Magnetic Properties of Geometrically Frustrated Double Perovskites, Ba_2YRuO_6, Ba_2YMoO_6 and La_2LiReO_6

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We have carried out neutron diffraction measurements on La_2LiReO_6, Ba_2YMoO_6 and Ba_2YRuO_6 using the C2 powder diffractometer at the Canadian Neutron Beam Centre. We expected that no magnetic peaks would appear in the first two compounds, since there were no signatures of long range ordering according to magnetic susceptibility measurements. On the other hand, Ba_2YRuO_6 was expected to show some feature of antiferromagnetic long range ordering since the heat capacity data shows a very sharp λ-type peak around 37 K, which is consistent with magnetic susceptibility results.

Ba_2YRuO_6 crystallizes in the cubic space group Fm-3m [1]. The refinement for the crystal structure with the data collected using longer neutron wavelength and FULLPROF program [2] was done and the cell constant is determined to be 8.332(1) Å at room temperature and 8.3194(7) Å at 2.7 K. R-factors in the refinements for each temperature are; R_p = 4.90, R_{wp}= 6.45 for room temperature, and R_p= 4.54, R_{wp}= 6.88, magnetic R-factor = 20.5 for 2.7 K. These factors are reasonably low, meaning these are good refinements.

The magnetic susceptibility results collected at McMaster University using SQUID magnetometer (Quantum Design) show two shoulder-like peaks at 47 K and 37 K for this compound, which may be the transition temperatures, and the Curie-Weiss constant is calculated to be θ = -522 K. However the heat capacity measurement shows a λ-type peak around 37 K only. This could be explained as antiferromagnetic short range ordering occurring first around 47 K, following by the formation of complete antiferromagnetic long range ordering at 37 K.

The evolution of magnetic peaks in the diffraction patterns of Ba_2YRuO_6 is shown in Figure 1. As one can see, two magnetic peaks appear around 2θ = 16° and 23° below about 35 K. The magnetic structure was also refined and the two magnetic peaks at 2θ = 16° and 23° could be indexed as k = (0, 0, 1), and (1, 1, 0), respectively. According to this result, the actual transition temperature of Ba_2YRuO_6 is determined to be 37 K. The magnetic structure of this compound was confirmed to be face-centered cubic and Type I structure, which was reported by Battle *et al.* [1].

As well as Ba_2YRuO_6, Ba_2YMoO_6 also crystallizes into the same cubic symmetry (space group Fm-3m) [3]. Since the magnetic susceptibility result shows the lack of long range ordering in the temperature region from 300 K to 2 K and Curie-Weiss constant was determined to be θ = -394 K, there should not be any new magnetic peaks appearing. This result was confirmed based on the comparison of diffraction patterns collected using longer neutron wavelength (2.37Å) at 2.7 K and room temperatures. The absence of antiferromagnetic long range ordering also agreed with the result of heat capacity collected at McMaster University. The crystal structures at 298 K and 2.7 K were refined and determined to have a cubic structure with cell constants, 8.3904(5) Å and 8.3784(5) Å, respectively. R factors are: R_p= 4.87, R_{wp}= 6.36 and χ^2 = 2.87. Since the Mo^{5+} ion has d^1 electronic structure, it could result in Jahn-Teller distortion and transform to a lower symmetry. However, the refinement with cubic symmetry shows good agreement and it is reasonable to say that Ba_2YMoO_6 remains cubic structure at 2.7 K.

We also have performed neutron diffraction measurements on the compound, La_2LiReO_6. The structure is monoclinic with space group $P2_1/n$. The magnetic susceptibility result shows a ZFC/FC divergence around 50 K, a broad maximum in the ZFC curve at 33 K and the Weiss temperature is θ=-163 K. Therefore, antiferromagnetic interaction exists. However, the heat capacity data did not show any peaks or shoulders. Thus, there are not any features indicating the presence of antiferromagnetic long or short range ordering. The comparison of two diffraction patterns collected at room and base temperatures supports the absence of antiferromagnetic long range ordering. Crystal structure refinement with the data collected at 298 K determined the structure parameters to be a = 5.5819(6) Å, b = 5.6743(6) Å, c = 7.8840(8) Å, β = 90.243° and total cell volume is 249.7117 $Å^3$. R factors are: R_p = 3.39, R_{wp} = 4.47 and χ^2 = 3.01. The structure refinement was also done with the data collected at 2.8 K and determined the cell parameters to be a = 5.5801(6) Å, b = 5.6859(6) Å, c = 7.8421(9) Å, β = 90.479° and total cell volume is 248.8079$Å^3$. R factors are: R_p = 4.17, R_{wp} = 5.72 and χ^2 = 6.10.

In summary, the neutron diffraction experiment confirmed the presence of antiferromagnetic long range ordering and revealed the transition temperature in Ba_2YRuO_6 and the absence of long range ordering with Ba_2YMoO_6 and La_2LiReO_6 is confirmed, as expected.

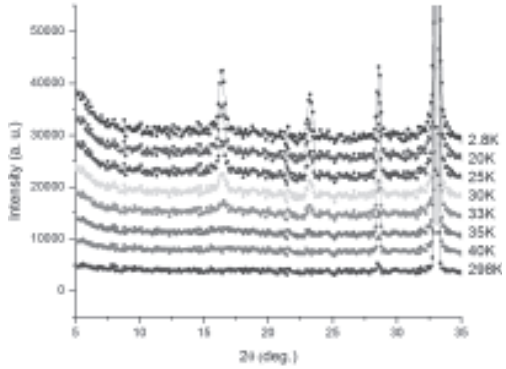

Fig. 1 Evolution of magnetic peaks of Ba_2YRuO_6

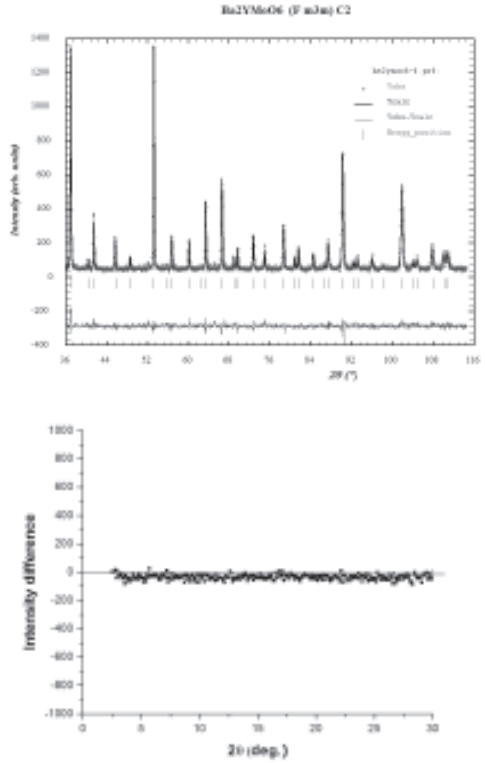

Fig. 2 Crystal structure refinement of Ba_2YMoO_6 at 2.7 K (upper) and the intensity difference of the diffraction patterns at 2.7 K and 298 K (lower). There is no difference between two patterns.

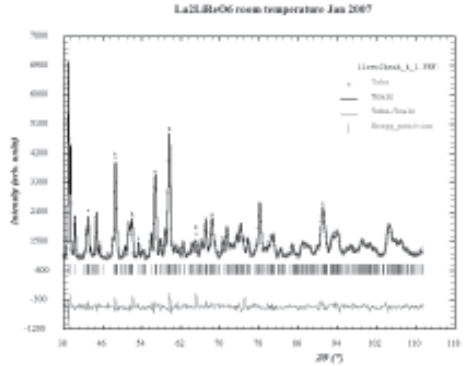

Fig. 3 Crystal structure refinement for La_2LiReO_6 (298 K)

References

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