Magnetic correlations in spin-3/2 perfect kagome Li2Cr3SbO8

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Network of corner sharing triangles forms kagome lattice. When the antiferromagnetic nearest-neighbor (NN) interaction exists the system shows geometrical frustration. Due to the geometrical frustration, classical kagome lattice Heisenberg antiferromagnet (CKLHAF) has infinitely degenerated ground states which are connected by zero-energy modes. Order by disorder theory predicted that quantum fluctuation favors so-called $\sqrt{3} \times \sqrt{3}$ order at $T \rightarrow 0$ among the many possible states. Experimentally, the magnetic long range order with a $\mathbf{q} = 0$ structure which has different spin chirality from that of the $\sqrt{3} \times \sqrt{3}$ structure was reported in several CKLHAF materials such as Cr and Fe jarosites. This is because Dzyloshinsky-Moriya (DM) interaction and/or single-ion anisotropy lead to $\mathbf{q} = 0$ structure. Therefore, experimental realization of $\sqrt{3} \times \sqrt{3}$ short-range order or classical spin-liquid ground state is highly desirable to understand the nature of the CKLHAF model.

Recently, CKLHAF material, $Li_2Cr_3SbO_8$ was synthesized. $Li_2Cr_3SbO_8$ has a $P6_3mc$ space group (No186, hexagonal), where magnetic Cr^{3+} ions (t_{2g}^3, $S = 3/2$) locate at $6c$ site [$(x, -x, z)$ with $x = 1/6$ and $z = 0.2478$] and form a "perfect" kagome lattice. Since there are non-magnetic Li^+ and Sb^{5+} layers between kagome layers, the kagome planes have good two-dimensional feature. In spite of the large negative Curie-Weiss temperature $\Theta_{CW} = -541.3$ K, no long-range order was observed down to 0.1 K confirmed by μSR. There is no difference between zero field cool and field cool in magnetic susceptibility, suggesting that spin glass state is ruled out from the ground state. Furthermore, magnetization measurements indicate the negligible DM interaction. The ground state of $Li_2Cr_3SbO_8$ is considered to be classical spin liquid.

Polycrystalline $Li_2Cr_3SbO_8$ was prepared by conventional solid state reactions for neutron experiments. 8 g powder was put into the annular can. Inelastic neutron scattering experiments were done at time-of-flight (TOF) neutron spectrometer PELICAN in ANSTO. Measurements were performed at 1.5, 15, 30, 60, and 120 K with $E_i = 10.9$ meV and 1.5, 4.5, 15, and 30 K with 3.63 meV. Empty can was also measured for both E_i, which was subtracted from the raw data. Vanadium was also measured for correction.

Fig. 1(a) shows neutron diffraction patterns with the energy window of $\hbar\omega = [-0.25, 0.25]$ meV at 1.5 and 15 K using $E_i = 10.9$ meV. Broad peaks was observed at $Q \sim 0.7$ and 1.4 Å^{-1}, corresponding to $(1/3, 1/3, 0)$ and $(2/3, 2/3, 0)$ positions, respectively. The broad peaks suggest the existence of short-range correlations. Fig. 1(b) shows inelastic neutron scattering map in $Li_2Cr_3SbO_8$ at $T = 1.5$ K with $E_i = 10.9$ meV. Quasielastic magnetic excitation centered at $Q = 1.3$ Å^{-1} was observed up to about 4 meV. Furthermore, additional magnetic excitation at $\hbar\omega = 7$ meV was also observed. To discuss the nature of the low-energy magnetic excitation, Q dependence of the magnetic excitations at $\hbar\omega = [1, 3]$ meV at 1.5 K is plotted in Fig. 1(c). The Q dependence can be well reproduced by the simulation assuming the 3×3 short-range magnetic order. This feature suggests that low-energy magnetic excitations observed in $Li_2Cr_3SbO_8$ originate in the classical spin liquid feature of $Li_2Cr_3SbO_8$. Interestingly, the maximum intensity of the Q dependence shifts to lower Q with increasing temperature (120 K), which is a common feature of the "quantum spin liquid" systems such as spin-1/2 kagome $ZnCu_3(OD)_6Cl_2$ and spin-1/2 triangular $YbMgGaO_4$. Note that peaks positions at $Q = 2.8$ and 3.6 Å^{-1} correspond to the nuclear Bragg peaks, suggesting that these peaks are phonon. On the other hand, Q dependences of the magnetic excitations at $\hbar\omega = [6.5, 8.5]$ meV at 1.5 and 120 K are also plotted in Fig. 1(d). These Q dependences cannot be explained by simple magnetic form factor of Cr^{3+} ions and we need to add another component from the classical spin liquid, indicating that the higher energy mode originates in combination of the crystal field and classical spin liquid. The energy dependences of the classical spin liquid excitation at $Q = [1, 1.8]$ Å^{-1} are also plotted in Fig. 1(e). Surprisingly, the energy spectrum at 1.5 K shows the spin gap which disappear above 15 K. Using the relationship, $\chi''(\hbar\omega) = I(\hbar\omega)(1 - e^{-\hbar\omega/k_BT})$, we also obtained the imaginary part of dynamical susceptibility $\chi''(\hbar\omega)$. Finally, we plotted the scaling plot ($\chi''(\omega)T$ vs ω/T) in Fig. 1(f). The experimental data well fitted by $\chi''(\hbar\omega)k_BT^{\alpha} \propto (k_BT/\hbar\omega)^{\alpha}\, tanh\,(\hbar\omega/\beta k_BT)$ with $\alpha = 0.840(1)$ and $\beta = 1.118(8)$.

Since $Li_2Cr_3SbO_8$ is the only material whose ground state is the classical spin liquid among the CKLHAF systems, we believe that our findings on the magnetic excitations in $Li_2Cr_3SbO_8$ can shed light on the classical spin liquid state.

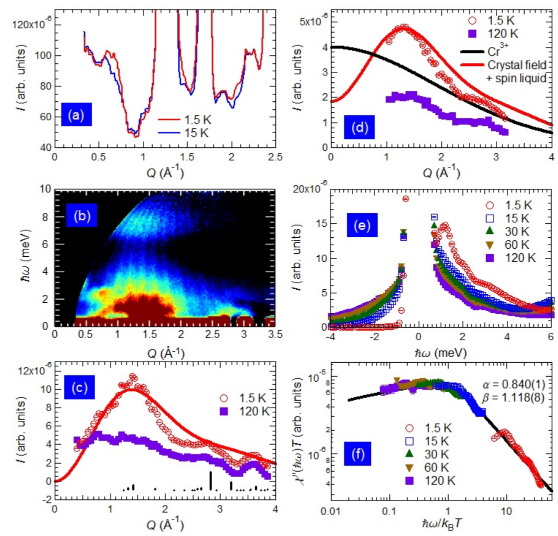

Fig. 1. (a) Neutron diffraction patterns. (b) Inelastic neutron scattering map. (c) Q dependences at 2 meV. (d) Q dependences at 7.5 meV. (e) Energy dependences at 1.4 Å^{-1}. (f) Scaling plot.