CuO}_{4+\delta}$

K. Miyagawa<sup>a,1</sup>, H. Kawamura<sup>a</sup>, K. Kanoda<sup>a</sup>, Y. Onose<sup>a</sup>, Y. Taguchi<sup>a</sup>, Y. Tokura<sup>a</sup>,

<sup>a</sup>*Department of Applied Physics, University of Tokyo, 7-3-1 Bunkyo-ku, Tokyo 113-8656, Japan*

---

## Abstract

The title material is known as an electron-doped high- $T_C$  superconductor, while the as-grown sample is a semiconductor. The  $^{63}\text{Cu}$  NMR studies for the as-grown and reduced  $\text{Nd}_{1.85}\text{Ce}_{0.15}\text{CuO}_{4+\delta}$  single crystals were performed. The NMR spectra of both the samples show small and continuous distribution of  $\nu_Q$ , which has nothing to do with the appearance/disappearance of superconductivity but may be structural in origin. Both crystals exhibit single-peaked central lines with different line width showing the single phase nature and no contamination of the other phase. The NMR signal of the as-grown sample disappears around 150 K due to the magnetic ordering. On the other hand, the line width of the reduced sample is narrower than that of the as-grown sample and shows weak temperature dependence. These behaviors indicate that the antiferromagnetic short range order is depressed when the sample is reduced.

*Key words:* High- $T_C$  superconductor ; electron dope ; NMR ;

---

## 1. Introduction

The relationship between magnetism and superconductivity is one of the main topics in the high- $T_C$  superconductors. The Nuclear Magnetic Resonance (NMR) is a useful probe to provide insight into this problem. Indeed, the pseudo-gapped behaviors in the relaxation rate and the Knight shift have been reported in the under-dope region of hole-doped superconductors. The  $\text{Nd}_{1.85}\text{Ce}_{0.15}\text{CuO}_{4+\delta}$  is known as an electron-doped superconductor, while the as-grown sample is a magnetic insulator with  $T_N$  of around 150 K. There is a small amount of apical oxygen in the as-grown sample. It is believed that by reducing the oxygen the superconductivity appears. The pseudo-gap state was observed for the as-grown  $\text{Nd}_{1.85}\text{Ce}_{0.15}\text{CuO}_{4+\delta}$  in the optical measurements, while it disappeared after reduced [1],[2]. Compared with the hole-doped system, there are small numbers of reports of NMR measurements for the electron-doped system.

In this paper, we report the  $^{63}\text{Cu}$  NMR study for single crystals of as-grown and reduced  $\text{Nd}_{1.85}\text{Ce}_{0.15}\text{CuO}_{4+\delta}$  to get informations about the electronic states before and after the oxygen reduction.

## 2. Experimental

The single crystal was prepared by TSFZ method. The NMR spectra were obtained by integrating the spin-echo signal while sweeping the external field applied parallel to the conducting *ab* plane. The NMR observation frequency was 90.515 MHz.

## 3. Results and Discussion

Figure 1 (a) shows the temperature dependence of the  $^{63}\text{Cu}$  NMR line shape for the as-grown sample. The spectra consist of sharp and rather broad components. The Cu nuclear spin is 3/2 and has a quadrupole moment of  $Q$ . In presence of the electric field gradi-

---

<sup>1</sup> E-mail:kazuya@ap.t.u-tokyo.ac.jp

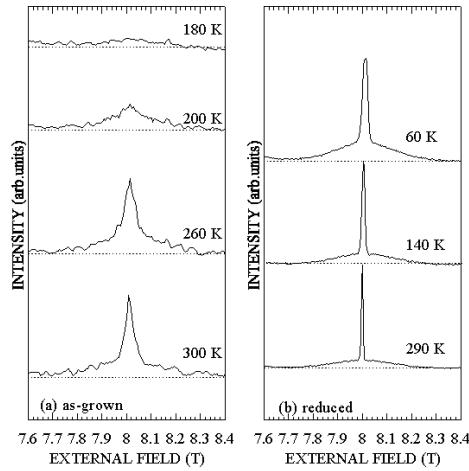


Fig. 1. Temperature dependence of  $^{63}\text{Cu}$  NMR spectra of (a) as-grown and (b) reduced  $\text{Nd}_{1.85}\text{Ce}_{0.15}\text{CuO}_{4+\delta}$

ent (EFG), the NMR spectrum should have symmetric satellite lines at both sides of a center line, of which the position is determined mainly by the magnetic interaction within the 1st-order EFG approximation. The separation,  $\nu_Q$ , of the satellites, which is determined by interaction of  $Q$  with EFG, should be insensitive to magnetic field. The sharp peak is reasonably assigned as the center line. The broad component is regarded as continuous distribution of the  $\nu_Q$  satellites. This assignment is supported by the field-dependence of the spectra; the distribution width of the broad component did not vary with magnetic field while the width of the center-line is roughly proportional to magnetic field. The former is a characteristic of the quadrupole interaction and the latter is a feature of the magnetic interaction with the paramagnetic electron spin.

With decreasing temperature, the center line is gradually broadened. The signals become weak and disappear around 150 K. These behaviors indicate that the short range magnetic order evolves into a three-dimensional ordering around 150 K as reported in the neutron study [3].

The line shape of the reduced sample is similar to that of the as-grown sample except for the line width of the center line (Fig. 1 (b)). The width of the continuously broadened  $\nu_Q$  satellites does not change even after the reduction. So, the apical oxygen has no involvement in the  $\nu_Q$  distribution.

Figure 2 shows the temperature dependence of the width of the center line. It is seen that the magnitude of the line width contrasts between the as-grown and reduced samples. This fact provides evidences that the reduced sample is a single phase and do not contain appreciable remain of the as-grown phase due to the imperfection of the reduction process. The line width of the reduced sample becomes still broader at lower

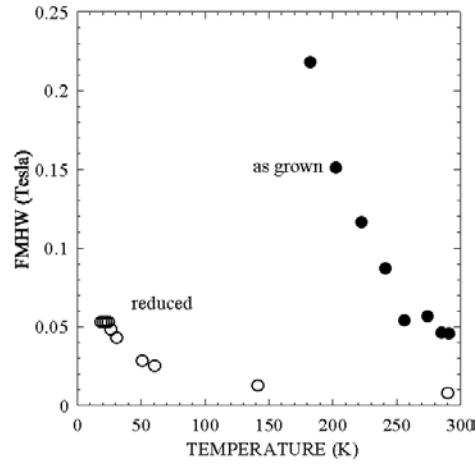


Fig. 2. Temperature dependence of  $^{63}\text{Cu}$  NMR center-line width of as-grown and reduced samples.

temperatures. However, its temperature-dependence is weak compared with the as-grown case. It is likely that the appreciable broadening in the reduced sample comes from the Curie-Weiss susceptibility of  $\text{Nd}^{3+}$  localized spins. These behaviors of the center line show that the enhanced antiferromagnetic short-range order in the as-grown sample is depressed in the reduced sample where superconductivity appears.

Finally, it is mentioned that because of the large spin fluctuations of  $\text{Nd}^{3+}$  in the so-called block layer, we could not get the informations from  $T_1^{-1}$  about the pseudo-gapped state as observed in the optical measurements.

## Acknowledgements

The authors would like to thank staff of Low Temperature Center in University of Tokyo for the technical support. This work is supported by the PRESTO program from JST Corporation and by Grant-in-Aid for Scientific Research from the Ministry of Education, Science and Sports, Japan.

## References

- [1] Y. Onose *et al.*, Phys. Rev. Lett. **82** (1999) 5120
- [2] Y. Onose *et al.*, Phys. Rev. Lett. **87** (2001) 217001
- [3] K. Yamada *et al.* J. Phys. Chem. Solid, **60** (1999) 1025