Investigation of hydrogen dynamics in hydroxyl salts $Co_2(OD)_3Cl$

X.L. Xu,[*1] X.G. Zheng,[*1] H.J. Guo,[*2] and I. Watanabe[*2]

Hydroxyl salts of the type $M_2(OH)_3X$ (X = Cl, Br, or I) have been known for a long time. These compounds containing magnetic ions are magnetic materials. However, only in recent years their magnetic properties have been clarified as a result of our research [1,2] and they are known as "frustrated magnets". Our latest finding is universal strong magnetic--dielectric--lattice coupling in all such compounds. Measurements of dielectric constants and lattice parameters revealed simultaneous changes at the respective T_N for all hydroxyl salts, which indicate strong magnetic--dielectric--lattice coupling. Moreover, we found that for $Co_2(OH)_3Cl$ and $Co_2(OH)_3Br$, which have the highest crystal symmetry in the hydroxyl salt series shown in Fig. 1, the corresponding deuterated compounds $Co_2(OD)_3Cl$ [Br] clearly exhibited a ferroelectric response at exceptionally high temperatures. Sharp peaks were observed at 229 K in both dielectric constants measured at 100 kHz for $Co_2(OD)_3Cl$.[3] Similar behaviors were observed in $Co_2(OD)_3Br$ (T_E = 224 K at 100 kHz).

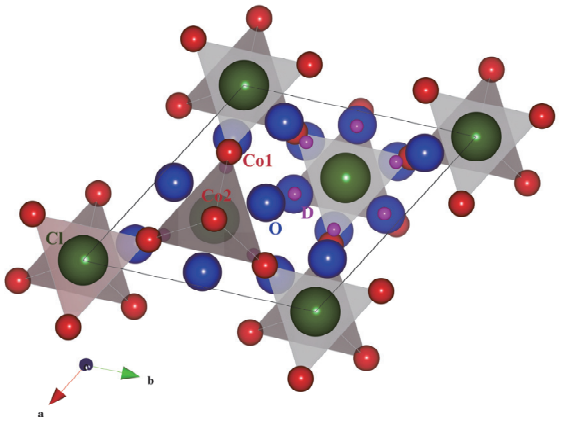

Fig. 1. High crystal symmetry of $Co_2(OD)_3Cl$.

Therefore, we performed μSR experiments on $Co_2(OD)_3Cl$ to reveal the mechanism of this unconventional ferroelectric response, using the muon facilities at RIEKN-RAL. We observed a change in the dynamics of D atoms in $Co_2(OD)_3Cl$ through the nuclear dipolar field of D (Fig. 2).

The asymmetry $a(t)$ of muon-spin-relaxation can be approximately expressed by a combination of the dynamic Kubo-Toyabe function and an exponential function.

The dynamic Kubo-Toyabe function represents the contribution from the nuclear dipolar field of D atoms, and the exponential one accounts for magnetic relaxation.

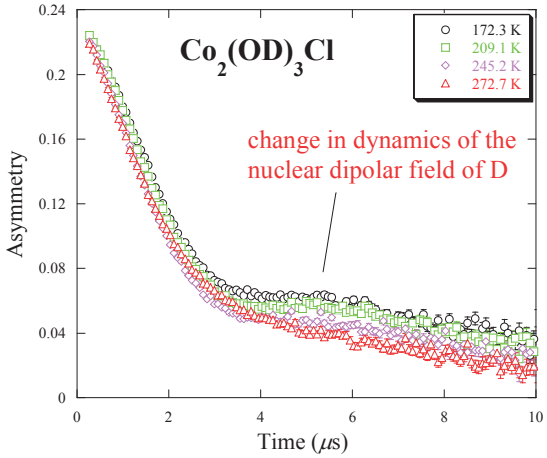

Fig. 2. Muon-spin-relaxation spectra indicating a change in the dynamics of D atoms in $Co_2(OD)_3Cl$.

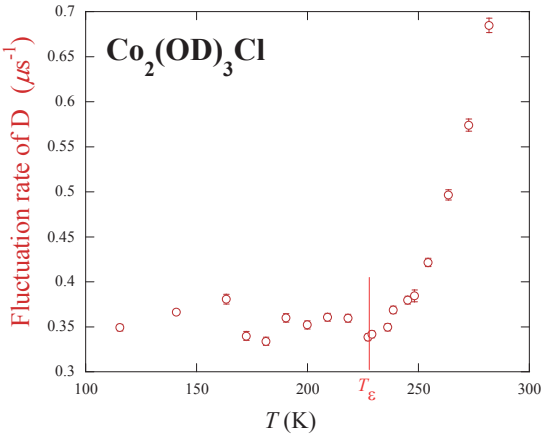

Fig.3. The analyzed fluctuation rate of the nuclear dipolar field of the D atoms in $Co_2(OD)_3Cl$.

The analyzed fluctuation rate of the nuclear dipolar field of the D atoms in $Co_2(OD)_3Cl$ shows an abrupt change around the ferroelectric transition temperature T_E = 230 K (Fig. 3), suggesting that the hydrogen (D) dynamics plays a critical role in the ferroelectric response of $Co_2(OD)_3Cl$.

References

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3) X.G. Zheng et al.: Phys. Rev. B 87, 174102 (2013).

[*1] Department of Physics, Saga University
[*2] RIKEN Nishina Center