Magnetic excitations in the spin-1/2 uniform triangular lattice $Li_2(In,Sc)Mo_3O_8$

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1. Introduction

Recently, molecular-based triangular lattice systems have attracted research attention, one of which is $Li_2AMo_3O_8$ (A = In or Sc) [Y. Haraguchi *et al*., Phys. Rev. B **92**, 014409 (2015).]. In $Li_2AMo_3O_8$, seven 4*d* electrons in the Mo_3O_{13} cluster occupy their orbitals, resulting in one unpaired spin (S = 1/2) per one Mo_3O_{13} cluster. Mo_3O_{13} clusters form a uniform triangular lattice. Since molecular-based magnets are robust against defects, site disorder, and Jahn-Teller distortion, $Li_2AMo_3O_8$ is believed to be ideal playgrounds for S = 1/2 uniform triangular lattice model. Inverse magnetic susceptibility clearly shows that magnetic long-range order develops below T_N = 12 K in $Li_2InMo_3O_8$, while shortrange order below 10 K in $Li_2ScMo_3O_8$. Similar difference in ground states in $Li_2AMo_3O_8$ was also found in heat capacity measurements; a lambda-shaped peak was observed at 12 K in $Li_2InMo_3O_8$, while $Li_2ScMo_3O_8$ shows a broad peak at ~10 K. ^{7}Li NMR measurement suggested that magnetic structure in $Li_2InMo_3O_8$ is so-called 120° structure as expected in a perfect triangular lattice system. In the meanwhile, magnetic specific heat in $Li_2ScMo_3O_8$ shows a T-linear dependence with γ = 35.7 mJK^{-2}mol^{-1} below 2 K, suggesting that $Li_2ScMo_3O_8$ undergoes a quantum spin liquid ground state. However, magnetic excitations in $Li_2AMo_3O_8$, which provides us more detailed information on the magnetism in $Li_2AMo_3O_8$, were never investigated, and thus, we have measured the magnetic excitations using the chopper spectrometer 4SEASONS.

2. Experiment

In the current experiment, we have measured $Li_2ScMo_3O_8$ with mass of about 4.5 g. Plate-type sample can was used to minimize the effect of neutron absorption by Li. Using the ^{3}He cryostat, we performed measurements at T = 0.3, 10, and 22 K. (We also measured at T = 1.7, 5, 15, and 40 K in June 2017.) Frequency of the Fermi chopper was 250 Hz, resulting in the combination of incident neutron energies of 7.5, 10.3, 15.0, and 23.9 meV. Empty can was also measured, and then subtracted from the raw data.

3. Results

No magnetic Bragg peak develop in the elastic channel down to 0.3 K in $Li_2ScMo_3O_8$ [Fig. 1(c)]. Furthermore, diffuse scattering which is the signature of a spin glass ground state was not observed at 0.3 K [Fig. 1(c)]. On the other hand, the magnetic fluctuations were observed at the inelastic channel as in other quantum spin liquid systems. To explore the spin dynamics in $Li_2ScMo_3O_8$, Figures 1(a) and 1(b) depict $I(Q, \hbar\omega)$ maps at 0.3 and 22 K. Although both magnetic excitations in $Li_2InMo_3O_8$ and $Li_2ScMo_3O_8$ are centered at $Q \sim 0.7$ Å^{-1}, the overall structures are completely different, representing the different ground states. In $Li_2ScMo_3O_8$, nondispersive magnetic excitation extends out to at least 5 meV. As shown in Fig. 1(d), the Q dependences of the magnetic excitations are invariant in the different energy windows. The nondispersive magnetic excitation is the characteristic feature of the magnetic excitations in the quantum spin liquid systems. Furthermore, spectrum weight of the Q dependence at 2 meV shifts to Q = 0 at high temperature [Fig. 1(e)], which is also observed in other quantum spin liquid system candidates.

To investigate the characteristic energy scale of the magnetic excitation, the dynamical spin susceptibilities,

$\chi''(\hbar\omega) = [1 - \exp(-\hbar\omega/k_BT)]/|F(Q)|^2 I(\hbar\omega)$, at Q = [0.6, 0.8] Å^{-1} where the magnetic signal is maximal are plotted for different temperatures in Fig. 1(f). The spectra are well fitted by the quasielastic Lorentzian $\chi''(\hbar\omega) = \chi'\hbar\omega\Gamma/[(\hbar\omega)^2+ \Gamma^2)]$ where χ' is the static susceptibility and Γ the inverse relaxation rate. The temperature dependences of the fitting parameters are summarized in Fig. 1(g). With temperature decreasing, the characteristic energy Γ (or peak position) decreases while the χ' increases. Contrary to the conventional long-range ordered magnets, no divergence behavior was observed in χ' and Γ. These fittings also exhibit two important features: (1) the dynamical spin susceptibilities survives up at least 9.5 meV which is about 1.6J_1 determined by the magnetic susceptibility measurement and (2) the magnetic excitation is gapless combined with the heat capacity measurement. The Q position which shows the maximum intensity (Q ~ 0.7 Å^{-1}) corresponds to the (100) position. Furthermore, as shown in Fig. 1(f), the magnetic excitation in $Li_2ScMo_3O_8$ is gapless. These two features are consistent with the Spinon Fermi surface model [B. Fak *et al*., Phys. Rev. B **95**, 060402(R) (2017).].

4. Conclusion

In summary, we have investigated the magnetic excitations of Mo_3O_{13} cluster-based spin-1/2 triangular lattice system, $Li_2ScMo_3O_8$. Gapless quantum spin liquid behaviors were observed in $Li_2InMo_3O_8$, which can be understood by the spinon Fermi surface model.

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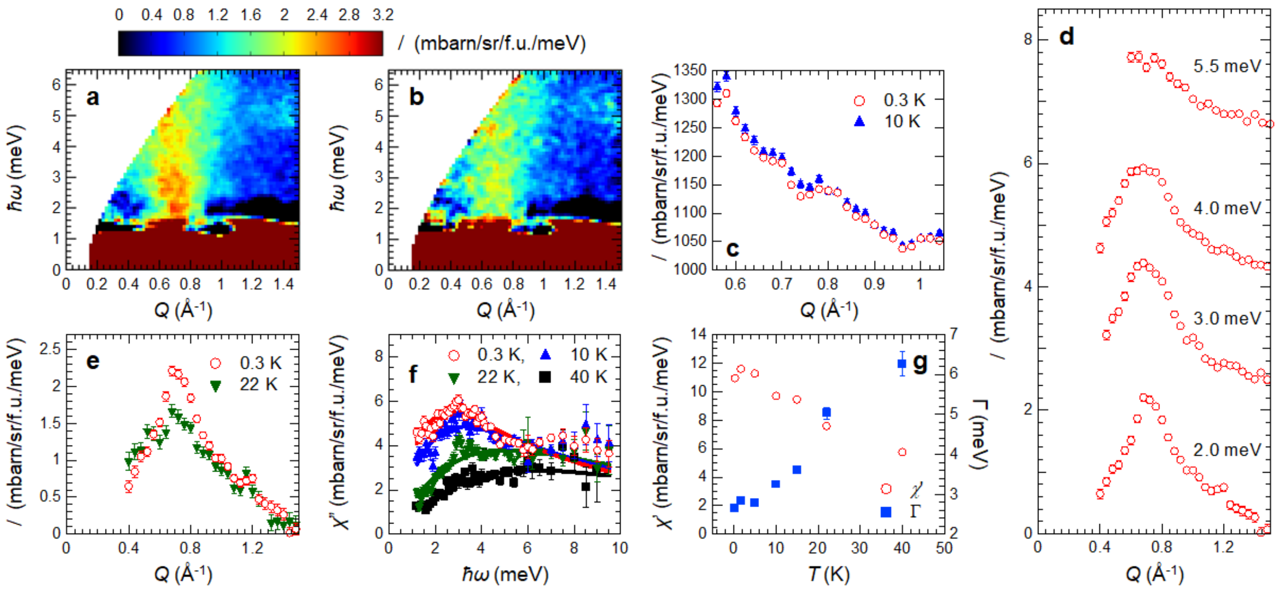

Figure 1. Neutron scattering results on $Li_2ScMo_3O_8$. Inelastic neutron scattering intensity maps with E_i = 10.3 meV measured at (a) 0.3 K and (b) 22 K. (c) Elastic neutron scattering intensities with energy window of [–0.075, 0.075] meV at 0.3 and 10 K with E_i = 10.3 meV. (d) Q dependences of the neutron scattering intensities at several energy windows at 0.3 K. Each energy window was [1.5, 2.5] meV with E_i = 7.5 meV, [2.5, 3.5] meV with E_i = 10.3 meV, [3.5, 4.5] meV with E_i = 15.0 meV, and [4.5, 6.5] meV with E_i = 23.9 meV, respectively. (e) Q dependences of the neutron scattering intensities at [1.5, 2.5] meV at 0.3 and 22 K with E_i = 7.5 meV. (f) Dynamical spin susceptibilities at Q = [0.6, 0.8] Å^{-1} for 0.3, 10, 22, and 40 K. Solid lines are the fitting results by the quasielastic Lorentzian as described in the main text. (g) Temperature dependences of static spin susceptibility χ' (left scale) and inverse relaxation rate Γ (right scale) obtained by the fitting in panel (f).