Electronic Supplementary Material (ESI) for Journal of Materials Chemistry C.

SUPPLEMENTARY INFORMATION

Insulating to Metallic behaviour in the Cation Ordered Perovskites $Ba_2Nd_{1-x}Fe_xMoO_6$

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Figure S5 The long-range ordered magnetic moment in $Ba_2Nd_{0.95}Fe_{0.05}MoO_6$ derived from fitting neutron diffraction data collected at various temperatures between 1.5 K and room temperature. The error bars indicate one standard deviation in the Rietveld refinement.

$Ba_2Nd_{0.9}Fe_{0.1}MoO_6$ Room Temperature Structure

8. Phase information from GSAS

```
_pd_phase_name
   "from C:/GSAS/MyWork/Strathclyde/PXRD_D500/BaNdFeMoO/FC305/FC305_03"
_cell_length_a                  5.99684(5)
_cell_length_b                  5.99684
_cell_length_c                  8.52234(11)
_cell_angle_alpha                 90.0
_cell_angle_beta                  90.0
_cell_angle_gamma                   90.0
_cell_volume                    306.482(7)
_symmetry_cell_setting            tetragonal
_symmetry_space_group_name_H-M      "I 4/m"

Ba
Ba1   0.0       0.5       0.25      1.0      Uiso  0.01185(18)  4
Mo
Mo1   0.0       0.0       0.5       1.0      Uiso  0.00554(15)  2
Nd
Nd1   0.0       0.0       0.0       0.9      Uiso  0.00554(15)  2
O-
O1    0.0       0.0       0.26920(18)  1.0      Uani  0.0185      4
O-
O2    0.24831(22)  0.29231(21)  0.0       1.0      Uani  0.01983     8
Fe
Fe1   0.0       0.0       0.0       0.1      Uiso  0.00554(15)  2

O1    0.0270(7)  0.0       0.0       0.0270(7)  0.0       0.0016(7)
O2    0.0134(8)  -0.0050(7)  0.0       0.0180(6)  0.0       0.0281(6)

loop_ _atom_type_symbol
    _atom_type_number_in_cell
            Ba  4.0
            Mo  2.0
            Nd  1.8
            O- 12.0
            Fe  0.2

# If you change Z, be sure to change all 3 of the following
_chemical_formula_sum             "Ba2 Fe0.10 Mo Nd0.90 O6"
_chemical_formula_weight           601.99
_cell_formula_units_Z             2
```

```
#==============================================================================

# 8. Phase information from GSAS

_pd_phase_name
   "from C:/GSAS/MyWork/Strathclyde/Neutron/D2B/FC313_Nd8Fe2/FC313_RT_"
_cell_length_a                  5.9897(4)
_cell_length_b                  5.9897
_cell_length_c                  8.4662(10)
_cell_angle_alpha                90.0
_cell_angle_beta                 90.0
_cell_angle_gamma                90.0
_cell_volume                    303.740(10)
_symmetry_cell_setting           tetragonal
_symmetry_space_group_name_H-M       "I 4/m"
# ATOMIC COORDINATES AND DISPLACEMENT PARAMETERS

loop_
    _atom_site_type_symbol
    _atom_site_label
    _atom_site_fract_x
    _atom_site_fract_y
    _atom_site_fract_z
    _atom_site_occupancy
    _atom_site_thermal_displace_type
    _atom_site_U_iso_or_equiv
    _atom_site_symmetry_multiplicity
Ba
Ba1    0.0        0.5        0.25       1.0       Uiso   0.01232(30)   4
Mo
Mo1    0.0        0.0        0.5        1.0       Uiso   0.00717(27)   2
Nd
Nd1    0.0        0.0        0.0        0.8       Uiso   0.00717(27)   2
O-
O1     0.0        0.0        0.2666(13)   1.0       Uiso   0.01978(30)   4
O-
O2     0.2483(7)    0.2921(8)    0.0        1.0       Uiso   0.01978(30)   8
Fe
Fe1    0.0        0.0        0.0        0.2       Uiso   0.00717(27)   2

    _atom_type_number_in_cell
            Ba  4.0
            Mo  2.0
            Nd  1.6
            O- 12.0
            Fe  0.4

# If you change Z, be sure to change all 3 of the following
_chemical_formula_sum              "Ba2 Fe0.20 Mo Nd0.80 O6"
_chemical_formula_weight            593.16
_cell_formula_units_Z              2
```

$Ba_2Nd_{0.75}Fe_{0.25}MoO_6$ Structure at Room Temperature

8. Phase information from GSAS

```
_pd_phase_name                  I4/m
_cell_length_a                  5.9808(4)
_cell_length_b                  5.9808
_cell_length_c                  8.4543(10)
_cell_angle_alpha               90.0
_cell_angle_beta                90.0
_cell_angle_gamma               90.0
_cell_volume                    302.410(10)
_symmetry_cell_setting          tetragonal
_symmetry_space_group_name_H-M      "I 4/m"

# ATOMIC COORDINATES AND DISPLACEMENT PARAMETERS

loop_
   _atom_site_type_symbol
   _atom_site_label
   _atom_site_fract_x
   _atom_site_fract_y
   _atom_site_fract_z
   _atom_site_occupancy
   _atom_site_thermal_displace_type
   _atom_site_U_iso_or_equiv
   _atom_site_symmetry_multiplicity
Ba
BA1   0.0       0.5       0.25      1.0      Uiso  0.01437(28)  4
Nd
ND1   0.0       0.0       0.0       0.75     Uiso  0.00728(20)  2
Fe
Fe1   0.0       0.0       0.0       0.25     Uiso  0.00728(20)  2
Mo
Mo2   0.0       0.0       0.5       1.0      Uiso  0.00728(20)  2
O
O(1)  0.0       0.0       0.2673(25)  1.0    Uiso  0.0246(23)   4
O
O(2)  0.2501(13)  0.2870(12)  0.0     1.0    Uiso  0.0184(9)    8

loop_ _atom_type_symbol
   _atom_type_number_in_cell
            Ba  4.0
            Nd  1.5
            Fe  0.5
            Mo  2.0
            O  12.0

# If you change Z, be sure to change all 3 of the following
_chemical_formula_sum           "Ba2 Fe0.25 Mo Nd0.75 O6"
_chemical_formula_weight        588.74
_cell_formula_units_Z           2
```

$Ba_2Nd_{0.75}Fe_{0.25}MoO_6$ Structure at 2 K

8. Phase information from GSAS

```
_pd_phase_name
   "from C:/GSAS/MyWork/Strathclyde/Neutron/D2B/FC308_Nd75Fe25/FC308_1"
_cell_length_a                  5.96349(21)
_cell_length_b                  5.96349
_cell_length_c                  8.4607(5)
_cell_angle_alpha                 90.0
_cell_angle_beta                  90.0
_cell_angle_gamma                   90.0
_cell_volume                    300.890(30)
_symmetry_cell_setting              tetragonal
_symmetry_space_group_name_H-M      "I 4/m"


# ATOMIC COORDINATES AND DISPLACEMENT PARAMETERS

loop_
    _atom_site_type_symbol
    _atom_site_label
    _atom_site_fract_x
    _atom_site_fract_y
    _atom_site_fract_z
    _atom_site_occupancy
    _atom_site_thermal_displace_type
    _atom_site_U_iso_or_equiv
    _atom_site_symmetry_multiplicity
Ba
Ba1   0.0       0.5       0.25      1.0      Uiso  0.0110(4)    4
Mo
Mo1    0.0       0.0       0.5       1.0      Uiso  0.00731(33)  2
Nd
Nd1    0.0       0.0       0.0       0.75     Uiso  0.00731(33)  2
O-
O1    0.0       0.0       0.2756(6)   1.0      Uani  0.01019     4
O-
O2    0.2425(4)   0.2892(5)   0.0       1.0      Uani  0.02476     8
Fe
Fe1    0.0       0.0       0.0       0.25     Uiso  0.00731(33)  2


O1    0.0101(16)  0.0       0.0       0.0101(16)  0.0       0.0103(16)
O2    0.0019(17) -0.0020(13)  0.0       0.0243(18)  0.0       0.0481(21)

loop_ _atom_type_symbol
    _atom_type_number_in_cell
            Ba  4.0
            Mo  2.0
            Nd  1.5
            O- 12.0
            Fe  0.5

# If you change Z, be sure to change all 3 of the following
_chemical_formula_sum               "Ba2 Fe0.25 Mo Nd0.75 O6"
_chemical_formula_weight            588.74
_cell_formula_units_Z
```

$Ba_2Nd_{0.10}Fe_{0.90}MoO_6$ Room Temperature Structure

8. Phase information from GSAS

```
_pd_phase_name
   "from C:/gsas/MyWork/Strathclyde/PXRD_D500/BaNdFeMoO/FC314/FC314_02"
_cell_length_a                  8.11566(4)
_cell_length_b                  8.11566
_cell_length_c                  8.11566
_cell_angle_alpha                 90.0
_cell_angle_beta                  90.0
_cell_angle_gamma                   90.0
_cell_volume                    534.530(9)
_symmetry_cell_setting            cubic
_symmetry_space_group_name_H-M      "F m -3 m"

# ATOMIC COORDINATES AND DISPLACEMENT PARAMETERS
loop_
    _atom_site_type_symbol
    _atom_site_label
    _atom_site_fract_x
    _atom_site_fract_y
    _atom_site_fract_z
    _atom_site_occupancy
    _atom_site_thermal_displace_type
    _atom_site_U_iso_or_equiv
    _atom_site_symmetry_multiplicity
Ba
BA1    0.25       0.25       0.25       1.0       Uiso   0.00558(21)   8
Nd
ND2    0.0        0.0        0.0        0.1       Uiso   0.00556(15)   4
Fe
FE3    0.0        0.0        0.0        0.9       Uiso   0.00556(15)   4
Mo
MO4    0.5        0.5        0.5        1.0       Uiso   0.00556(15)   4
O
O5     0.25885(13)  0.0        0.0        1.0       Uani   0.00939     24

loop_ _atom_site_aniso_label
    _atom_site_aniso_U_11
    _atom_site_aniso_U_12
    _atom_site_aniso_U_13
    _atom_site_aniso_U_22
    _atom_site_aniso_U_23
    _atom_site_aniso_U_33
O5     0.0078(4)   0.0        0.0        0.01018(24  0.0        0.01018(24

loop_ _atom_type_symbol
    _atom_type_number_in_cell
            Ba  8.0
            Nd  0.4
            Fe  3.6
            Mo  4.0
            O  24.0
# If you change Z, be sure to change all 3 of the following
_chemical_formula_sum             "Ba2 Fe0.90 Mo Nd0.10 O6"
_chemical_formula_weight           531.28
_cell_formula_units_Z             4
```

$Ba_2Nd_{0.05}Fe_{0.95}MoO_6$ Room Temperature Structure

8. Phase information from GSAS

```
_pd_phase_name
   "from C:/gsas/MyWork/Strathclyde/PXRD_D500/BaNdFeMoO/FC316/FC316_02"
_cell_length_a                  8.09519(4)
_cell_length_b                  8.09519
_cell_length_c                  8.09519
_cell_angle_alpha                 90.0
_cell_angle_beta                  90.0
_cell_angle_gamma                   90.0
_cell_volume                    530.496(8)
_symmetry_cell_setting              cubic
_symmetry_space_group_name_H-M      "F m -3 m"

# ATOMIC COORDINATES AND DISPLACEMENT PARAMETERS
loop_
    _atom_site_type_symbol
    _atom_site_label
    _atom_site_fract_x
    _atom_site_fract_y
    _atom_site_fract_z
    _atom_site_occupancy
    _atom_site_thermal_displace_type
    _atom_site_U_iso_or_equiv
    _atom_site_symmetry_multiplicity
Ba
BA1    0.25       0.25       0.25       1.0       Uiso   0.00322(22)   8
Nd
ND2    0.0        0.0        0.0        0.05      Uiso   0.00329(17)   4
Fe
FE3    0.0        0.0        0.0        0.95      Uiso   0.00329(17)   4
Mo
MO4    0.5        0.5        0.5        1.0       Uiso   0.00329(17)   4
O
O5     0.25786(13)  0.0        0.0        1.0       Uani   0.00656     24

loop_ _atom_site_aniso_label
    _atom_site_aniso_U_11
    _atom_site_aniso_U_12
    _atom_site_aniso_U_13
    _atom_site_aniso_U_22
    _atom_site_aniso_U_23
    _atom_site_aniso_U_33
O5     0.0047(4)  0.0        0.0        0.00750(25  0.0        0.00750(25

loop_ _atom_type_symbol
    _atom_type_number_in_cell
            Ba  8.0
            Nd  0.2
            Fe  3.8
            Mo  4.0
            O  24.0

# If you change Z, be sure to change all 3 of the following
_chemical_formula_sum               "Ba2 Fe0.95 Mo Nd0.05 O6"
_chemical_formula_weight             526.86
_cell_formula_units_Z               4
```

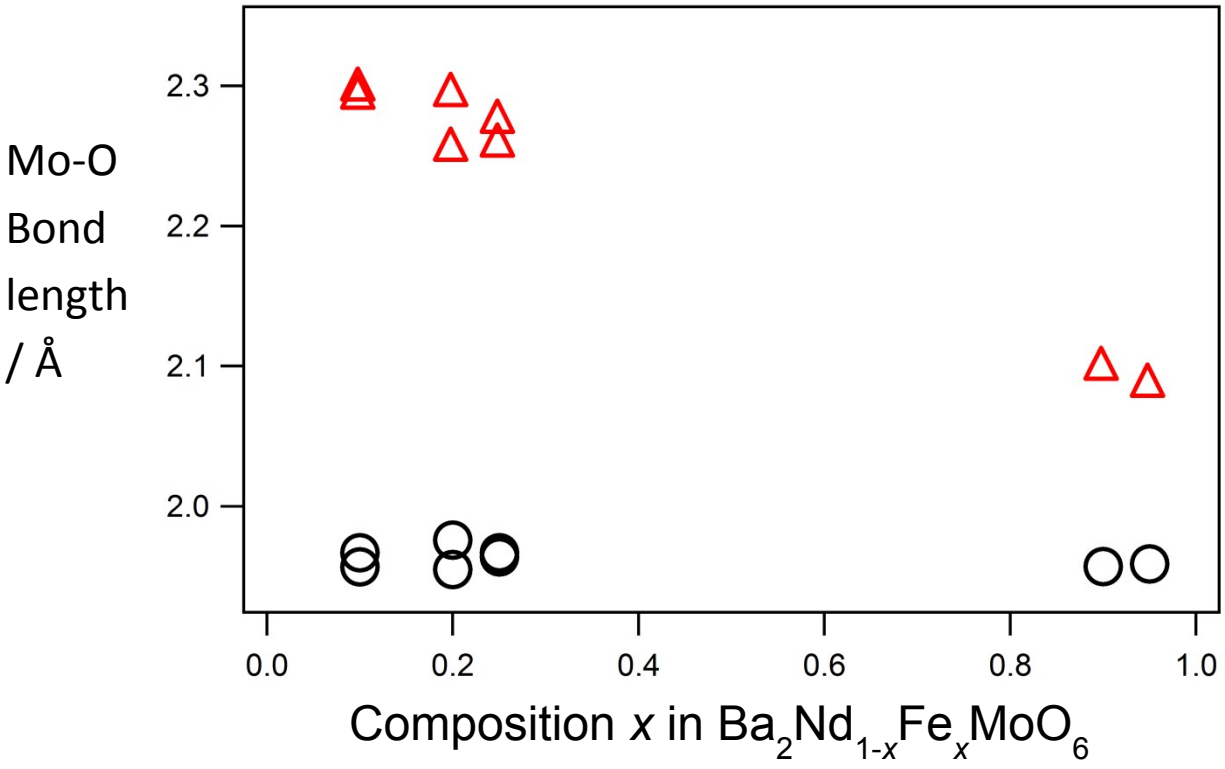

Figure S1
The bond lengths for Mo-O (black circles) and Nd/Fe-O (triangles) derived from refinement against neutron diffraction data collected at room temperature.

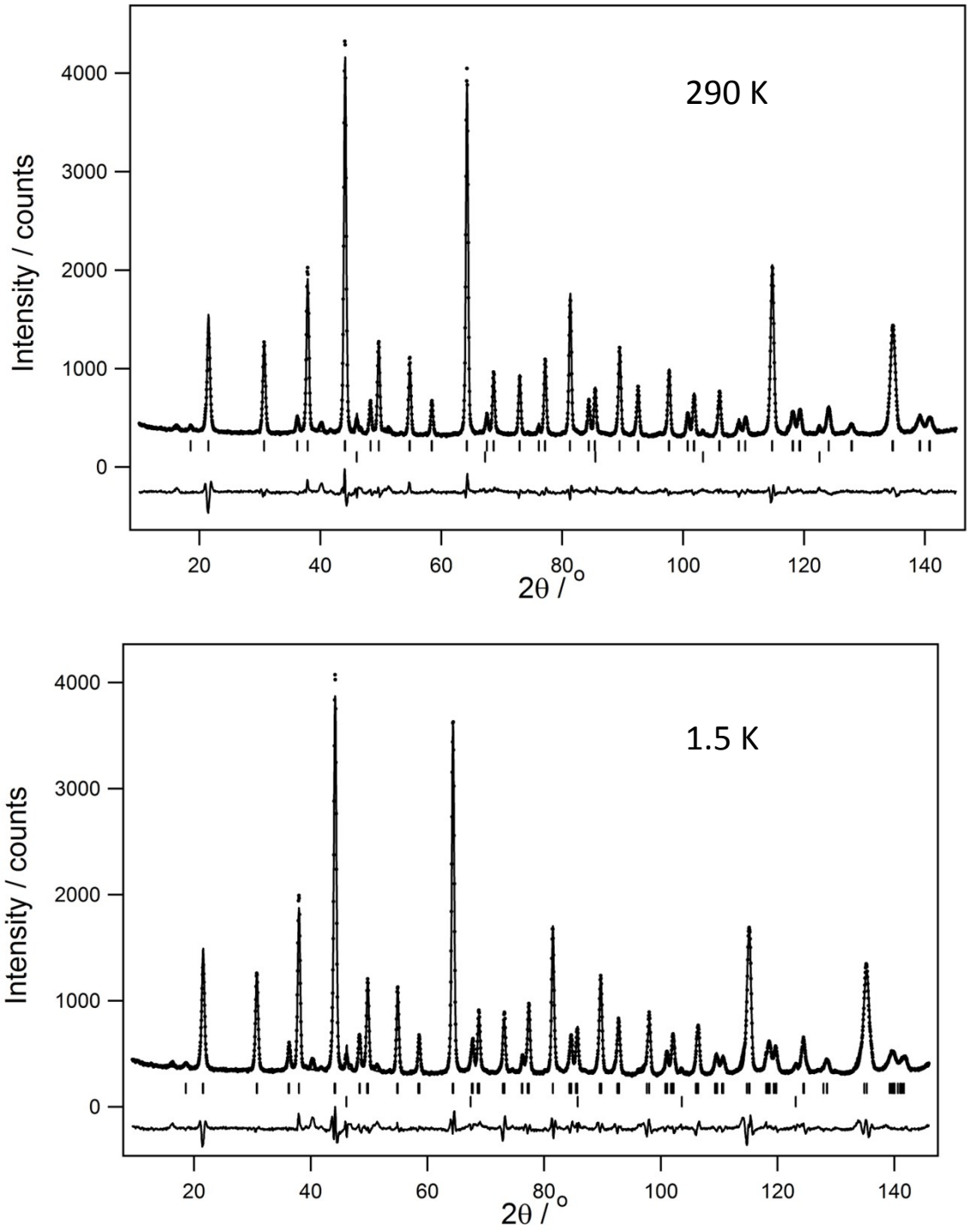

Figure S2 Neutron diffraction profiles collected at room temperature and 1.5 K from $Ba_2Nd_{0.75}Fe_{0.25}MoO_6$. Experimental and calculated profiles are shown as points and lines respectively. The vertical markers indicate the positions of allowed Bragg reflections from the tetragonal perovskite (upper marks) and the iron impurity (lower marks). The quality of fit is indicated by the parameters R_{wp} = 3.90, χ^2 = 6.94 for 290 K and R_{wp} = 5.14, χ^2 = 12.07 for the data collected at 1.5 K.

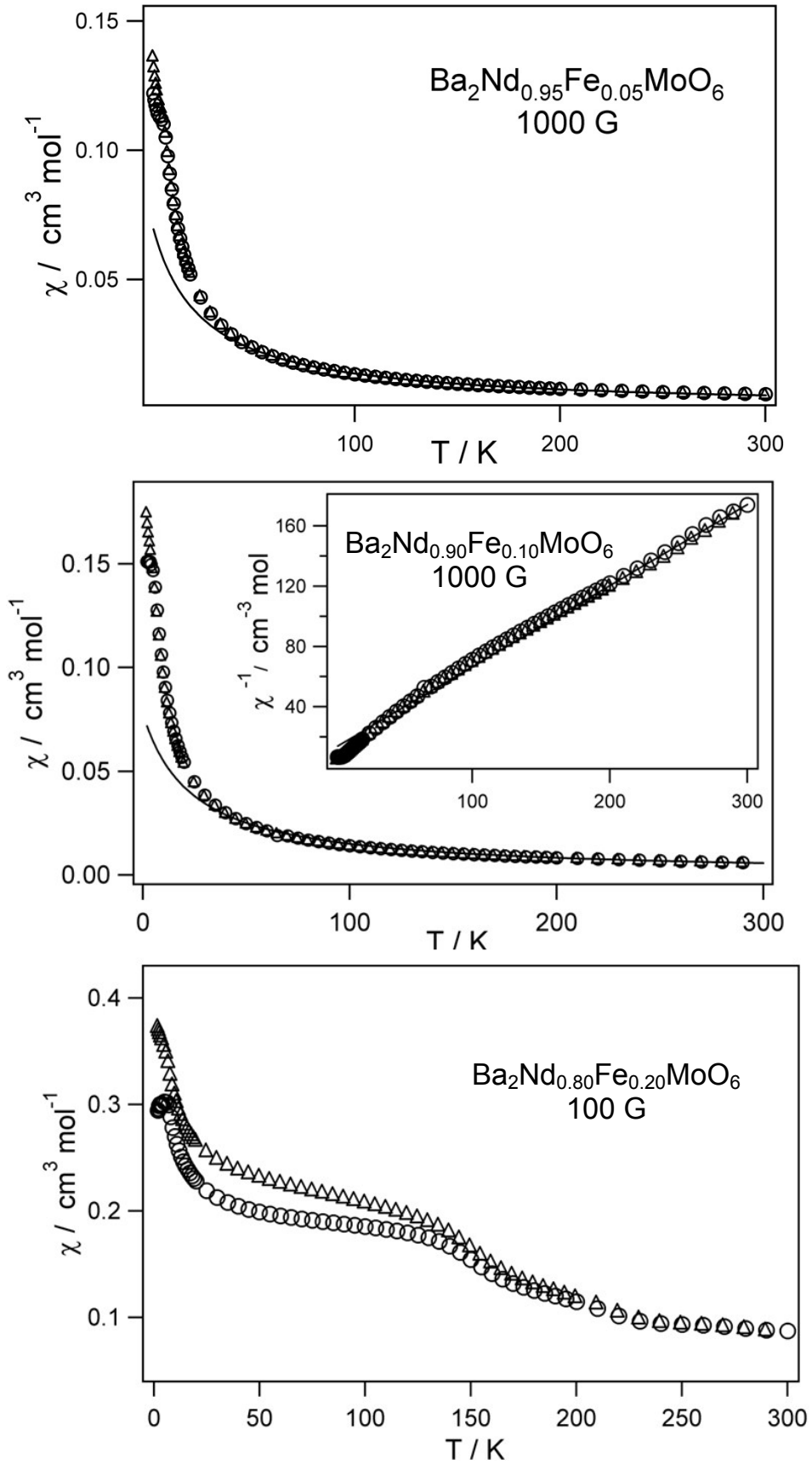

Figure S3 Magnetic susceptibility data collected from (top) $Ba_2Nd_{0.95}Fe_{0.05}MoO_6$ in 1000 G field, (middle) $Ba_2Nd_{0.90}Fe_{0.10}MoO_6$ in 1000 G field and (bottom) $Ba_2Nd_{0.80}Fe_{0.20}MoO_6$ in 100 G field. Data are were collected after cooling in either zero applied magnetic field (circles) or after cooling in the measuring field cooled (triangles). The inset shows the Curie Weiss fit to the inverse susceptibility.

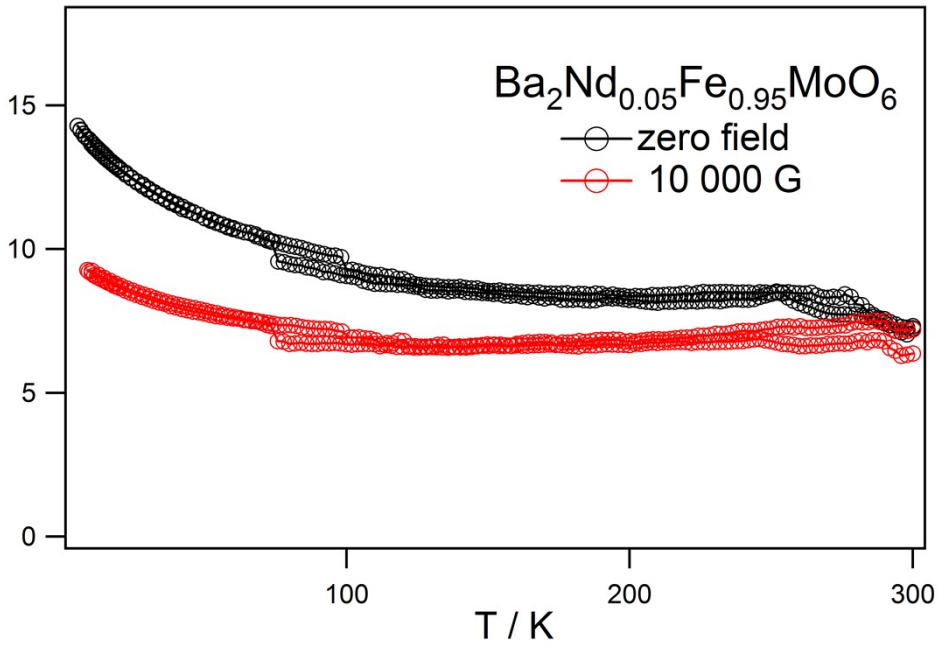

Figure S4 The temperature dependence of the resistivity of $Ba_2Nd_{0.05}Fe_{0.95}MoO_6$ as a function of applied magnetic field. Data collected in zero applied field and 10 000 G are represented as black and red circles respectively.

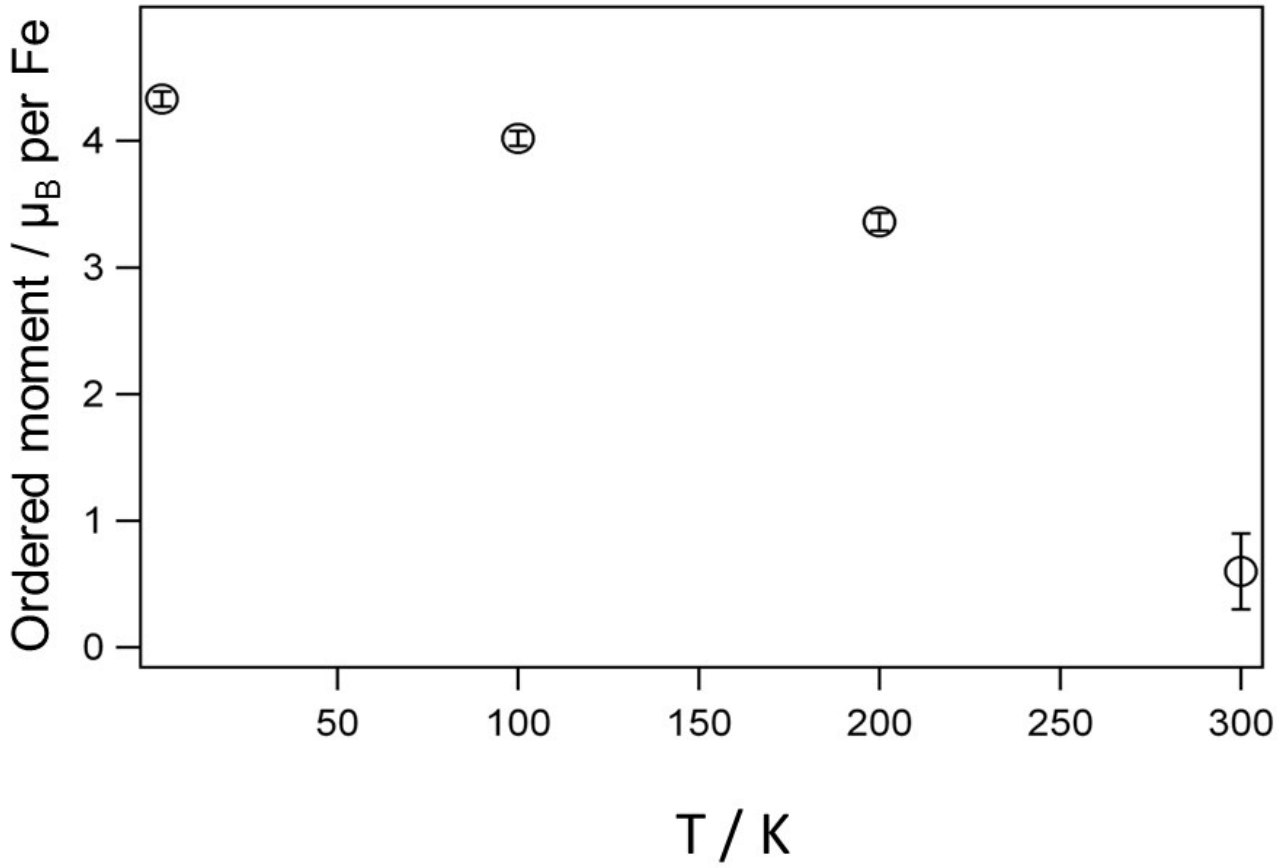

Figure S5 The long-range ordered magnetic moment in $Ba_2Nd_{0.95}Fe_{0.05}MoO_6$ derived from fitting neutron diffraction data collected at various temperatures between 1.5 K and room temperature. The error bars indicate one standard deviation in the Rietveld refinement.