MLF Experimental Report	Date of Report
Project No. 2011B0064 Title of experiment Temperature dependence of spin gap in hexagonal multiferroic YMnO3 Name of principal investigator Je-Geun Park Affiliation Seoul National University	Name of person responsible for instrument Kenji Nakajima Name of Instrument/(BL No.) AMATERAS/(BL 14) Date of Experiment 2012/05/03 ~2012/05/07

Please report your samples, experimental method and results, discussion and conclusions. Please add figures and tables for better explanation.

1. Name of sample(s) and chemical formula, or compositions including physical form.

Yttrium manganite($YMnO_3$) powder

Lutetium manganite($LuMnO_3$) powder

2. Experimental method and results. If you failed to conduct experiment as planned, pleas describe reasons.

The main focus of the experiment was to explain the anomalous broad feature in the magnetic specific heat of (Y/Lu)MnO_3 around 50 K by measuring the temperature dependence of magnetic density of states (DOS). The standard repetition rate multiplication (RRM) method was used with incident beam energies of 42, 15, 8, and 5 meV. The temperature was varied from 10 K to approximately 150 K in steps of 5 K. Typical spectra are shown in figure 1. In the E_i=42meV data, the magnetic signal was taken from the Q range between 1~2 $Å^{-1}$. The background signal was estimated from the Q range between 5~6 $Å^{-1}$ and subtracted. In the 15meV data, the magnetic signal was also taken from the Q range between 1~2 $Å^{-1}$. However, as the background appeared to be minimal in this case, no background subtraction was carried out. Whilst the 42 meV data is necessary to obtain information on the full dispersion of the spin waves, the low energy gapped feature is better seen in the 15 meV data. Therefore, the derived DOS shown in figures 2 and 3 used the 15meV data for the energy range 2~8 meV and 42 meV data for larger energy. In contrast to our expectations, no drastic change in the DOS is observed around 50 K.

The data was modeled assuming the system behaved as a 2D triangular Heisenberg antiferromagnet with easy plane anisotropy D_1 and easy axis anisotropy D_2. Figure 4 shows the calculated dynamical structure factor $S(Q,\omega)$ for both $YMnO_3$ and $LuMnO_3$. The parameters used are shown in the table 1. The calculations show many similarities with the experimental data except for the strong peak at the maximum energy of the spin waves. This may be related to the spontaneous magnon decay observed in a recent single crystal $LuMnO_3$ inelastic neutron scattering measuremnt.

The low momentum transfer Q-integrated intensity measured by inelastic neutron scattering $S(\omega)$, is approximately proportional to the product of the structure factor f and the magnon density of states. We estimated f from the ratio of calculated S(q,w) and DOS, and subsequently used this to derive the measured DOS from the measured $S(\omega)$. This is compared to the DOS calculated from the model Hamiltonian in figures 5 and 6. The specific heat calculated from these DOS is compared to the measured specific heat in the figures 7 and 8. For $YMnO_3$, the estimated magnon contribution to the specific heat is in a good agreement up to 20 K. However, the measured magnetic specific heat of $LuMnO_3$ is much larger than the estimated magnon contribution from the measured DOS even at low temperatures. This puzzling deviation

may point to an explanation of the 50 K heat capacity anomaly and will require further study.

We also calculated the total magnetic energy of (Y/Lu)MnO_3 from the model Hamiltonian. The ground state energy of 2D triangular Heisenberg antiferromagnet is about $-3JS^2/2$ per spin[1], which corresponds to −1440J per mole for $YMnO_3$ and −1900J per mole for $LuMnO_3$. However, the total magnetic energy calculated from the magnetic specific heat is 817J per mole for $YMnO_3$ and 1028J per mole for $LuMnO_3$. This may either be due to an overestimation of the phonon heat capacity in the Debye model, or because the large spin–lattice coupling observed in these compounds may play an important role in specific heat[2].

In conclusion, no drastic change in the magnetic DOS was observed which could otherwise account for the broad anomaly in the specific heat. Nonetheless, there are several discrepancies between the microscopic view from inelastic neutron scattering and the macroscopic specific heat, which necessitates further investigation.

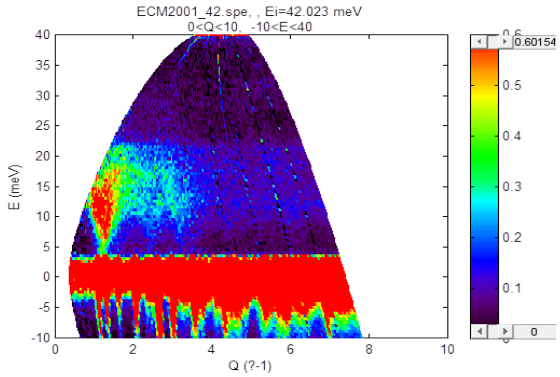

Fig 1. Data of $YMnO_3$ at 10 K with Ei=42 meV.

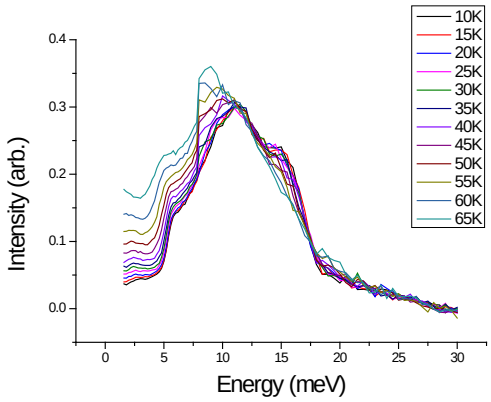

Fig 2. Combined data for $YMnO_3$.

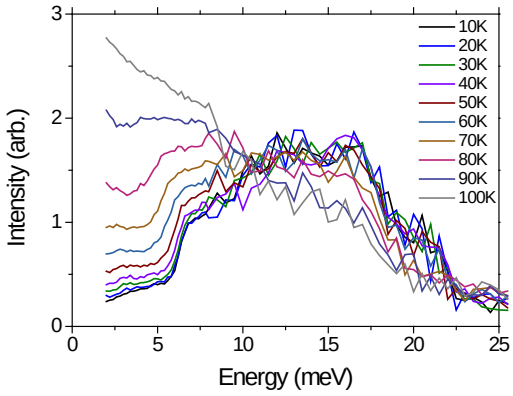

Fig 3. Data of $LuMnO_3$.

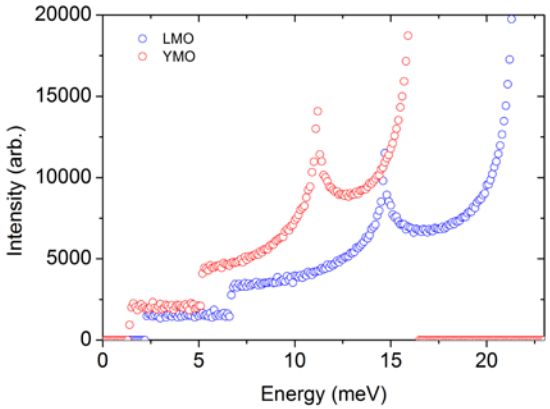

Fig 4. Calculated structure factor using model Hamiltonian.

[1] A.L.Chernyshev *et al.*, Phys. Rev. B **79**, 144416 (2009)
[2] S.Lee *et al.*, Nature **451**, 805 (2008)

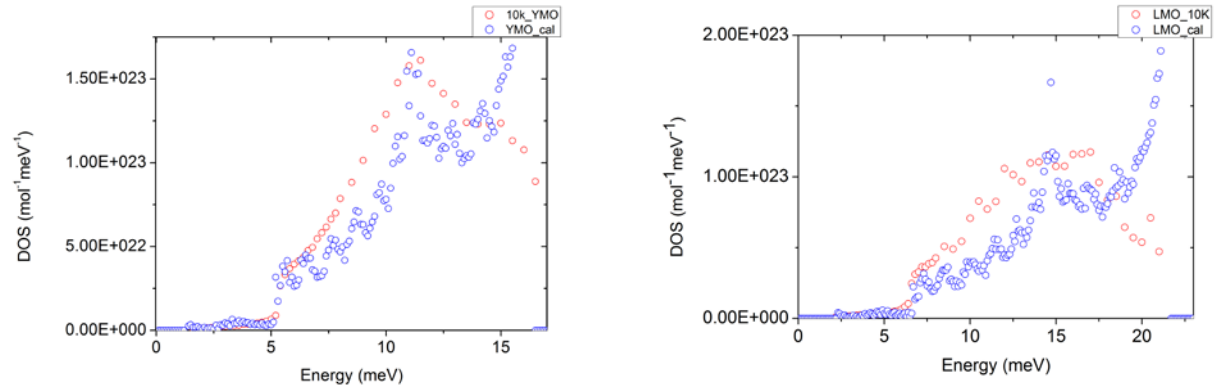

Fig 5 & 6. Calculated DOS from the model Hamiltonian (blue) and estimated DOS from the 10 K intensity (red) for $YMnO_3$ (left) and $LuMnO_3$ (right).

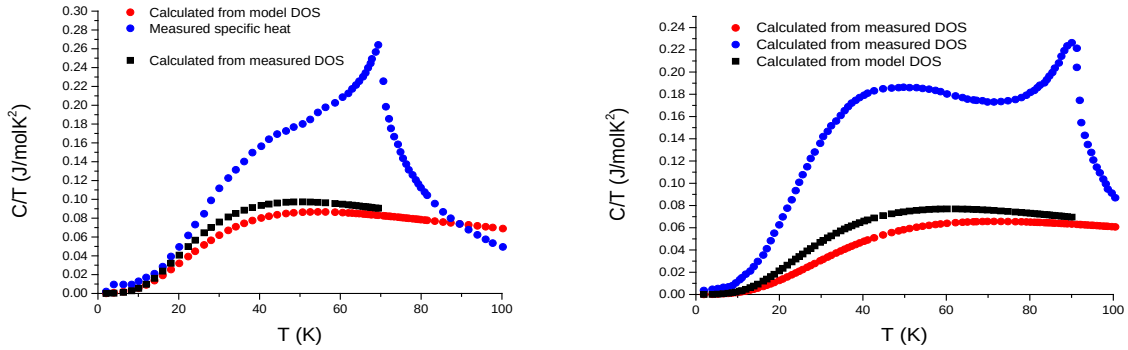

Fig 7 & 8. The measured magnetic specific heat[3] (blue) compared to the magnon contribution calculated using DOS from the 10K data (black) and from the model Hamiltonian (red) for $YMnO_3$ (left) and $LuMnO_3$ (right)

	J (meV)	D_1 (meV)	D_2 (meV)
$YMnO_3$	-2.5	-0.28	0.01
$LuMnO_3$	-3.3	-0.35	0.02

Table 1. The parameters of model Hamiltonian. The parameters for YMnO3 is similar to the previous report[4].

[3] D. G. Tomuta *et al.*, J. Phys. Condens. Matter **13** (2001) 4543–4552
[4] T. Chatterji *et al.*, Phys. Rev. B 76, 144406 (2007)