Supplementary Information for "Valence bond glass state in the 4*d*^{1} fcc antiferromagnet Ba_2LuMoO_6"

O. H. J. Mustonen[1,2], H. M. Mutch[1], H. C. Walker[3], P. J. Baker[3], F. C. Coomer[4], R. S. Perry[5], C. Pughe[1], G. B. G. Stenning[3], C. Liu[6], S. E. Dutton[6] and E. J. Cussen[1*]

[1]*Department of Material Science and Engineering, University of Sheffield, Mappin Street, Sheffield, S1 3JD, United Kingdom*

[2]*School of Chemistry, University of Birmingham, Edgbaston, Birmingham B15 2TT, United Kingdom*

[3]*ISIS Pulsed Neutron and Muon Source, STFC Rutherford Appleton Laboratory, Harwell Campus, Didcot, OX11 0QX, United Kingdom*

[4]*Johnson Matthey Battery Materials, Blount's Court, Sonning Common, Reading, RG4 9NH United Kingdom*

[5]*London Centre for Nanotechnology and Department of Physics and Astronomy, University College London, Gower Street, London, WC1E 6BT, United Kingdom*

[6]*Cavendish Laboratory, University of Cambridge, Cambridge CB3 0HE, United Kingdom*

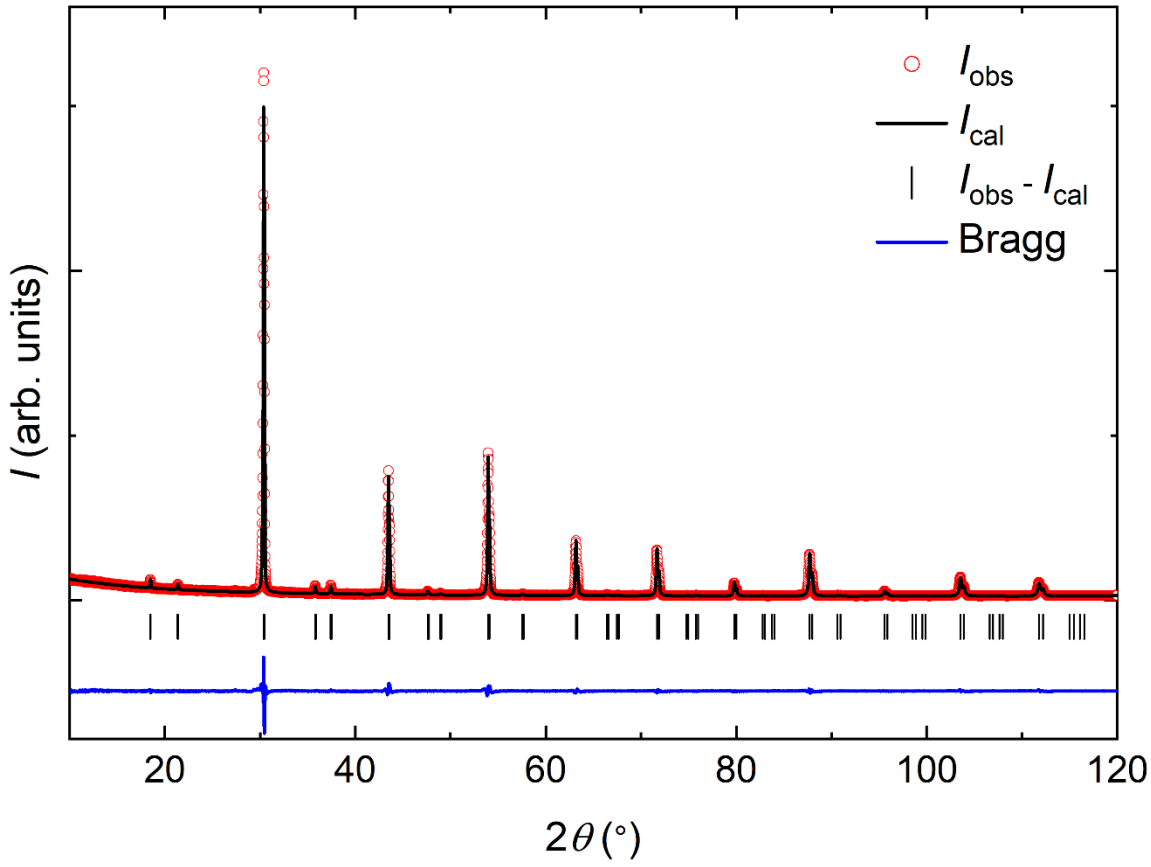
[*] Corresponding author

e.j.cussen@sheffield.ac.uk

Supplementary Note 1: X-ray Diffraction

Laboratory X-ray diffraction was used to evaluate the phase purity and crystal structure of our Ba_2LuMoO_6 sample. The material crystallises in the cubic $Fm\bar{3}m$ space group corresponding to an ordered double perovskite structure. No additional impurity peaks are observed in the diffraction pattern (Supplementary Figure 1).

Results of our Rietveld refinement are presented in Supplementary Table 1. The refined structure is in agreement with reported low-temperature neutron diffraction data[1]. The degree of *B*-site cation order was evaluated by refining Mo and Lu site occupancies with the total occupancies of Mo and Lu constrained to nominal stoichiometry and Mo on the Lu-site constrained to be the same as Lu on the Mo-site. Atomic displacement parameters of both *B*-sites were constrained to be the same. The displacement parameters are similar to those of Ba_2YMoO_6 at room temperature[2].



Supplementary Figure 1. Laboratory X-ray powder diffraction pattern of Ba_2LuMoO_6 at room temperature. No impurity peaks are observed.

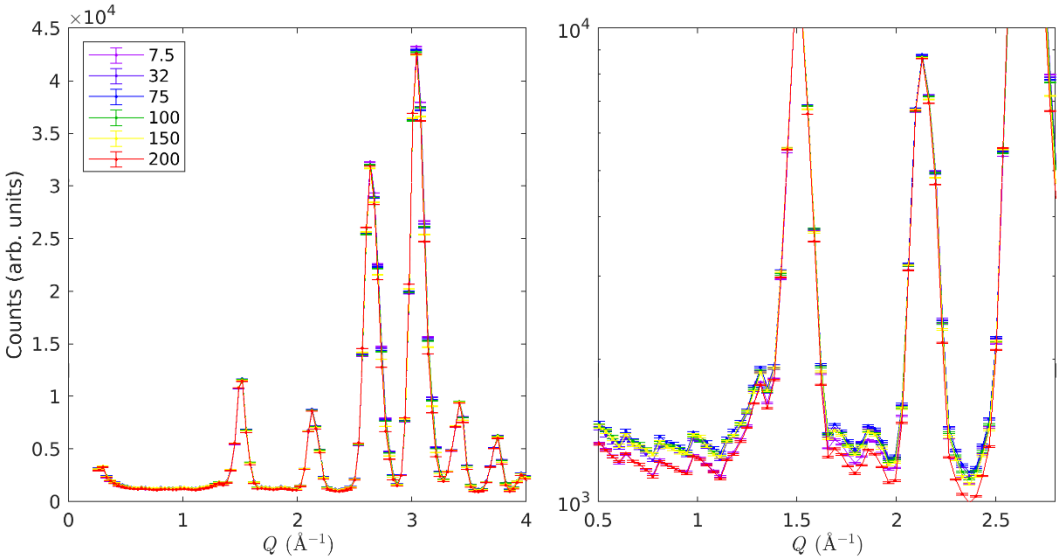
Supplementary Table 1. Refined crystal structure of Ba_2LuMoO_6 at room temperature. Space group $Fm\bar{3}m$ with a = 8.32058(3) Å, R_p = 13.7%, R_{wp} = 10.8% and χ^2 = 3.05.

Atom	X	y	z	B_{iso} (Å^2)	Occupancy
Ba	0.25	0.25	0.25	0.50(4)	1.00
Lu1	0	0	0	0.41(4)	0.99(1)
Mo1	0	0	0	0.41(4)	0.01(1)
Mo2	0.5	0.5	0.5	0.41(4)	0.99(1)
Lu2	0.5	0.5	0.5	0.41(4)	0.01(1)
O	0.2633(7)	0	0	0.7(1)	1.00

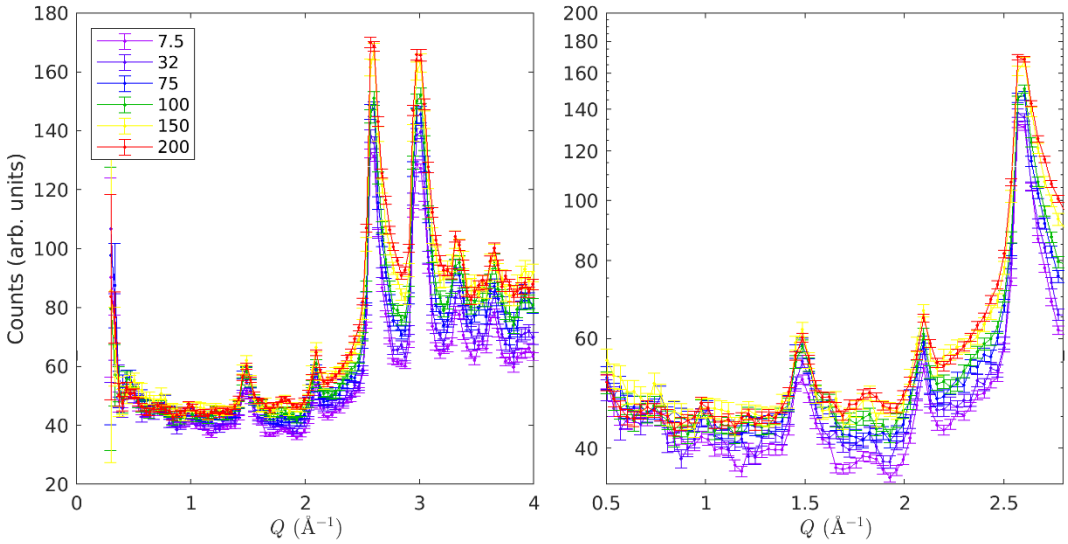
Supplementary Note 2: Cuts of the inelastic neutron scattering data

Our inelastic neutron scattering data shows a clear flat excitation at 28 meV. We have interpreted this as a singlet-triplet excitation consistent with a valence bond glass ground state. Another possible ground state for Ba_2LuMoO_6 is the disorder-induced random singlet state. The most structurally similar random singlet candidate is $Sr_2CuTe_{1-x}W_xO_6$, which is also a double perovskite. In $Sr_2CuTe_{1-x}W_xO_6$, significant scattering is observed at the $|Q|$ positions of the Bragg peaks of the magnetically ordered parent phases.[3] This scattering is very noticeable at $|Q|$ ~0.7-0.9 at low energies.

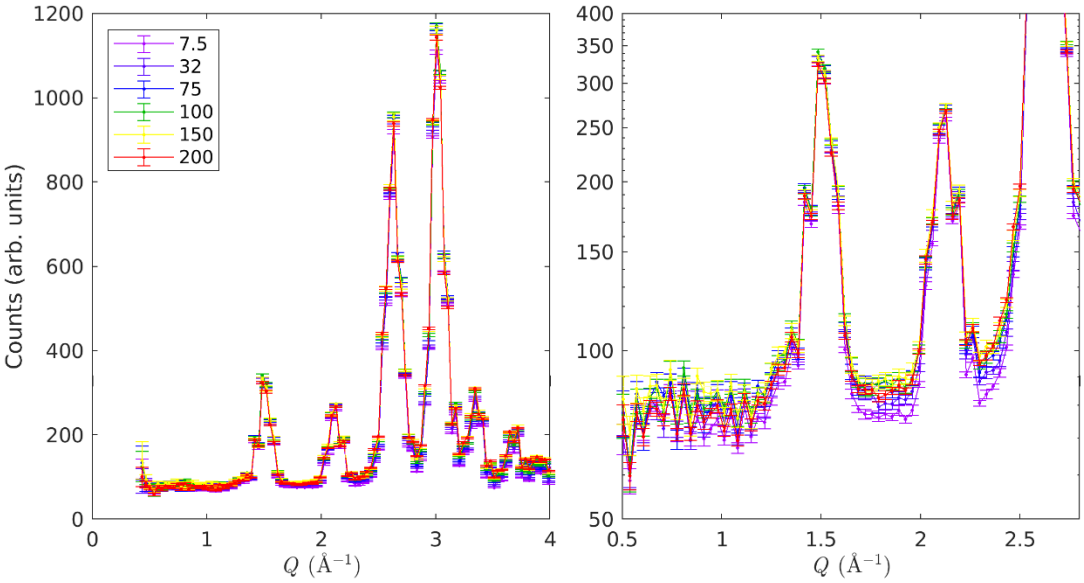
We do not observe such magnetic scattering for Ba_2LuMoO_6. The $|Q|$ positions of an ordered fcc antiferromagnet with the lattice parameter of Ba_2LuMoO_6 correspond to $|Q|$ = 0.75 Å^{-1} for Type I order and $|Q|$ = 0.65 Å^{-1} for Type II order. We do not observe any magnetic scattering at these positions in cuts of the elastic line (Supplementary Figure 2), in cuts between 3 meV < E < 5 meV (Supplementary Figure 3), cuts between 5 meV < E < 8 meV (Supplementary Figure 4) or in cuts between 8 meV < E < 12 meV (Supplementary Figure 5).



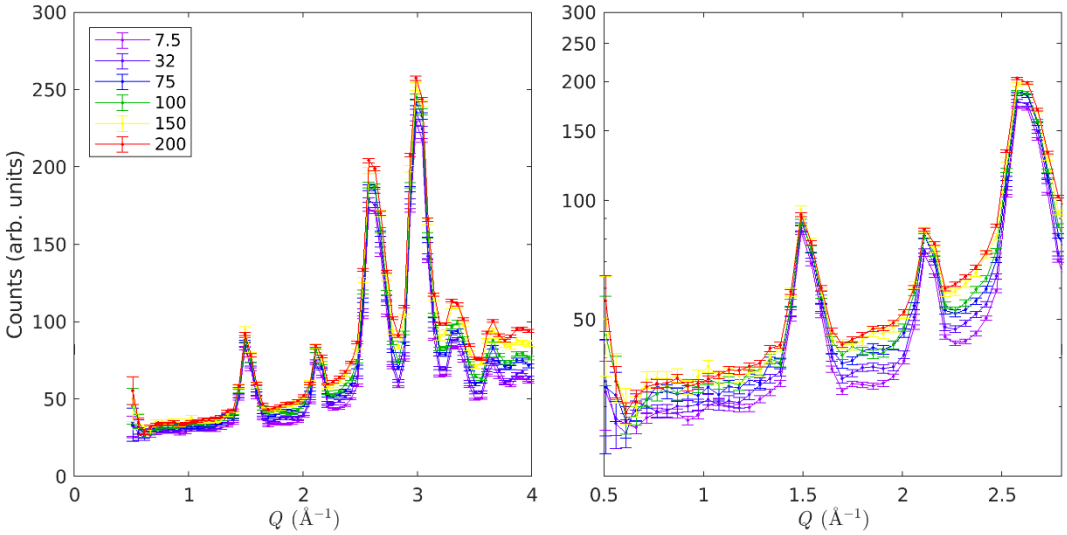
Supplementary Figure 2. Cuts of the elastic line from E_i = 30 meV data MERLIN data at different temperatures. No magnetic Bragg peaks are observed. The nuclear scattering from the elastic line bleeds into the inelastic data at low energies, which is why we present it for comparison.



Supplementary Figure 3. Cuts of the E_i = 30 meV MERLIN data between 3 and 5 meV at different temperatures. We do not observe magnetic scattering at the positions expected from a magnetically ordered fcc antiferromagnet at $|Q|$ = 0.75 Å^{-1} or $|Q|$ = 0.65 Å^{-1}.



Supplementary Figure 4. Cuts of the E_i = 70 meV MERLIN data between 5 and 8 meV at different temperatures. We do not observe magnetic scattering at the positions expected from a magnetically ordered fcc antiferromagnet at $|Q|$ = 0.75 Å^{-1} or $|Q|$ = 0.65 Å^{-1}.



Supplementary Figure 5. Cuts of the E_i = 70 meV MERLIN data between 8 and 12 meV at different temperatures. We do not observe magnetic scattering at the positions expected from a magnetically ordered fcc antiferromagnet at $|Q|$ = 0.75 Å^{-1} or $|Q|$ = 0.65 Å^{-1}.

Supplementary References

1. Coomer, F. C. & Cussen, E. J. Structural and magnetic properties of Ba_2LuMoO_6: A valence bond glass. *J. Phys. Condens. Matter* **25**, 082202 (2013).

2. Mclaughlin, A. C., De Vries, M. A. & Bos, J. W. G. Persistence of the valence bond glass state in the double perovskites $Ba_{2-x}Sr_xYMoO_6$. *Phys. Rev. B - Condens. Matter Mater. Phys.* **82**, 094424 (2010).

3. Fogh, E. *et al.* Randomness and Frustration in a *S* = 1/2 Square-Lattice Heisenberg Antiferromagnet. *Phys. Rev. B* **105**, 184410 (2022).