

NMR studies of non-centrosymmetric phases in the spin-orbit coupled metal $\text{Cd}_2\text{Re}_2\text{O}_7$

NMRによるスピン軌道結合金属 $\text{Cd}_2\text{Re}_2\text{O}_7$ の非反転対称相の研究

Department of Advanced Materials Science, 47-186135, Xia Jingding

Supervisors: Professor Takigawa Masashi

Keyword: Superconductivity, Pyrochlore oxide, NMR, Phase transitions.

Introduction

There has been increasing interest in novel phenomena caused by strong spin-orbit coupling in metals which do not have inversion symmetry. In such materials, spin degeneracy of the conduction electrons is lifted even without magnetic order. This results in spin-split energy bands and spin-polarized Fermi surfaces, which may cause unusual magneto-current effects and non-reciprocal transport in magnetic fields.

$\text{Cd}_2\text{Re}_2\text{O}_7$ is a metallic pyrochlore compound, in which both Cd and Re atoms form a network of corner sharing tetrahedra known as the pyrochlore lattice shown in Fig. 1. It exhibits sequential phase transitions as a function of temperature as shown in Fig. 2. At $T_{s1} \sim 200$ K, the space group of the crystal structure changes from the high temperature cubic $Fd\bar{3}m$ to the low temperature tetragonal $I\bar{4}m2$, which breaks inversion symmetry. At $T_{s2} \sim 110$ K, the second transition takes place from $I\bar{4}m2$ to another tetragonal and non-centrosymmetric $I4_122$ structure [1,2]. Although the changes in the lattice parameters are extremely small across these transitions, of the order of 10^{-4} , electronic properties such as resistivity and magnetic susceptibility shows pronounced anomalies at T_{s1} and T_{s2} . This suggests that the phase transitions in $\text{Cd}_2\text{Re}_2\text{O}_7$ are driven by electronic instability not by lattice instability.

Indeed, Lian Fu proposed theoretically that interacting electrons with strong spin-orbit coupling may break inversion symmetry spontaneously, leading to orders of odd-parity multipole moments [3]. Hayami et al. pointed out that the structural changes mentioned above are compatible with order of electric toroidal quadrupoles with E_u symmetry [4].

Previous NMR results

Motivated by the recent development mentioned above, NMR experiments on $\text{Cd}_2\text{Re}_2\text{O}_7$ have been carried out in Takigawa lab. in order to identify the order parameters in the non-centrosymmetric phases based on the analysis of local magnetic response.

In Fig. 3, the NMR spectra at the Cd sites for the magnetic field of 7 T along [001] are

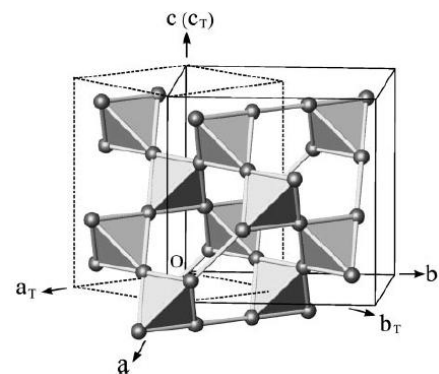


Fig. 1. The pyrochlore lattice.

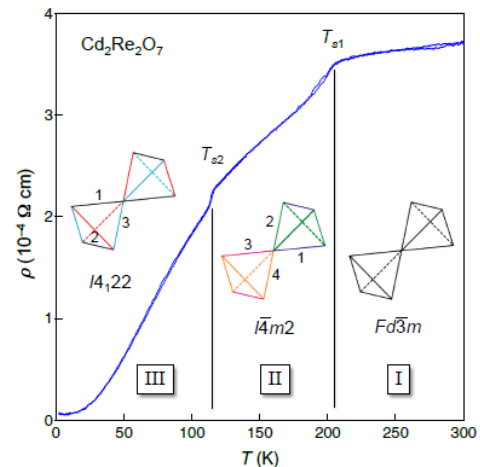


Fig. 2. The sequential transitions [2].

shown at various temperatures in the vicinity of T_{s1} [5]. The single peak observed at high temperatures splits into two peaks at low temperatures. This splitting is due to the cubic-to-tetragonal symmetry change. By fitting the temperature dependence of the splitting to a power law, the cubic-to-tetragonal transition temperature is determined to be 201.5 K (Fig. 4). The average Knight shift determined by the center of gravity of the spectra, on the other hand, shows a kink at a slightly higher temperature of 203.1 K and no anomaly at 201.5 K. These results indicate that the transition at T_{s1} occurs in two steps, i.e. there is an intermediate phase in a very narrow range of temperature between phase I and phase II. This new phase is likely to have non-centrosymmetric cubic symmetry, pointing to the $F\bar{4}3m$ space group.

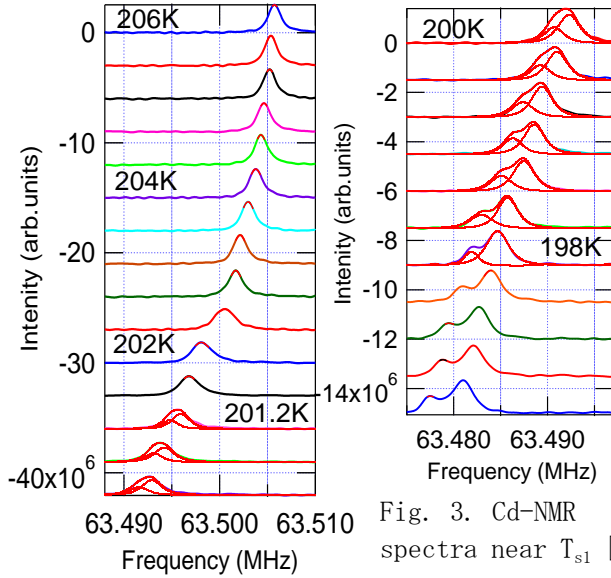


Fig. 3. Cd-NMR spectra near T_{s1} [5].

Experiments

Since there is one Cd site in the $F\bar{4}3m$ structure, Cd-NMR does not allow us to distinguish the $Fd\bar{3}m$ and the $F\bar{4}3m$ structures. However, the oxygen site located near the center of the Re-Re bonds should be split into two sites. Thus, we expect the O-NMR spectra to split into two peaks, should the $Fd\bar{3}m$ to $F\bar{4}3m$ transition occur. Therefore, we decided to perform O-NMR experiments. Single crystals of $Cd_2Re_2O_7$ enriched with ^{17}O isotope with the nuclear spin 5/2 have been successfully synthesized and we are now conducting O-NMR measurements.

For this field orientation, there are two types of oxygen sites marked by the red and blue circles. We have indeed observed two sets of NMR spectra, each of which consists of a quadrupole split five lines. The green line is the NMR spectra of O' site. We have observed ^{17}O NMR signal. Fig.6 is the NMR spectrum at 220K with the magnetic field applied along [111] direction, and Fig.7 is the NMR spectrum with the magnetic field applied along [001] direction. We will make further analysis in the paper.

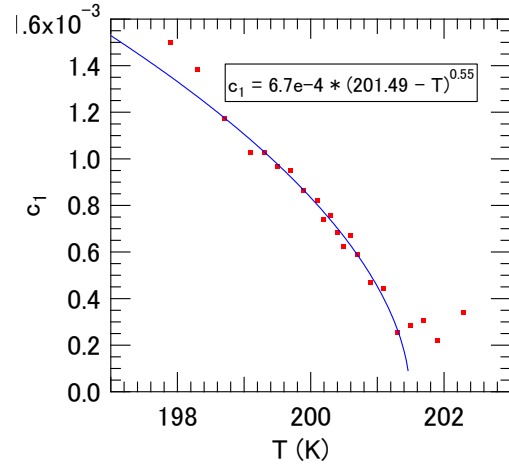


Fig. 4. Splitting of the Cd NMR spectra [5].

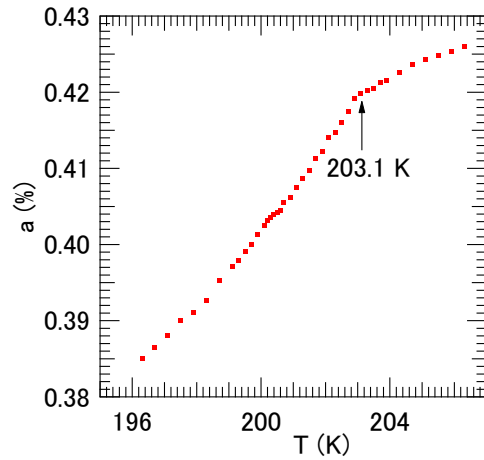


Fig. 4. Average shift of the Cd NMR spectra [5].

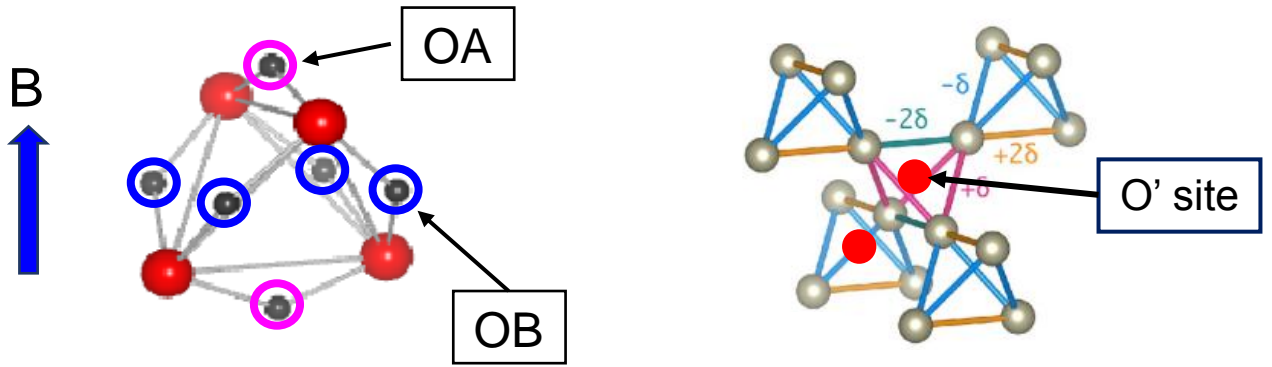


Fig. 5. Crystal structure of $\text{Cd}_2\text{Re}_2\text{O}_7$

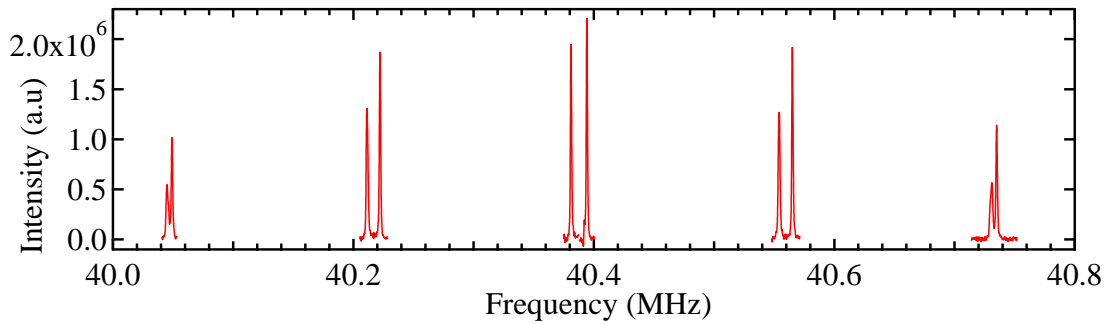


Fig. 6. ^{17}O NMR spectra for $B // [111]$ ($T = 220\text{K}$ $H = 7$)

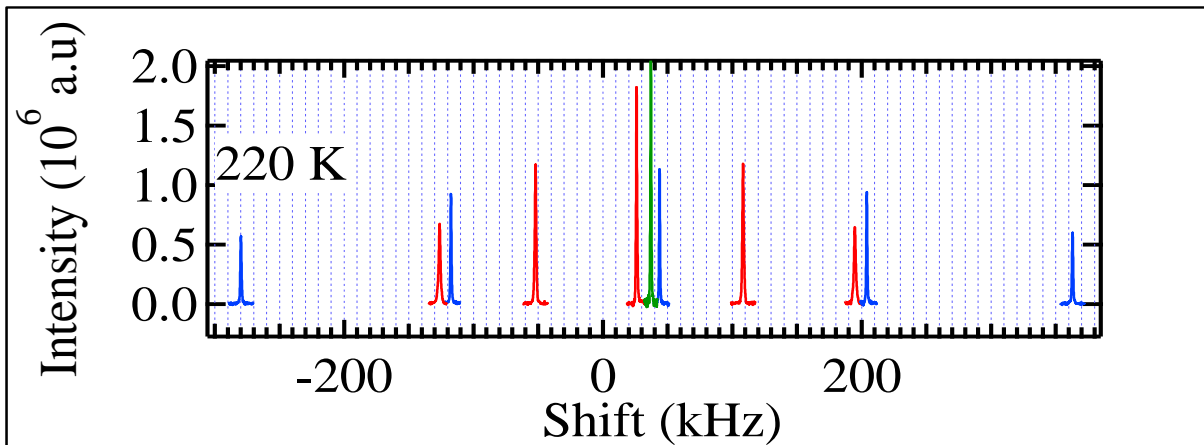


Fig. 7 ^{17}O NMR spectra for $B // [001]$ ($T = 220\text{K}$ $H = 7$)

References

- [1] Z. Hiroi and J. Yamaura, *J. Phys. Soc. Jpn.* **71** (2002) 2598.
- [2] Z. Hiroi, J. Yamaura, T. C. Kobayashi, Y. Matsubayashi, and D. Hirai, *J. Phys. Soc. Jpn.* **87** (2018) 024702.
- [3] Liang Fu, *Phys. Rev. Lett.* **114** (2015) 026401.
- [4] S. Hayami, Y. Yanagi, H. Kusunose, and Y. Motome, *Phys. Rev. Lett.* **122** (2019) 147602.
- [5] N. Mera, M. Takigawa et al., unpublished.