Determination of the magnetic structure of perfect quantum kagome lattice antiferromagnet $CaCu_3(OD)_6Cl_2 \cdot 0.6D_2O$

Hokkaido University Hiroyuki Yoshida

1. Introduction

To investigate the ground state of the perfect kagome lattice with quantum spins in the absence of intersite disorder is very important because all reported perfect kagome lattice quantum antiferromagnets exhibit such intersite disorder. Recently we have succeeded in synthesizing a new kagome material, $CaCu_3(OH)_6Cl_2 \cdot 0.6H_2O$ [1]. By means of single-crystal X-ray diffraction, we have determined a crystal structure. $CaCu_3(OH)_6Cl_2 \cdot 0.6H_2O$ has a *P*-3*m*1 space group (No164, trigonal). Magnetic Cu^{2+} ions locate at the 3*f* site [(0.5, 0.5, 0.5)] and form a "perfect" kagome lattice. Since Cu^{2+} ions are in octahedral environment, the Cu^{2+} ion has S = 1/2. On the other hand, there are non-magnetic H_2O layers between Cu-kagome layers, and kagome layers are separated by 5.8 Å. The kagome planes are thus expected to have good two-dimensional feature. We address that Ca/Cu site disorder in $CaCu_3(OH)_6Cl_2 \cdot 0.6H_2O$ was "not" confirmed by our single crystal X-ray measurement using synchrotron radiation. Therefore, $CaCu_3(OH)_6Cl_2 \cdot 0.6H_2O$ can be a good candidate for perfect kagome lattice quantum antiferromagnets.

Magnetic susceptibility (M/H) and heat capacity measurements in single crystal $CaCu_3(OH)_6Cl_2 \cdot 0.6H_2O$ were reported [1]. A magnetic anomaly observed in the *ab*-plane direction at T_N = 7 K in the susceptibility and a small peak at T_N in the heat capacity indicates the occurrence of a magnetic transition at the temperature. Furthermore, our recent NMR measurements also supported the existence of the magnetic transition at T_N = 7 K; the $1/T_1$ showed the small peak at T_N which exhibited the critical slowing down of the spins on the kagome lattice. As discussed in ref. [1], $CaCu_3(OH)_6Cl_2 \cdot 0.6H_2O$ includes magnetic interactions of a nearest-neighbor J_1, a next-nearest-neighbor J_2, and a third-nearest-neighbor J_d on the kagome lattice which dominate the ground state of the system. Theoretically, the $\sqrt{3} \times \sqrt{3}$ structure, the q = 0 order, and an unusual noncoplanar magnetic ordered state called Cuboc1 state with 12-sublattice compete with each other in the J_1-J_2-J_d magnetic phase diagram at T = 0 [2]. To determine the magnetic ground state of $CaCu_3(OH)_6Cl_2 \cdot 0.6H_2O$ contributes to the general understanding of intrinsic magnetic properties of quantum kagome lattice antiferromagnet. Therefore, we performed single-crystal neutron diffraction experiments to study magnetic structure in $CaCu_3(OH)_6Cl_2 \cdot 0.6H_2O$ below and above T_N using SENJU.

References

[1] H. Yoshida *et al.*, JPSJ **86**, 033704 (2017).
[2] B. Bernu *et al.*, PRB 87, 155107 (2013).

2. Experiment

In the current experiment, we measured a single-crystal $CaCu_3(OD)_6Cl_2 \cdot 0.6D_2O$ with volume of about 1 mm^3. Using the conventional bottom-loading cryostat at SENJU, we performed measurements at 4 K and also investigated the temperature dependence of the magnetic Bragg peak of (1 0 0.5) between 4 and 20 K. The refinement of the magnetic structure is done by JANA.

3. Results

The magnetic Bragg peaks were observed at several positions such as (1 0 0.5) and (1 1 0.5). The corresponding magnetic propagation vector is $\mathbf{k}$ = (0, 0, 0.5). Temperature evolution of the magnetic Bragg peak at (1 0 0.5) is shown in Fig. 1(a). Below T_N = 7 K, the magnetic long-range order develops. As shown in Fig. 1(b), the corresponding magnetic structure of $CaCu_3(OD)_6Cl_2 \cdot 0.6D_2O$ is the $\mathbf{q}$ = 0 structure which is often realized in the kagome lattice antiferromagnet. The $\mathbf{q}$ = 0 magnetic structure is stacked antiferromagnetically along the *c* axis. The estimated ordered moment at 4.2 K is $0.43\mu_B$, and from the temperature dependence shown in Fig. 1(a) we estimated that the ordered moment at 0 K is $0.57\mu_B$, which is strongly reduced compared with the expected value $1\mu_B$. This is probably due to the combination of quantum effects and geometrical frustration. Also, the $\mathbf{q}$ = 0 structure may indicate the Dzyaloshinsky-Moriya interaction is not negligible in $CaCu_3(OD)_6Cl_2 \cdot 0.6D_2O$ because Dzyaloshinsky-Moriya interaction favors the $\mathbf{q}$ = 0 magnetic structure.

4. Conclusion

In summary, we have investigated the magnetic structure of the perfect kagome lattice antiferromagnet $CaCu_3(OD)_6Cl_2 \cdot 0.6D_2O$. Obtained magnetic structure is so-called $\mathbf{q}$ = 0 structure with the reduced magnetic moment of $0.43\mu_B$ at 4.2 K.

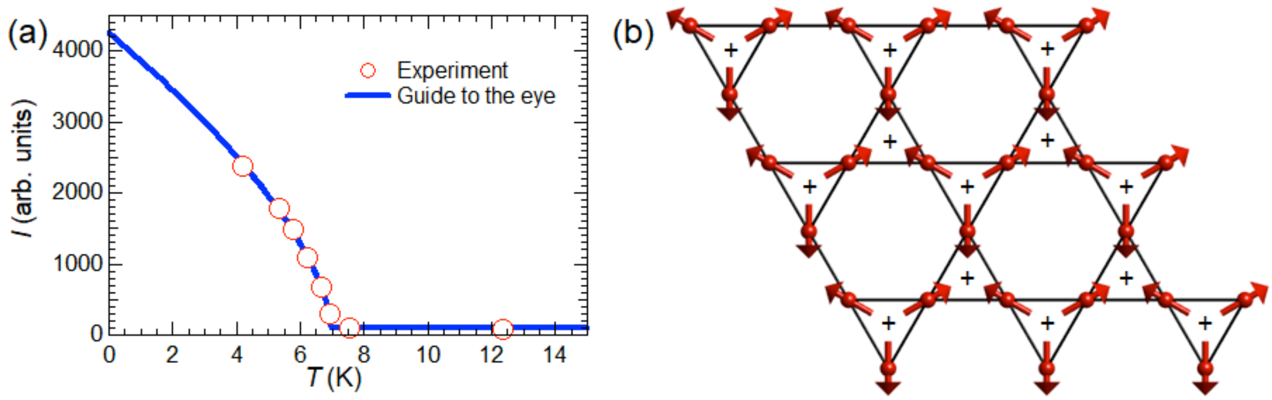

Figure 1. (a) Temperature evolution of the magnetic Bragg peak at (1 0 0.5) in $CaCu_3(OD)_6Cl_2 \cdot 0.6D_2O$. (b) Obtained magnetic structure of $CaCu_3(OD)_6Cl_2 \cdot 0.6D_2O$ with the magnetic propagation vector of $\mathbf{k}$ = (0, 0, 0.5).