Using Muon Spin Relaxation to Investigate Unconventional Magnetism in Double Perovskites

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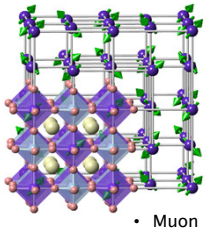
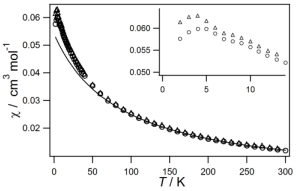
Competing magnetic interactions within a lattice, often arising from spins arranged on triangular plaquettes, is known as geometric frustration and can lead to highly unusual magnetic ground states. The face-centred cubic lattice, containing edge sharing tetrahedra, exhibits a high degree of frustration. The *B*-site cation ordered double perovskite ($A_2BB'O_6$) is one such physical realisation of a face-centred cubic lattice, which affords a great degree of diversity due to the variety of cations that can be accommodated within the structure.

Ba_2MnMoO_6

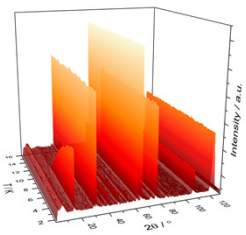
- Double perovskite.
- Mo^{6+} ($S = 0$) & Mn^{2+} ($S = ⅝$) ordered on *B*-site.
- Mn^{2+} on geometrically frustrated, face-centred cubic (FCC) lattice - edge sharing tetrahedra.





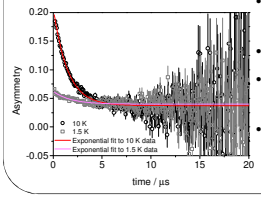
D.C.-susceptibility measurements (in 1000 G):

- Mn^{2+} cations strongly antiferromagnetically coupled, $\theta = -85$ K.
- Small cusp at 5 K.

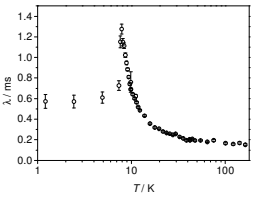


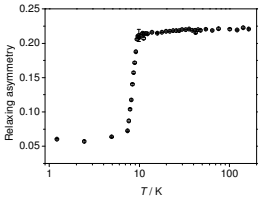
Neutron powder diffraction:

- Cubic space group $Fm\bar{3}m$.
- Cubic symmetry maintained on cooling to 1.5 K.
- Magnetic Bragg peaks appear at 9.4 K.
- Propagation vector: **k** = (½, ½, ½).
- Non-collinear spin structure:
  - 4 interpenetrating G-type afm lattices.
  - orientated along 4 different <111> directions.
  - contains spin compensated tetrahedra.



- Muon relaxation fitted to exponential function $T > 7.8$ K - paramagnetic behaviour.
- Slowing down of relaxation on cooling to T_N.
- $T < 7.8$ K - significant loss of relaxing asymmetry, but no oscillations are observed.
- Indicative of muons oscillating on timescale faster than can be detected at ISIS, due to strong local magnetic fields.

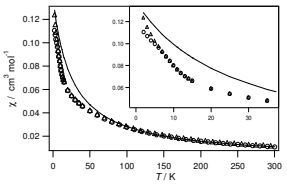




- μSR (μs timescale) shows T_N in Ba_2MnMoO_6 intermediate between susceptibility and neutron diffraction measurements:
  - degeneracy in ground state.

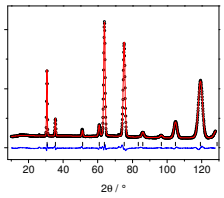
$Ba_2Nd_{0.3}Mn_{0.7}MoO_6$

- Double perovskite.
- Nd^{3+} & Mn^{2+} solid solution on one *B*-site.
- $Mo^{5.7+}$ on *B'*-site.[1]



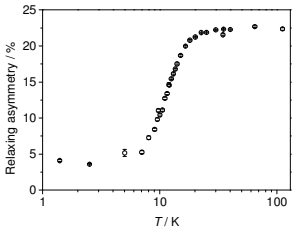
D.C.-susceptibility measurements (in 1000 G):

- Curie-Weiss behaviour, antiferromagnetic: $\theta = -26$ K.
- Small deviation of field cooled and zero field cooled susceptibility at ~5 K.



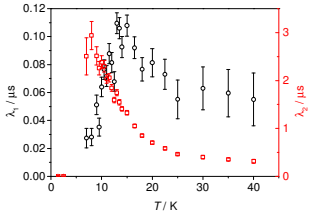
Neutron powder diffraction:

- Cubic space group $Fm\bar{3}m$ despite presence of Jahn-Teller active Mo^{5+}.
- Cubic symmetry maintained on cooling to 1.5 K.
- No evidence of long range magnetic order $T > 1.5$ K.

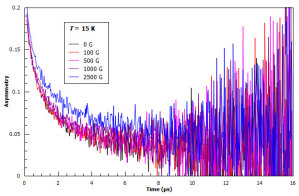


μSR:

- $Ba_2Nd_{0.3}Mn_{0.7}MoO_6$, shows a similar loss in asymmetry as Ba_2MnMoO_6, over broader temperature range.
- Two relaxing components required to fit data.

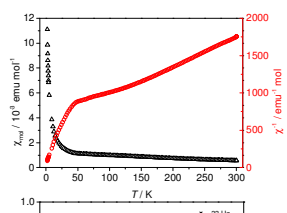


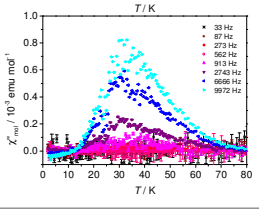
- Two relaxation rates exhibit maxima at two different temperatures (8 K & 14 K).
- At 15 K (paramagnetic regime in susceptibility data), muon spin still strongly coupled – not decoupled in applied longitudinal field of 2500 G.

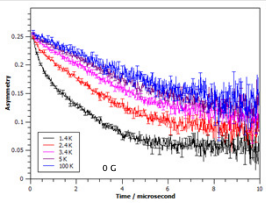


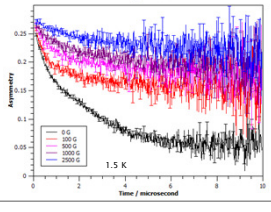
Ba_2LuMoO_6

- Double perovskite.
- Mo^{5+} ($S = ½$) & Lu^{3+} ($S = 0$) ordered on *B*-site.
- Mn^{2+} on geometrically frustrated, FCC lattice - edge sharing tetrahedra.
- Cubic symmetry maintained & no long range magnetic order at $T \geq 1.5$ K.
- Strong antiferromagnetic interactions:
  - $\theta = -129$ K at $T > 50$ K.
- Large loss of moment at $T \sim 50$ K.[2]
- A.c.-susceptibility shows peak in imaginary part at $T \sim 50$ K:
  - sharpens with increasing frequency.









- Muon relaxation fitted to exponential function at $T = 100$ K - paramagnetic behaviour.
- Slowing down of relaxation on cooling to 1.5 K.
- At low *T*, 2 components to relaxing asymmetry – 1 component showing glassy behaviour.
- At $T = 1.5$ K, muon spin strongly couples to Mo^{5+} spin – not decoupled in applied longitudinal field of 2500 G.
- Behaviour indicative of formation of a sea of spin singlets and remaining isolated spins, exhibiting glassy dynamics:
  - so called valence bond glass behaviour.

References

1. F. C. Coomer & E. J. Cussen, *Inorg. Chem.*, 2014, **53**(2), 746-755.
2. F. C. Coomer & E. J. Cussen, *J. Phys.: Condens. Matter*, 2013, **25**, 082202.

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