Supporting Information

Magnetic Ground State Crossover in a Series of Glaserite Systems with Triangular Magnetic Lattices

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Table SI 1. Atomic positional parameters of $Na_2BaMn(VO_4)_2$ at 298 K and 125 K respectively.

Table SI 2. Atomic positional parameters of $Na_2BaCo(VO_4)_2$ at 298 K.

Table SI 3. Atomic positional parameters of $Na_2BaMn_{0.6}Co_{0.4}(VO_4)_2$ at 298 K and 150 K respectively.

Table SI 4. EDS analysis of $Na_2BaMn(VO_4)_2$, $Na_2BaCo(VO_4)_2$ and $Na_2BaMn_{0.6}Co_{0.4}(VO_4)_2$.

Figure SI 1. A comparison between (a) calculated PXRD pattern of $Na_2BaMn(VO_4)_2$ and (b) precursor that was used to synthesize $Na_2BaMn_{0.6}Co_{0.4}(VO_4)_2$ single crystal.

Figure SI 2. PXRD pattern of $Na_2BaMn_{0.6}Co_{0.4}(VO_4)_2$ at 20 K. The open circle is the best fit from the Rietveld refinement by using the program FULLPROF. The refinements were done based on the single crystal structure data obtain at 153 K for $Na_2BaMn_{0.6}Co_{0.4}(VO_4)_2$ with the space group of C2/c. The cell parameters are a = 9.6001(5) Å, b = 5.5619(8) Å, c = 14.1488(13) Å , β = 90.04(2)°, V = 753.20(4) Å^3, Rp = 3.31% and Rwp = 4.52%.

Figure SI 3. The diffuse magnetic scattering at T = 1.5 K, obtained by subtracting the paramagnetic contribution measured at 12 K, corrected for the Mn magnetic form factor decay with the momentum transfer Q. An estimate of the correlation length (ξ) was determined by fitting the Q dependence of the diffuse intensity to a Lorentzian function shown by solid red line.

Figure SI 4. Evolution of magnetic scattering at 1.5 K, 1K and 0.3 K, obtained by subtracting the paramagnetic contribution measured at 12 K. Due to the overlap of the diffuse magnetic scattering with magnetic Bragg peaks it is difficult to make a quantitative estimation of the change with the temperature of the diffuse scattering. However, a visual inspection of data suggests that the diffuse scattering does not get suppressed when the magnetic Bragg peaks develop.

Figure SI 5. The pure ferro-magnetic scattering observed for Na2BaCo(VO4)2 by subtracting the paramagnetic 5 K scattering from 0.3 K data.

Table SI 1. Atomic positional parameters of $Na_2BaMn(VO_4)_2$ at 298 K and 125 K respectively.

Atom	Wyck.	*x*	*y*	*z*
***P*-3*m*1 – 298 K**				
Na(1)	2*d*	-0.33333(0)	0.33333(0)	-0.3339(12)
Ba(1)	1*a*	0	0	0
Mn(1)	1*b*	0	0	0.50000(0)
V(1)	2*d*	-0.66667(0)	0.66667(0)	-0.2329(4)
O(1)	2*d*	-0.33333(0)	0.33333(0)	-0.0021(18)
O(2)	6*i*	0.341(2)	0.1706(11)	0.3150(13)
***C*2/*c* – 125 K**				
Na(1)	8*f*	0.3323(5)	0.4750(10)	0.0858(4)
Ba(1)	4*e*	0.50000	-0.0505(2)	0.25000
Mn(1)	4*b*	0.50000	0	0.50000
V(1)	8*f*	0.3317(2)	0.4776(4)	0.36703(15)
O(1)	8*f*	0.3229(10)	0.5224(19)	0.2503(6)
O(2)	8*f*	0.3846(10)	0.7393(19)	0.4179(7)
O(3)	8*f*	0.4503(10)	0.2565(18)	0.3946(7)
O(4)	8*f*	0.1709(10)	0.4006(18)	0.4101(7)

Table SI 2. Atomic positional parameters of $Na_2BaCo(VO_4)_2$ at 298 K.

Atom	Wyck.	*x*	*y*	*z*
***P*-3*m*1 – 298 K**				
Na(1)	2*d*	-0.33333(0)	0.33333(0)	-0.3339(12)
Ba(1)	1*a*	0	0	0
Mn(1)	1*b*	0	0	0.50000(0)
V(1)	2*d*	-0.66667(0)	0.66667(0)	-0.2329(4)
O(1)	2*d*	-0.33333(0)	0.33333(0)	-0.0021(18)
O(2)	6*i*	0.341(2)	0.1706(11)	0.3150(13)

Table SI 3. Atomic positional parameters of $Na_2BaMn_{0.6}Co_{0.4}(VO_4)_2$ at 298 K and 150 K respectively.

Atom	Wyck.	*x*	*y*	*z*
***P*-3*m*1 – 298 K**				
Na(1)	2*d*	-0.33333(0)	0.33333(0)	-0.3339(12)
Ba(1)	1*a*	0	0	0
Mn(1)	1*b*	0	0	0.50000(0)
V(1)	2*d*	-0.66667(0)	0.66667(0)	-0.2329(4)
O(1)	2*d*	-0.33333(0)	0.33333(0)	-0.0021(18)
O(2)	6*i*	0.341(2)	0.1706(11)	0.3150(13)
***C*2/*c* – 150 K**				
Na(1)	8*f*	0.3323(5)	0.4750(10)	0.0858(4)
Ba(1)	4*e*	0.50000	-0.0505(2)	0.25000
Mn(1)	4*b*	0.50000	0	0.50000
V(1)	8*f*	0.3317(2)	0.4776(4)	0.36703(15)
O(1)	8*f*	0.3229(10)	0.5224(19)	0.2503(6)
O(2)	8*f*	0.3846(10)	0.7393(19)	0.4179(7)
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Table SI 4. EDS analysis of $Na_2BaMn(VO_4)_2$, $Na_2BaCo(VO_4)_2$ and $Na_2BaMn_{0.6}Co_{0.4}(VO_4)_2$.

Element Type	$Na_2BaMn(VO_4)_2$	$Na_2BaCo(VO_4)_2$	$Na_2BaMn_{0.6}Co_{0.4}(VO_4)_2$
Na	14.01	13.98	14.32
Ba	7.19	7.18	7.21
Mn	6.09	--	4.32
Co	--	7.11	2.89
V	14.12	14.08	14.56
O	58.59	57.65	56.70

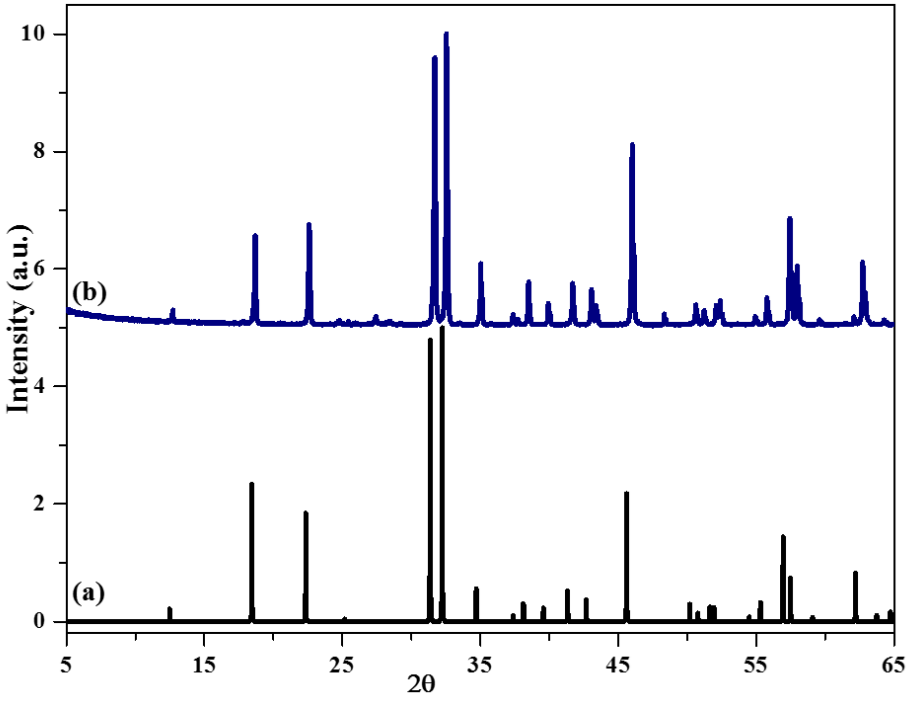


Figure SI 1. A comparison between (a) calculated PXRD pattern of $Na_2BaMn(VO_4)_2$ and (b) precursor that was used to synthesize $Na_2BaMn_{0.6}Co_{0.4}(VO_4)_2$ single crystal.

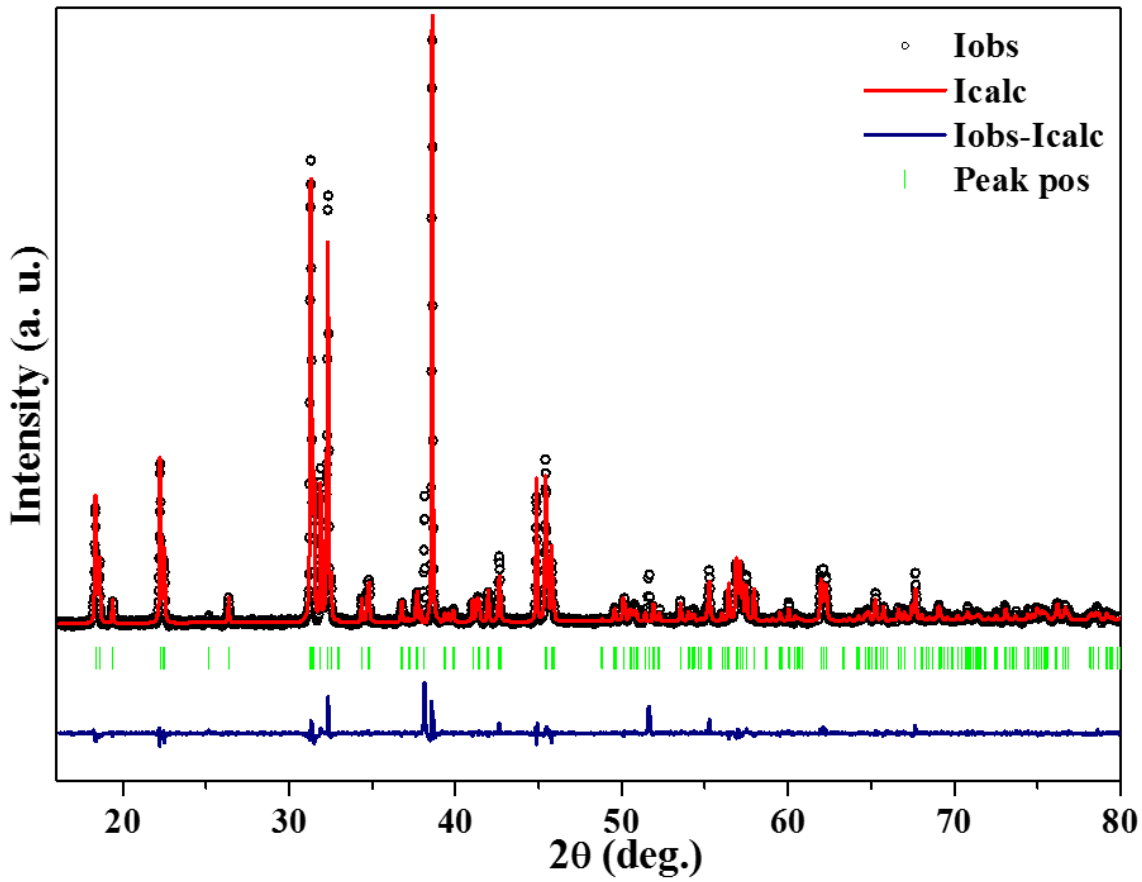
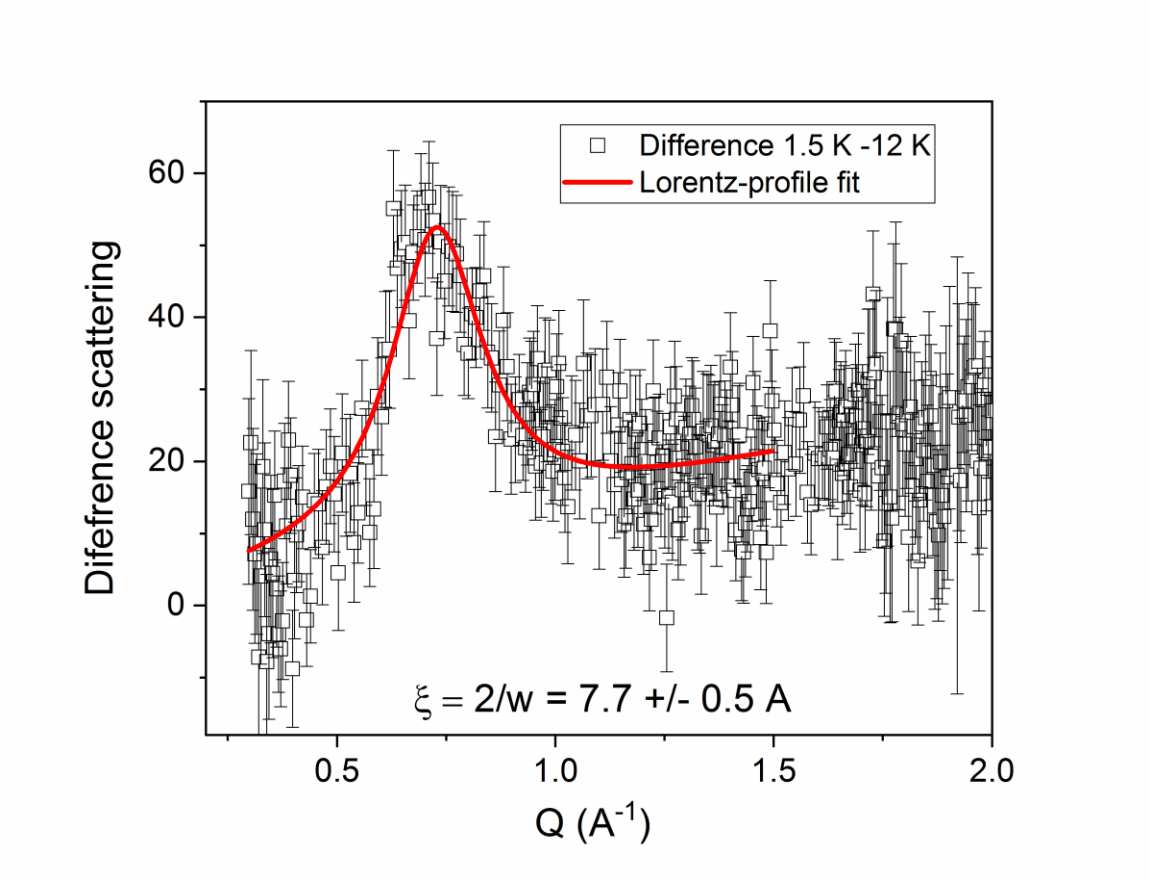


Figure SI 2. PXRD pattern of $Na_2BaMn_{0.6}Co_{0.4}(VO_4)_2$ at 20 K. The open circle is the best fit from the Rietveld refinement by using the program FULLPROF. The refinements were done based on the single crystal structure data obtain at 153 K for $Na_2BaMn_{0.6}Co_{0.4}(VO_4)_2$ with the space group of C2/c. The cell parameters are a = 9.6001(5) Å, b = 5.5619(8) Å, c = 14.1488(13) Å , β = 90.04(2)°, V = 753.20(4) Å^3, Rp = 3.31% and Rwp = 4.52%.



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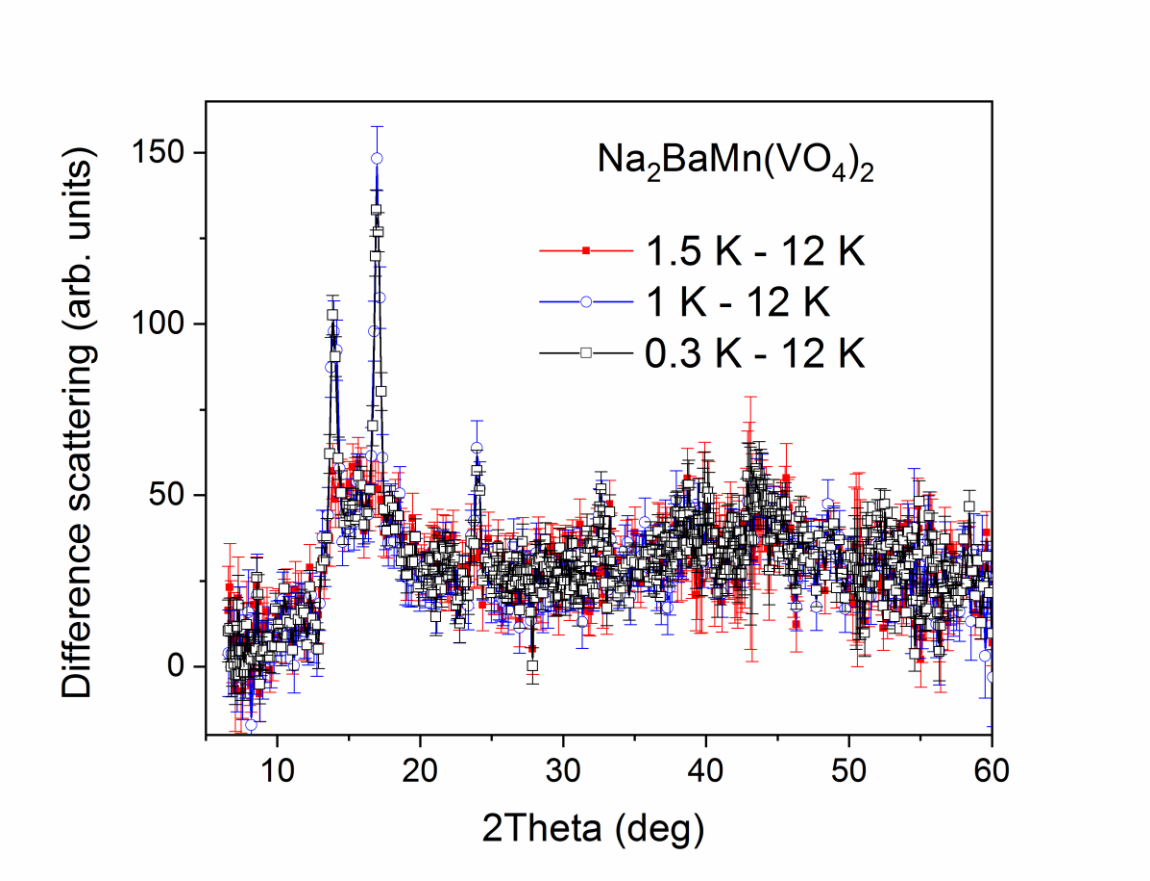

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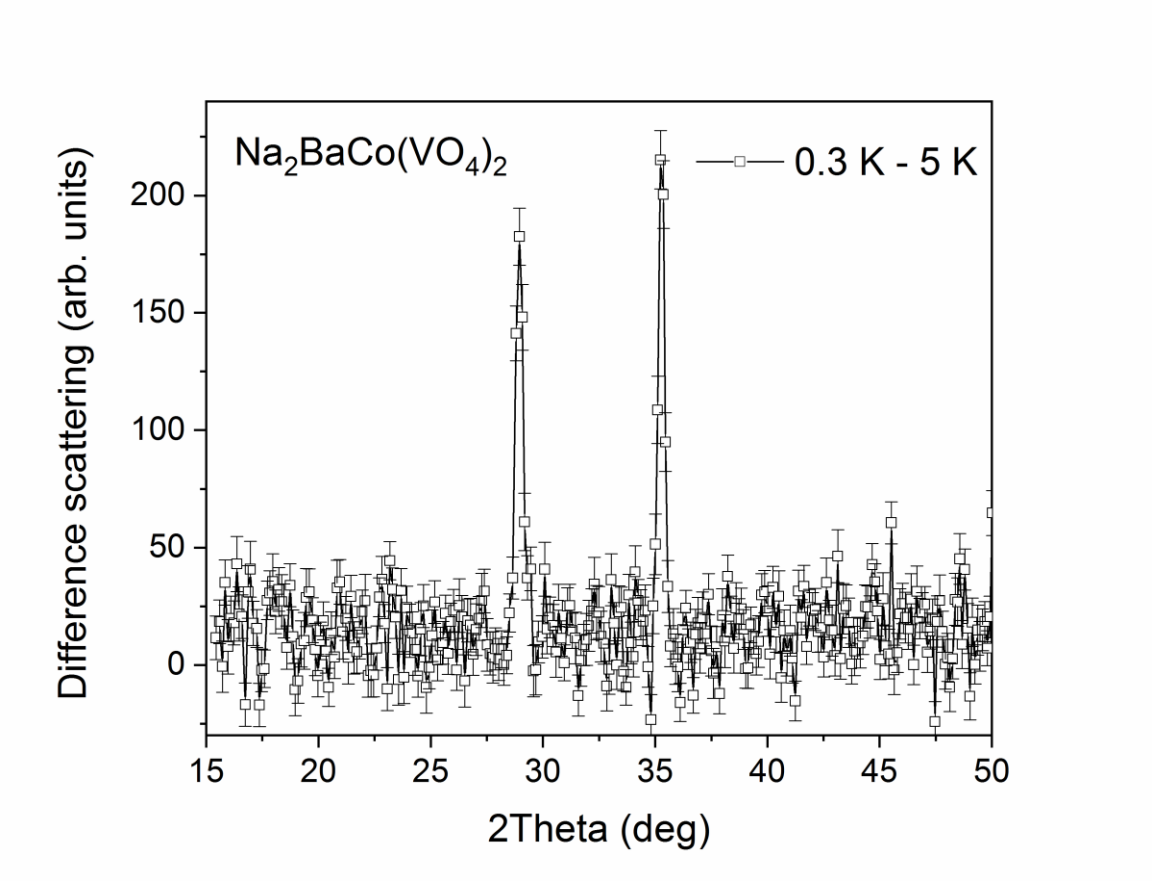

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