Supporting Information

$NaKV_4O_9{\cdot}2H_2O$: A New 2D Magnetic Compound with 1/5-depleted Square Lattice

Meiyan Cui[a,b], Zhangzhen He[a,*], Nannan Wang[a], Yingying Tang[a], Wenbin Guo[a], Suyun Zhang[a], Lin Wang[a], and Hongping Xiang[a]

[a]State Key Laboratory of Structural Chemistry, Fujian Institute of Research on the Structure of Matter, Chinese Academy of Sciences, Fuzhou, Fujian 350002, China.

[b]University of the Chinese Academy of Sciences, Beijing, 100039, P. R. China.

*E-mail: hezz@fjirsm.ac.cn

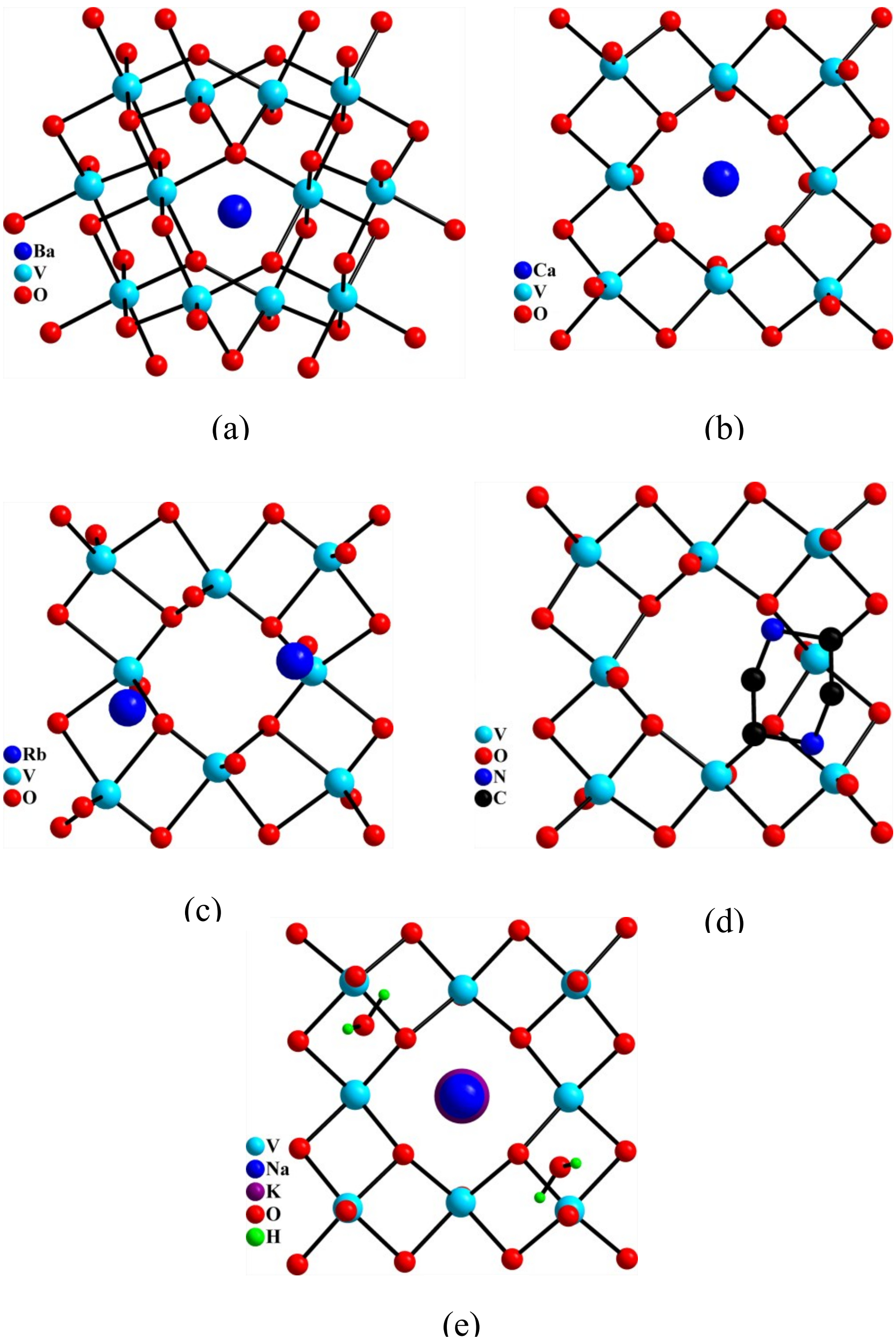

Figure S1. The positions of alkaline, alkaline-earth, or organic cations in the interlayer space of $MV_4O_9/A_2V_2O_9$ family with a unique spin-lattice for (a) BaV_4O_9, (b) Ca/SrV_4O_9, (c) $Rb_2/Cs_2V_4O_9$, (d) $[H_2N(CH_2)_4NH_2]V_4O_9$, and (e) $NaKV_4O_9{\cdot}2H_2O$.

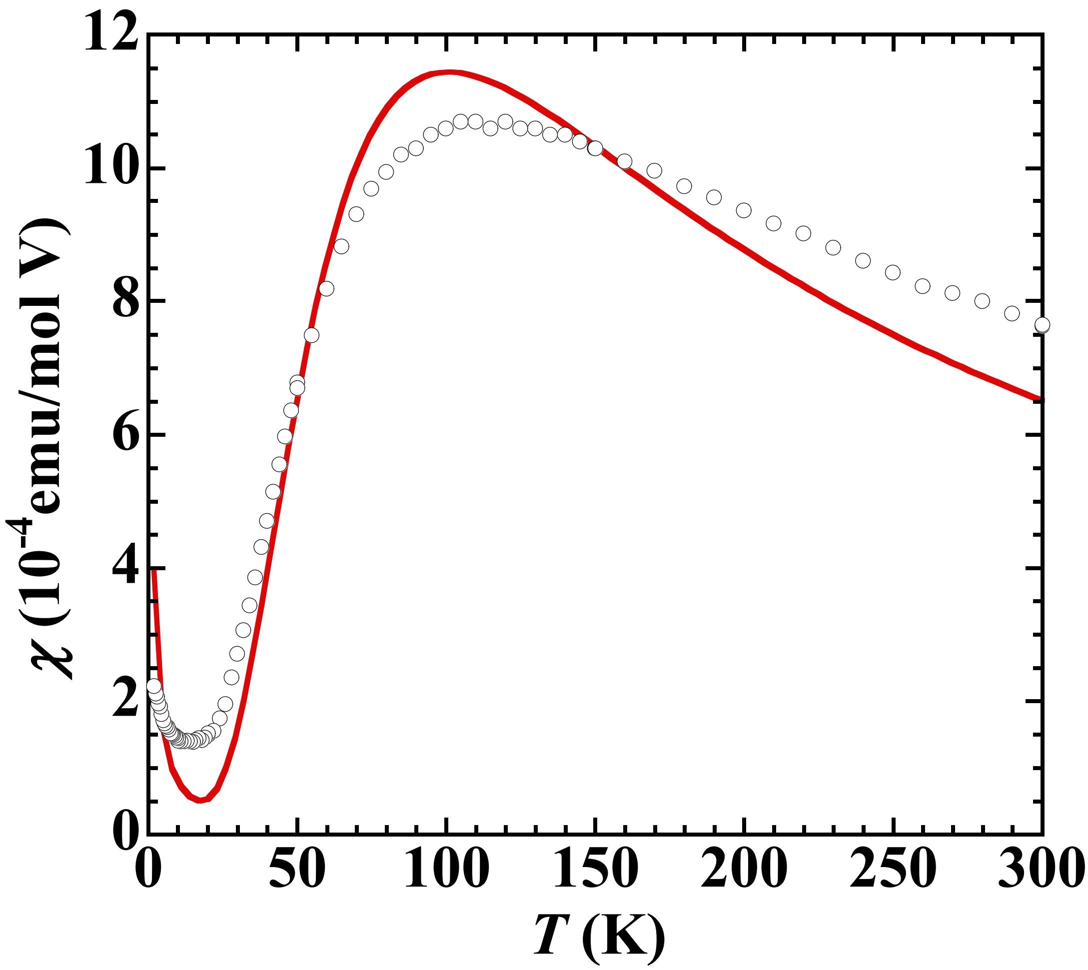

Figure S2. The susceptibility fitted using a Bleaney Bowers equation. The red line is a fit using a Bleaney Bowers equation expressed as $\chi_{raw} = Ng^2\mu_B{}^2/kT[3+\exp(\Delta/T)] + C'/(T-\theta') + \chi_{0,}$.

Table S1. Atomic coordinates (×10^4) and equivalent isotropic displacement parameters (Å^2×10^3) for $NaKV_4O_9 \cdot 2H_2O$. U(eq) is defined as one third of the trace of the orthogonalized Uij tensor.

atom	x	y	z	U(eq)
V(1)	2068(1)	2102(1)	8436(1)	10(1)
V(2)	973(1)	2954(1)	5384(1)	10(1)
Na	5000	6524(2)	2500	22(1)
K	0	4871(1)	2500	26(1)
O(1)	3947(3)	2515(2)	9608(3)	13(1)
O(2)	2913(3)	2541(2)	6450(3)	12(1)
O(3)	0	2584(3)	7500	13(1)
O(4)	2084(4)	1064(2)	8345(3)	20(1)
O(5)	960(4)	3992(2)	5230(3)	22(1)
H(6A)	3753	4939	3278	55(1)
H(6B)	3089	5405	4534	55(1)

Table S2. Anisotropic displacement parameters (Å^2×10^3) for $NaKV_4O_9 \cdot 2H_2O$. The anisotropic displacement factor exponent takes the form: $-2 \pi^2$ [h^2 a^{*2} U11 + ... + 2 h k a* b* U12].

atom	U11	U22	U33	U23	U13	U12
V(1)	8(1)	17(1)	5(1)	-1(1)	-1(1)	0(1)
V(2)	7(1)	17(1)	7(1)	1(1)	-1(1)	0(1)
Na	19(1)	28(1)	20(1)	0	3(1)	0
K	32(1)	28(1)	18(1)	0	1(1)	0
O(1)	8(1)	27(2)	3(1)	-3(1)	-2(1)	1(1)
O(2)	7(1)	21(2)	8(1)	0(1)	1(1)	1(1)
O(3)	8(2)	25(2)	5(2)	0	-1(1)	0
O(4)	28(2)	18(2)	15(1)	-2(1)	-2(1)	-2(1)
O(5)	26(2)	19(2)	20(1)	0(1)	1(1)	1(1)
O(6)	70(3)	35(2)	32(2)	-11(2)	19(2)	-20(2)

Table S3. Bond lengths [Å] and angles [deg] for $NaKV_4O_9 \cdot 2H_2O$.

V(1)-O(4)	1.609(3)	V(1)-O(2)	1.932(3)
V(1)-O(1)	1.937(3)	V(1)-O(1)#1	1.938(3)
V(1)-O(3)	2.016(2)	V(2)-O(5)	1.611(3)
V(2)-O(1)#5	1.931(3)	V(2)-O(2)	1.936(3)
V(2)-O(2)#4	1.953(3)	V(2)-O(3)	2.033(2)
Na-O(6)	2.327(4)	Na-O(6)#8	2.327(4)
Na-O(2)#3	2.412(3)	Na-O(2)#9	2.412(3)
Na-O(1)#9	2.471(3)	Na-O(1)#3	2.471(3)
K-O(5)#7	2.714(4)	K-O(5)#9	2.714(4)
K-O(5)#12	2.771(4)	K-O(5)	2.771(4)
K-O(4)#5	2.901(4)	K-O(4)#4	2.901(4)
K-O(6)	3.026(5)	K-O(6)#12	3.026(5)
O(4)-V(1)-O(2)	107.87(14)	O(4)-V(1)-O(1)	110.26(14)
O(2)-V(1)-O(1)	91.53(12)	O(4)-V(1)-O(1)#1	110.44(14)
O(2)-V(1)-O(1)#1	141.40(15)	O(1)-V(1)-O(1)#1	79.70(12)
O(4)-V(1)-O(3)	110.99(15)	O(2)-V(1)-O(3)	81.13(10)
O(1)-V(1)-O(3)	138.38(16)	O(1)#1-V(1)-O(3)	81.10(10)
O(5)-V(2)-O(1)#5	110.04(14)	O(5)-V(2)-O(2)	111.76(14)
O(1)#5-V(2)-O(2)	137.93(14)	O(5)-V(2)-O(2)#4	109.30(14)
O(1)#5-V(2)-O(2)#4	90.03(12)	O(2)-V(2)-O(2)#4	80.77(12)
O(5)-V(2)-O(3)	110.37(15)	O(1)#5-V(2)-O(3)	80.83(10)
O(2)-V(2)-O(3)	80.57(10)	O(2)#4-V(2)-O(3)	140.05(15)
O(6)-Na-O(6)#8	87.6(2)	O(6)-Na-O(2)#3	135.69(13)
O(6)#8-Na-O(2)#3	98.70(15)	O(6)-Na-O(2)#9	98.70(15)
O(6)#8-Na-O(2)#9	135.69(13)	O(2)#3-Na-O(2)#9	106.21(18)
O(6)-Na-O(1)#9	87.81(13)	O(6)#8-Na-O(1)#9	155.14(12)
O(2)#3-Na-O(1)#9	68.45(11)	O(2)#9-Na-O(1)#9	69.17(11)
O(6)-Na-O(1)#3	155.14(12)	O(6)#8-Na-O(1)#3	87.81(13)
O(2)#3-Na-O(1)#3	69.17(11)	O(2)#9-Na-O(1)#3	68.45(11)
O(1)#9-Na-O(1)#3	105.97(17)	O(5)#7-K-O(5)#9	99.11(15)
O(5)#7-K-O(5)#12	144.41(12)	O(5)#9-K-O(5)#12	79.86(12)
O(5)#7-K-O(5)	79.86(12)	O(5)#9-K-O(5)	144.41(12)
O(5)#12-K-O(5)	121.17(15)	O(5)#7-K-O(4)#5	84.54(10)
O(5)#9-K-O(4)#5	137.90(9)	O(5)#12-K-O(4)#5	73.97(10)
O(5)-K-O(4)#5	77.67(10)	O(5)#7-K-O(4)#4	137.90(9)
O(5)#9-K-O(4)#4	84.54(10)	O(5)#12-K-O(4)#4	77.67(10)
O(5)-K-O(4)#4	73.97(10)	O(4)#5-K-O(4)#4	120.19(14)
O(5)#7-K-O(6)	83.41(11)	O(5)#9-K-O(6)	74.84(11)
O(5)#12-K-O(6)	129.20(9)	O(5)-K-O(6)	69.68(10)
O(4)#5-K-O(6)	146.68(10)	O(4)#4-K-O(6)	56.88(11)
O(5)#7-K-O(6)#12	74.84(11)	O(5)#9-K-O(6)#12	83.41(11)

O(5)#12-K-O(6)#12	69.68(10)	O(5)-K-O(6)#12	129.20(9)
O(4)#5-K-O(6)#12	56.88(11)	O(4)#4-K-O(6)#12	146.68(10)
O(6)-K-O(6)#12	146.28(17)		

Symmetry transformations used to generate equivalent atoms:

#1 -x+1/2,-y+1/2,-z+2 #2 -x, y,-z+3/2
#3 -x+1,-y+1,-z+1 #4 -x+1/2,-y+1/2,-z+1
#5 x-1/2,-y+1/2, z-1/2 #6 x-1/2, y-1/2,z
#7 -x,-y+1,-z+1 #8 -x+1, y,-z+1/2
#9 x,-y+1, z-1/2 #10 x+1/2, y+1/2,z
#11 -x+1/2, y+1/2,-z+1/2 #12 -x, y,-z+1/2
#13 x+1/2,-y+1/2, z+1/2