Supplementary Information:

Magnetic order arising from structural distortion: the Structure and Magnetic Properties of $\text{Ba}_2\text{LnMoO}_6$

Edmund J. Cussen, Danny R. Lynham and Joseph Rogers
The University of Nottingham*

Contents

- 1) Structural data for $\text{Ba}_2\text{NdMoO}_6$ at 10 K
- 2) Structural data for $\text{Ba}_2\text{NdMoO}_6$ at 18 K
- 3) Structural data for $\text{Ba}_2\text{NdMoO}_6$ at 32 K
- 4) Structural data for $\text{Ba}_2\text{SmMoO}_6$ at RT
- 5) Refined structural parameters for cubic phases $\text{Ba}_2\text{LnMoO}_6$ ($\text{Ln} = \text{Eu, Gd, Dy, Er, Y, Yb}$)
- 6) AC Molar Magnetic Susceptibility data for $\text{Ba}_2\text{LnMoO}_6$ ($\text{Ln} = \text{Eu, Dy, Er, Y, Yb}$)

Ba₂NdMoO₆ 10 K

Refined against neutron powder diffraction data

$I\bar{1}$: $a = 5.9758(2) \text{ \AA}$, $b = 5.9808(2) \text{ \AA}$, $c = 8.5978(2) \text{ \AA}$,

$\alpha = 89.855(2)^\circ$, $\beta = 90.049(5)^\circ$, $\gamma = 90.002(5)^\circ$ Vol. = $307.286(11) \text{ \AA}^3$

Moments: Nd $2.07(6) \mu_B$, Mo $0.17(5) \mu_B$ Ferrimagnetic in xy plane stacked antiferromagnetically along

z

	x	y	z	100U _{iso}
Ba	-0.0027(13)	0.493(2)	0.2574(6)	0.26(4)
Mo	0	0	0.5	0.39(3)
Nd	0	0	0	0.39(3)
O(1)	0.000(4)	0.003(2)	0.2659(3)	1.25(5)
O(2)	0.2349(12)	0.3112(12)	0.0051(6)	1.27(14)
O(3)	0.6926(11)	0.2333(12)	-0.0035(8)	0.63(12)

Fit parameters: $R_{wp} = 3.20$, $R_p = 2.54$, $R(F^2) = 4.60$,

D-W $d = 0.485$ (<1.910 confidence limit)

$\chi^2_{red} = 0.5439$ for 42 variables (6 lattice parameters, 12 positional parameters, 5 displacement parameters, 1 scale factor, 1 zero point error, 5 profile parameters, 10 background parameters, 2 magnetic moment)

Ba — O(1)	2.937(14)	Mo — O(1)	$2.012(3) \times 2$
Ba — O(1)	2.98(2)	Mo — O(2)	$1.945(7) \times 2$
Ba — O(1)	3.00(2)	Mo — O(3)	$1.967(7) \times 2$
Ba — O(1)	3.046(14)	Nd — O(1)	$2.287(3) \times 2$
Ba — O(2)	2.814(9)	Nd — O(2)	$2.333(7) \times 2$
Ba — O(2)	2.892(10)	Nd — O(3)	$2.307(7) \times 2$
Ba — O(2)	3.167(11)		
Ba — O(2)	3.262(8)	O(1) — Mo — O(2)	88.4(7)
Ba — O(3)	2.756(11)	O(1) — Mo — O(3)	89.3(3)
Ba — O(3)	2.767(8)	O(2) — Mo — O(3)	89.6(4)
Ba — O(3)	3.283(8)	O(1) — Mo — O(1)	180
Ba — O(3)	3.295(11)	O(1) — Nd — O(2)	88.5(5)
Mo — O(1) — Nd	179.1(8)	O(1) — Nd — O(3)	89.5(4)
Mo — O(2) — Nd	162.3(4)	O(2) — Nd — O(1)	89.5(4)
Mo — O(3) — Nd	163.0(4)	O(1) — Nd — O(1)	180

Ba₂NdMoO₆ 18 K

Refined against neutron powder diffraction data

$I\bar{1}$: $a = 5.9755(2) \text{ \AA}$, $b = 5.9806(2) \text{ \AA}$, $c = 8.5982(2) \text{ \AA}$,
 $\alpha = 89.854(2)^\circ$, $\beta = 90.058(4)^\circ$, $\gamma = 90.002(4)^\circ$ Vol. = $307.277(11) \text{ \AA}^3$

	x	y	z	100U _{iso}
Ba	-0.0009(12)	0.493(13)	0.2562(6)	0.28(4)
Mo	0	0	0.5	0.37(3)
Nd	0	0	0	0.37(3)
O(1)	0.003(3)	0.0044(13)	0.2652(3)	1.20(5)
O(2)	0.2297(11)	0.3128(10)	0.0049(6)	1.50(11)
O(3)	0.6934(8)	0.2381(10)	-0.0044(6)	0.40(8)

Fit parameters: $R_{wp} = 2.88$, $R_p = 2.23$, $R(F^2) = 3.82$,

D-W d = 0.436 (<1.908 confidence limit)

$\chi^2_{red} = 0.5439$ for 39 variables (6 lattice parameters, 12 positional parameters, 5 displacement parameters, 1 scale factor, 1 zero point error, 4 profile parameters, 10 background parameters)

Ba — O(1)	2.926(10)	Mo — O(1)	$2.019(3) \times 2$
Ba — O(1)	2.98(2)	Mo — O(2)	$1.947(7) \times 2$
Ba — O(1)	3.00(2)	Mo — O(3)	$1.966(7) \times 2$
Ba — O(1)	3.057(11)	Nd — O(1)	$2.280(3) \times 2$
Ba — O(2)	2.784(8)	Nd — O(2)	$2.320(7) \times 2$
Ba — O(2)	2.870(9)	Nd — O(3)	$2.320(7) \times 2$
Ba — O(2)	3.189(10)		
Ba — O(2)	3.292(8)	O(1) — Mo — O(2)	87.9(4)
Ba — O(3)	2.781(7)	O(1) — Mo — O(3)	89.0(3)
Ba — O(3)	2.790(10)	O(2) — Mo — O(3)	88.3(3)
Ba — O(3)	3.261(10)	O(1) — Mo — O(1)	180
Ba — O(3)	3.271(7)	O(1) — Nd — O(2)	88.0(3)
Mo — O(1) — Nd	178.3(7)	O(1) — Nd — O(3)	89.2(3)
Mo — O(2) — Nd	160.9(3)	O(2) — Nd — O(1)	88.4(3)
Mo — O(3) — Nd	164.2(3)	O(1) — Nd — O(1)	180

Ba₂NdMoO₆ 32 K

Refined against neutron powder diffraction data

$I\bar{1}$: $a = 5.9764(3) \text{ \AA}$, $b = 5.9795(3) \text{ \AA}$, $c = 8.5988(2) \text{ \AA}$,
 $\alpha = 89.860(4)^\circ$, $\beta = 90.041(9)^\circ$, $\gamma = 90.013(8)^\circ$ Vol. = $307.23(2) \text{ \AA}^3$

	x	y	z	100U _{iso}
Ba	-0.0003(2)	0.492(2)	0.2584(7)	0.22(7)
Mo	0	0	0.5	0.28(4)
Nd	0	0	0	0.28(4)
O(1)	0.001(8)	0.003(3)	0.2661(5)	1.35(8)
O(2)	0.253(2)	0.3074(14)	0.0049(9)	0.7(2)
O(3)	0.689(2)	0.225(2)	-0.0002(14)	1.1(2)

Fit parameters: $R_{wp} = 4.89$, $R_p = 3.85$, $R(F^2) = 4.93$, D-W d = 0.955 (<1.908 confidence limit)

$\chi^2_{red} = 0.5439$ for 39 variables (6 lattice parameters, 12 positional parameters, 5 displacement parameters, 1 scale factor, 1 zero point error, 4 profile parameters, 10 background parameters)

Ba — O(1)	2.929(2)	Mo — O(1)	$2.011(4) \times 2$
Ba — O(1)	2.98(5)	Mo — O(2)	$1.922(9) \times 2$
Ba — O(1)	3.01(5)	Mo — O(3)	$1.995(11) \times 2$
Ba — O(1)	3.05(2)	Nd — O(1)	$2.288(4) \times 2$
Ba — O(2)	2.854(13)	Nd — O(2)	$2.341(9) \times 2$
Ba — O(2)	2.932(14)	Nd — O(3)	$2.294(11) \times 2$
Ba — O(2)	3.123(16)		
Ba — O(2)	3.221(14)	O(1) — Mo — O(2)	88.4(14)
Ba — O(3)	2.694(15)	O(1) — Mo — O(3)	89.9(6)
Ba — O(3)	2.750(15)	O(2) — Mo — O(3)	87.7(5)
Ba — O(3)	3.302(15)	O(1) — Mo — O(1)	180
Ba — O(3)	3.356(16)	O(1) — Nd — O(2)	88.5(11)
Mo — O(1) — Nd	179.1(15)	O(1) — Nd — O(3)	89.8(8)
Mo — O(2) — Nd	164.9(5)	O(2) — Nd — O(3)	87.7(5)
Mo — O(3) — Nd	160.5(5)	O(1) — Nd — O(1)	180

Ba₂SmMoO₆ RT

Refined against X-ray powder diffraction data

I 4/mmm: a = b = 5.98954(6), c = 8.50030(12) Å Vol. = 304.945(8) Å³

	x	y	z	100 U _{iso}
Ba	0	¹ / ₂	¹ / ₄	0.58(4)
Mo	0	0	0	0.17(4)
Sm	0	0	¹ / ₂	0.17(4)
O(1)	0	0	0.232(2)	1.7(2)
O(2)	0.2341(14)	0.2341(14)	0	1.7(2)

R_{wp} = 11.62, R_p = 7.21, for all 50 reflections

$\chi^2_{\text{red}} = 2.459$ for 17 variables (2 lattice parameters, 2 positional parameters, 3 displacement parameters, scale factor, zero point, 2 profile parameters, 6 background parameters)

Ba – O(1)	2.9986(10) x 4	Ba – O(2)	3.0031(5) x 8
Mo – O(1)	1.97(2) x 2	Mo – O(2)	1.983(12) x 4
Sm – O(1)	2.28(2) x 2	Sm – O(2)	2.253(12) x 4
O(1) – O(2)	2.798(12) x 4		

Table S1 Structural variables for Ba₂LnMoO₆ compounds modelled in space group Fm $\bar{3}$ m

	O _x	100Ba _{Uiso}	100Mo/Ln _{Uiso}	100O _{U11}	100O _{U22,U33}
Ba ₂ EuMoO ₆ ^a	0.2305(9)	1.07(4)	0.96 (9)	2.2(2)*	
Ba ₂ GdMoO ₆ ^a	0.2335(13)	0.19(7)	-0.0(1)	0.4(3) *	
Ba ₂ DyMoO ₆ ^a	0.2364(14)	0.16(7)	-0.14(8)	1.0(3) *	
Ba ₂ YMoO ₆ ^b	0.23455(9)	1.01(3)	0.65(5)	0.78(4)	1.76(3)
Ba ₂ ErMoO ₆ ^b	0.2359(2)	0.95(5)	0.68(4)	0.8(1)	1.71(7)
Ba ₂ YbMoO ₆ ^b	0.2373(2)	0.82(2)	0.69(5)	0.85(5)	1.50(2)

^a Refinement against X-ray diffraction data^b Refinement against neutron diffraction data

* Isotropic displacement parameter

Fig S1 The in-phase component of the AC molar magnetic susceptibility of $\text{Ba}_2\text{LnMoO}_6$

Magnetic order arising from structural distortion: the Structure and Magnetic Properties of $\text{Ba}_2\text{LnMoO}_6$

Edmund J. Cussen, Danny R. Lynham and Joseph Rogers*
The University of Nottingham