Spin dynamics of quantum spin liquid state in kapelasite

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Ground state of the spin-1/2 perfect kagome lattice is believed to be a quantum spin liquid (QSL) state. It is of great interest whether or not intrinsic spin gap exists in the QSL state [S. Yan *et al.*, Science **332**, 1173 (2011).], which gives us strong constrain to construct the QSL theory. So far, all reported spin-1/2 perfect kagome-lattice compounds have impurity (i.e., atomic intersite randomness between Cu^{2+} and nonmagnetic ions), which prevents us to investigate the existence of spin gap because impurity gives rise to low-energy scattering [T.-H. Han *et al.*, PRB **94**, 201111 (2016).]. To discuss the spin gap problem in the spin-1/2 perfect kagome lattice, we performed single-crystal inelastic neutron scattering measurements on newly discovered spin-1/2 perfect kagome compound $CaCu_3(OH)_6Cl_2{\cdot}0.6H_2O$.

Recently, we have succeeded in growing large single crystals of the new kagome material, $CaCu_3(OH)_6Cl_2{\cdot}0.6H_2O$ [H. Yoshida *et al.*, submitted]. By means of single-crystal X-ray diffraction measurements, we determined its crystal structure. $CaCu_3(OH)_6Cl_2{\cdot}0.6H_2O$ has the $P3m1$ space group (No156, trigonal), and magnetic Cu^{2+} ions with $S = 1/2$ form a perfect kagome lattice. We address that Ca^{2+}/Cu^{2+} intersite disorder was " not " observed in our single crystal X-ray diffraction measurement because of the large difference between their ionic radii. No signature of magnetic long-range order or spin glass was observed in magnetic susceptibility and μSR measurements down to 0.1 K. Currie-Weiss fit gives S = 1/2 and $\Theta_{CW} = -60$ K, indicating antiferromagnetic interactions. In heat capacity measurement, T-linear term ($\gamma = 10.2$ mJ/Cu mol K^2) was observed, suggesting that the ground state is QSL. Therefore, $CaCu_3(OH)_6Cl_2{\cdot}0.6H_2O$ is an ideal compound to investigate the spin gap problem in the spin-1/2 perfect kagome-lattice antiferromagnet.

Very recently, we measured inelastic neutron scattering (INS) on powder $CaCu_3(OD)_6Cl_2{\cdot}0.6D_2O$ at the cold-neutron chopper spectrometer LET in ISIS. Clear spin gap of 0.78(2) meV was observed at 0.15 K. To investigate in detail the momentum vector dependence of the spin dynamics as well as the spin gap in $CaCu_3(OD)_6Cl_2{\cdot}0.6D_2O$, single crystal INS measurements are necessary. We therefore performed single-crystal INS measurements on the QSL state in $CaCu_3(OH)_6Cl_2{\cdot}0.6H_2O$ using the time-of-flight (TOF) chopper spectrometer CNCS in SNS. About four hundreds of single-crystal $CaCu_3(OH)_6Cl_2{\cdot}0.6H_2O$ with total mass of $\sim$ 2 g were co-aligned [Fig. 1(a)]. TOF measurements were performed at 1.5 and 50 K with $E_i = 12$ meV. We rotate crystals by 225° to observe the whole picture of magnetic excitations.

Figure 1(b) shows neutron diffraction pattern at 1.5 K. No magnetic excitation was observed, indicating the absence of symmetry-breaking magnetic long-range order. Figure 1(c)–1(f) show constant-energy maps. Hexagonal-shaped excitation, which is typical magnetic excitation in the kagome QSL state, was observed at 1.5 K above 1 meV. Similar magnetic excitation survives at least 8.5 meV, which can be also seen in the powder results. In the meanwhile, no hexagonal-shaped magnetic excitation was observed below 1 meV. These results indicate that spin gap with about 0.5 meV exists at 1.5 K. Therefore, we have succeeded observing the spin gap in quantum spin liquid kagome $CaCu_3(OH)_6Cl_2{\cdot}0.6H_2O$. The size of the spin gap is consistent with the powder result.

Since $CaCu_3(OH)_6Cl_2 \cdot 0.6H_2O$ is the most clean system to investigate QSL, we believe that our findings on the spin gap in $CaCu_3(OH)_6Cl_2 \cdot 0.6H_2O$ can shed light on QSL and encourage further theoretical works.

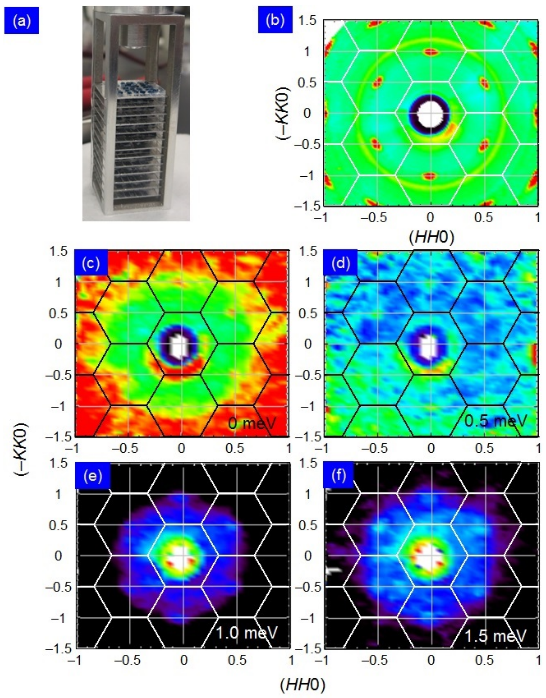

Fig. 1. (a) Picture of co-aligned single crystals. (b) Neutron diffraction pattern at 1.5 K. Constant-energy maps by subtracting the high temperature data (50 K) from the low temperature data (1.5 K) at (c) 0, (d) 0.5, (e) 1, and (f) 1.5 meV.