μSR study of spin dynamics in Cu-based organic-inorganic hybrid systems

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The muon-site is a very important parameter to study the hyperfine interaction between muon and electrons and to discuss ordered states of electronic spins in magnetic materials. In the previous report,[1)] we have estimated muon-sites in organic-inorganic hybrid materials of $(C_6H_5CH_2CH_2NH_3)_2CuCl_4$ (PEA) and $(C_2H_5NH_3)_2CuCl_4$ (EA) by taking into account the minimum electrostatic potential energy based on the density functional theory (DFT). Accordingly, we estimated six muon-site candidates in EA and eight candidates in PEA; nevertheless, only one site was experimentally achieved on both materials with an internal field of around 200 G at 4 K.[2)] A new achievement for PEA has been obtained at the RIKEN-RAL muon facilities. The internal field of PEA increases by decreasing the temperature with 240 G at 1.5 K. This result can be used to discuss the magnetic interaction mechanism between magnetic layers in PEA system.

In order to understand the magnetic interaction in EA and PEA systems and to explain the experimental results, we performed DFT and dipole field calculations by using the HOKUSAI, RIKEN supercomputer. To begin with, we put a muon at the minimum potential position in a bulk structure of 2 × 2 × 1 supercell to study the localization of the muon. To obtain the final stable muon position, we made a self-consistency test by using the generalized gradient approximation (GGA) and augmented plane wave (APW) pseudopotential methods as implemented in the VASP package.[3)] During the self-consistency, the muon moved to the side of $CuCl_4$ octahedral, pushed the nearest Cu^{2+} ion and deformed the local crystal structure due to change in the local potential around the muon, as shown in Fig. 1 and 2.

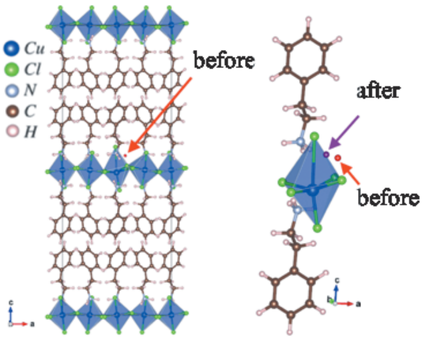

Fig. 1. Local structure deformation of $(C_6H_5CH_2CH_2NH_3)_2$ $CuCl_4$ (PEA) by the injected muon.

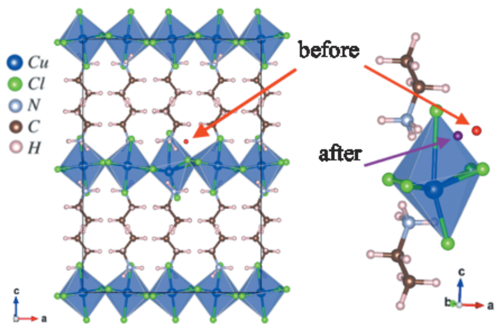

Fig. 2. Local structure deformation of $(C_2H_5NH_3)_2CuCl_4$ (EA) by the injected muon.

In order to confirm our muon-site candidates, we calculated electronic dipole fields at the muon position coming to the magnetic moment of Cu^{2+} spins. We assumed the ferromagnetic spin alignment as previously suggested from the susceptibility measurement[4)] and calculated electronic dipolar fields in three different orientations along the a-, b-, and c- axes to find the ground-state condition. We also considered other quantum effects such as the zero-point vibration motion of the muon, the spatial distribution of the magnetic moments, and the deformation of the surrounding electronic orbital by the injected muon.

Our DFT calculation results on both Cu-hybrid systems show the appearance of magnetic moment distribution of Cl^- as well as that of Cu^{2+} ions. We estimated the size of magnetic moment around 0.5 μ_B and 0.1 μ_B for Cu^{2+} and Cl^- ions, respectively, to calculate dipole fields at the muon site. In order to obtain the accurate muon position, we suggested the supercell calculation to see local deformation of the ionic position due to injected muon. Finally, by including those effects, we can compare the experimental results of internal fields at the muon-site in order to determine the spin structure of Cu-hybrid EA and PEA. Detailed calculations and further discussion of the magnetic interaction mechanism between magnetic layers on those hybrids are under way and we expect to obtain the final results and conclusions very soon.

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