Supporting Information

Experimental Realization of a Unique Class of Compounds: *XY*-Antiferromagnetic Triangular Lattices, $KAg_2Fe[VO_4]_2$ and $RbAg_2Fe[VO_4]_2$, with Ferroelectric Ground States

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S1. Crystallographic data and details of the refinement for $RbAg_2Fe[VO_4]_2$.

$P\bar{3}$, $Z = 1$	λ = 2.4062 Å, H = 0 T		λ = 1.542 Å (XRD)
T	**20K**	**1.5 K**	**293 K**
a	5.47563(3) Å	5.47556(6) Å	5.4827(1) Å
c	7.35696(8) Å	7.35685(9) Å	7.3826(1) Å
V	191.028(3) $Å^3$	191.022(3) $Å^3$	192.19(1) $Å^3$

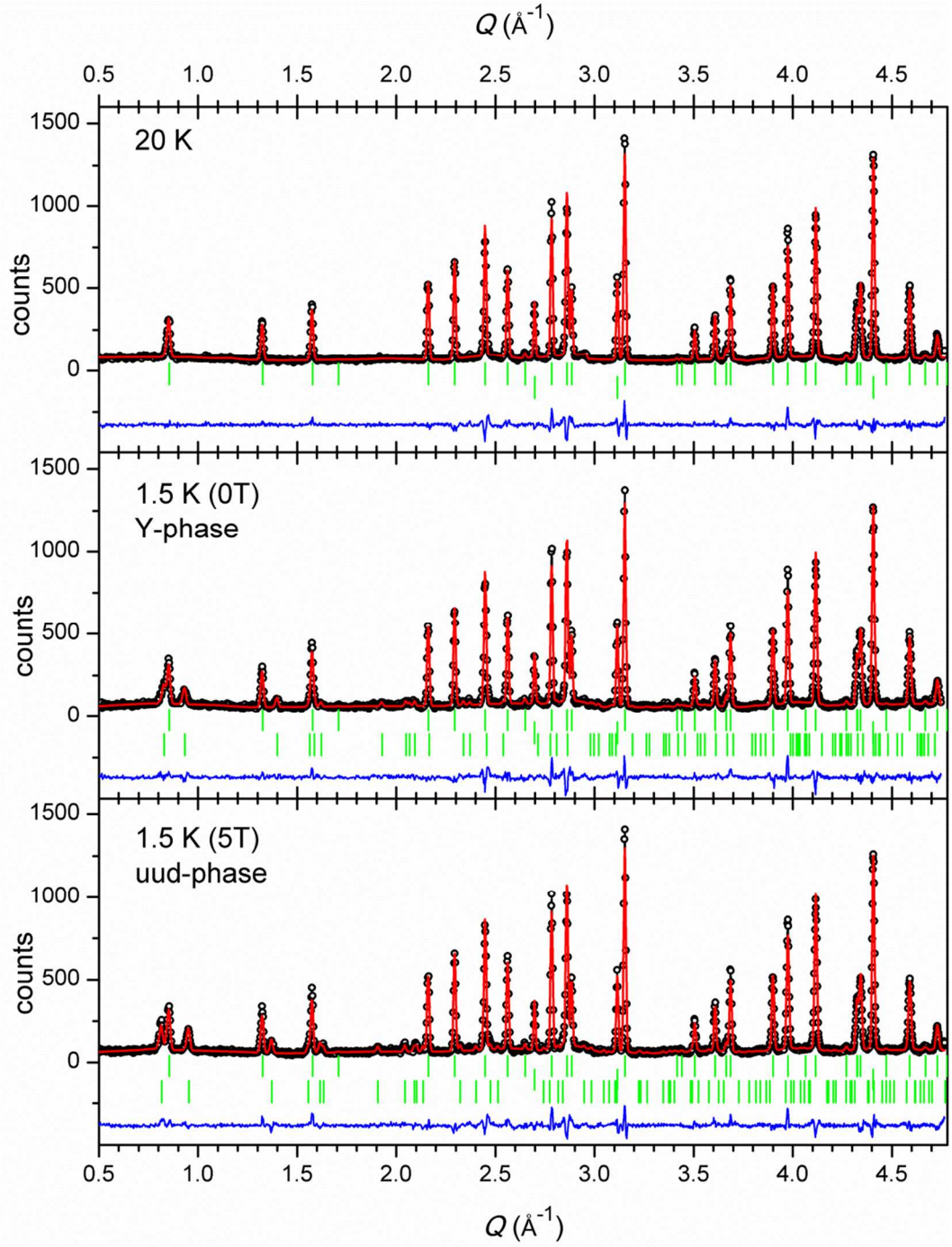
T	**20 K nuclear**	**1.5 K Y-phase, H = 0 T**	**1.5 K uud-phase, H = 5 T**
R_p	6.25	7.34	7.19
R_{wp}	7.98	9.37	8.97
R_{exp}	5.20	5.70	5.18
R_{Bragg}	3.41	4.33	4.49
χ^2	2.35	2.811	3.10
k_z		0.3724(3)	1/3
$R_{mag.}$		27.50	23.88
μ_B/Fe^{3+}		3.32	3.44

Fractional atomic coordinates and isotropic displacement parameters ($Å^2$)

T	**20 K**				**1.5 K**			
Atom (Wyckoff)	***x***	***y***	***z***	B_{iso}	***x***	***y***	***z***	B_{iso}
Ag (*2d*)	1/3	2/3	0.2818(7)	0.13(2)	1/3	2/3	0.2815(8)	0.10(3)
Rb (*1a*)	0	0	0	0.36(2)	0	0	0	0.33(3)
Fe (*1b*)	0	0	1/2	0.21(2)	0	0	1/2	0.19(3)
V (*2d*)	1/3	2/3	0.728(3)	0.12(2)	1/3	2/3	0.730(3)	0.09(3)
O1 (*6g*)	0.6674(7)	0.9120(5)	0.6590(5)	0.92(2)	0.6681(8)	0.9130(6)	0.6583(6)	0.90(3)
O2 (*2d*)	1/3	2/3	0.9580(6)	0.66(2)	1/3	2/3	0.9591(7)	0.64(3)

S2. $RbAg_2Fe[VO_4]_2$: Neutron diffraction data refinement (nuclear, 20 K) and magnetic phases (1.5 K) in magnetic fields, 0T (Y) and 5T (uud).

Experiment (black), refinement (red), difference (blue). Sequence of Bragg positions (green) from top to bottom: nuclear, Al-container, magnetic phase.



S3. Crystallographic data and details of the refinement for $KAg_2Fe[VO_4]_2$.

$P\bar{3}$, $Z = 1$	$\lambda = 2.4063$ Å, $H = 0$ T		$\lambda = 1.542$ Å (XRD)
T	**20K**	**1.5 K**	**293 K**
a	5.48099(7) Å	5.4811(1) Å	5.4896(1) Å
c	7.2119(1) Å	7.2119(1) Å	7.2430(1) Å
V	187.629(5) $Å^3$	187.631(5)$Å^3$	189.03(1) Å

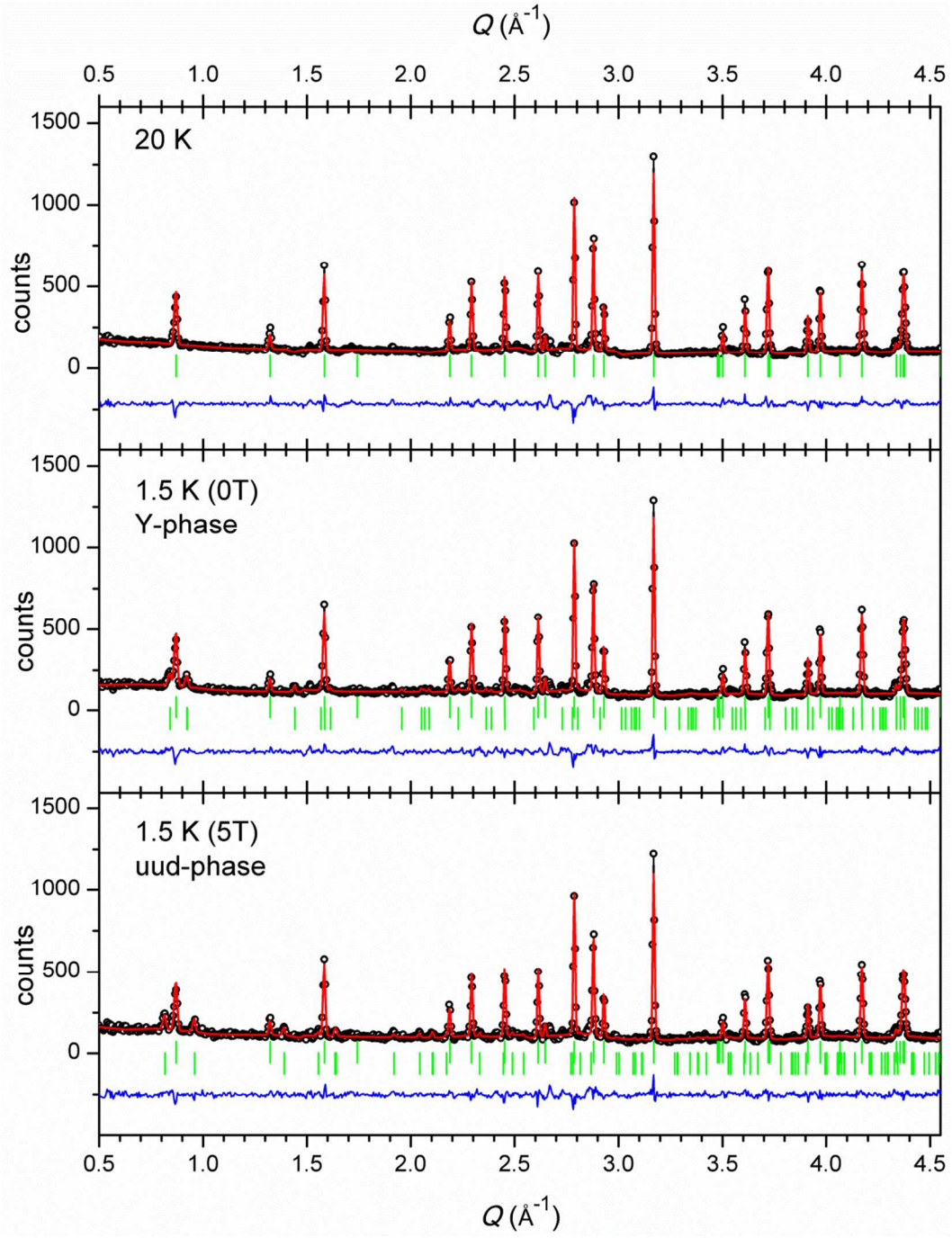
T	**20 K nuclear**	**1.5 K Y-phase, $H = 0$ T**	**1.5 K uud-phase, $H = 5$ T**
R_p	6.19	6.08	7.42
R_{wp}	8.15	7.96	9.32
R_{exp}	3.67	3.58	3.76
R_{Bragg}	7.67	7.25	5.75
χ^2	5.75	4.94	6.31
k_z		0.4035(6)	1/3
$R_{mag.}$		44.56	33.34
μ_B/Fe^{3+}		2.97	3.13

Fractional atomic coordinates and isotropic displacement parameters ($Å^2$)

T	**20 K**				**1.5 K**			
Atom (Wyckoff)	***x***	***y***	***z***	B_{iso}	***x***	***y***	***z***	B_{iso}
Ag (*2d*)	1/3	2/3	0.297(2)	0.14(5)	1/3	2/3	0.296(1)	0.24(5)
K (*1a*)	0	0	0	0.44(5)	0	0	0	0.54(5)
Fe (*1b*)	0	0	1/2	0.15(5)	0	0	1/2	0.25(5)
V (*2d*)	1/3	2/3	0.729(1)	0.17(5)	1/3	2/3	0.729(1)	0.27(5)
O1 (*6g*)	0.669(1)	0.901(1)	0.664(1)	0.71(5)	0.666(1)	0.901(1)	0.663(1)	0.82(5)
O2 (*2d*)	1/3	2/3	0.965(1)	0.50(5)	1/3	2/3	0.965(1)	0.60(5)

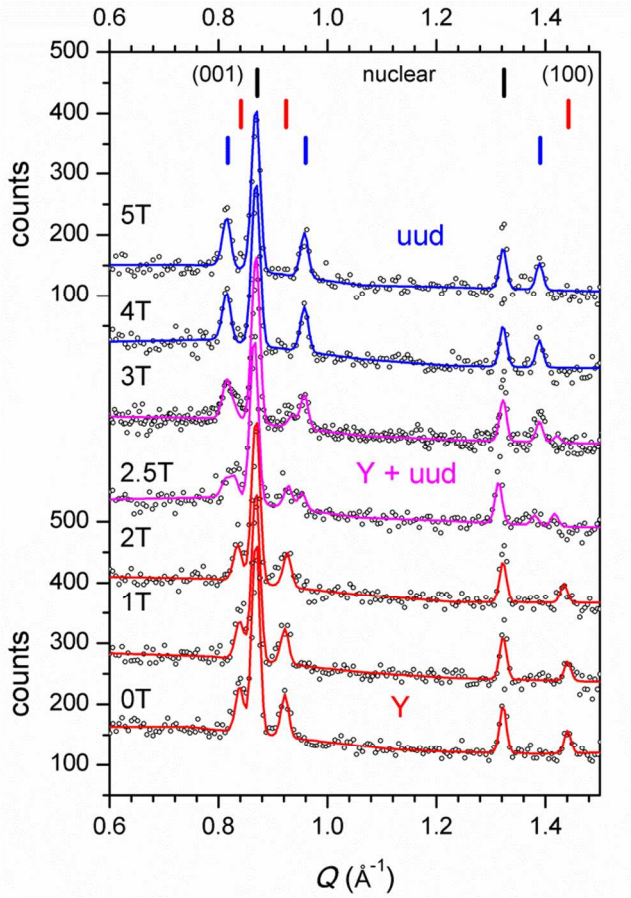
S4. $KAg_2Fe[VO_4]_2$: Neutron diffraction data refinement (nuclear, 20 K) and magnetic phases (1.5 K) in magnetic fields, 0T (Y) and 5T (uud).

Experiment (black), refinement (red), difference (blue). Sequence of Bragg positions (green) from top to bottom: nuclear, magnetic phase.



S4. Continued.

$KAg_2Fe[VO_4]_2$: Neutron diffraction data refinement at 1.5 K in applied fields.

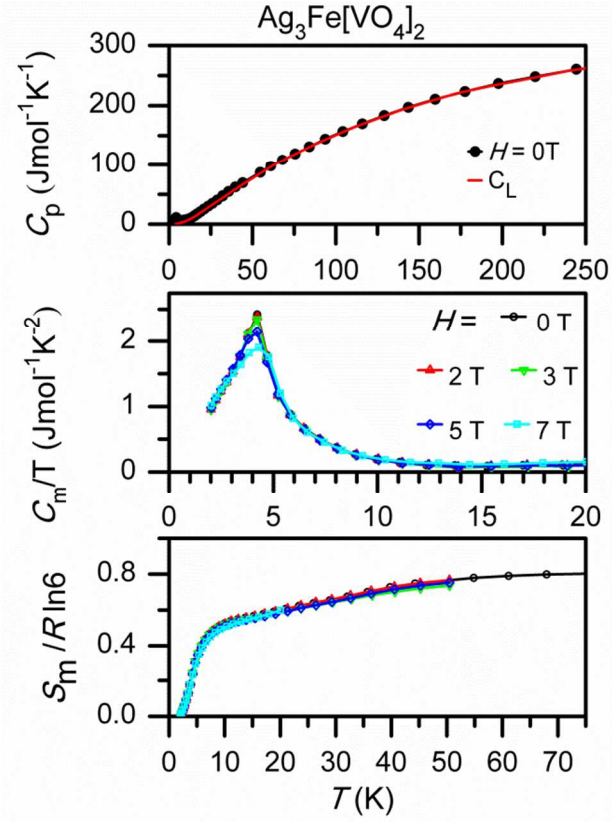


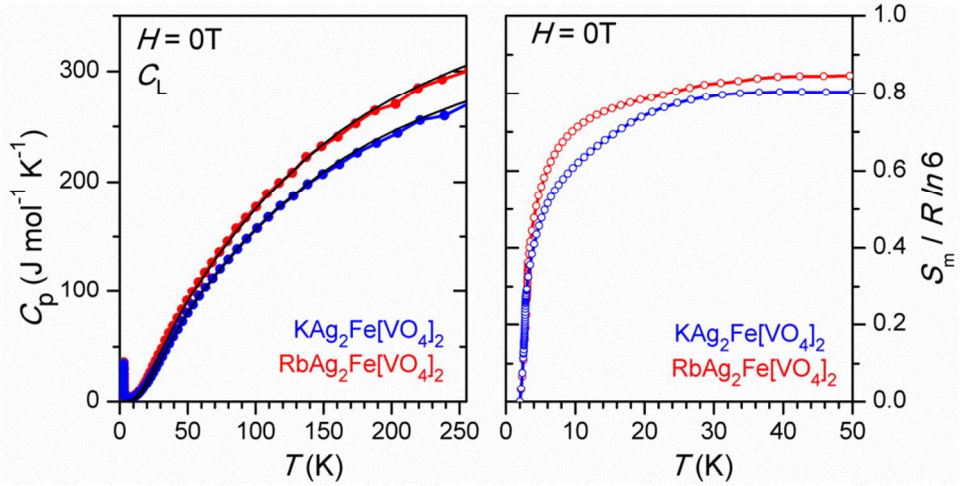
S5. Selected interatomic distances and angles for $AAg_2Fe[VO_4]_2$ with A = K or Rb

$P\ \bar{3}$, $Z = 1$	$T = 20K$, $H = 0$ T	
Distances in Å	**$KAg_2Fe[VO_4]_2$**	**$RbAg_2Fe[VO_4]_2$**
Fe – O1 (×6)	2.000(7)	2.010(3)
V – O1 (×3)	1.70(6)	1.718(8)
V – O2 (×1)	1.70(1)	1.69(2)
Ag – O1 (×3)	2.394(8)	2.350(4)
Ag – O2 (×1)	2.39(1)	2.382(7)
A – O1 (×6)	2.915(7)	2.994(3)
A – O2 (×6)	3.1745(6)	3.1766(4)
Angles in degrees		
O1 – Fe – O1	88.7(2)	89.5(1)
O1 – V – O1	112.6(3)	111.7(1)
O1 – V – O2	106.2(2)	107.2(1)
Φ_{S1}	121.6(3)	121.1(2)
Φ_{S2}	154.1(4)	149.6(2)

S6. Specific heat data for $AAg_2Fe[VO_4]_2$ (A = K, Rb, Ag).

The measured specific heat data was corrected for the lattice contribution using the diamagnetic references: $BaAg_2Mg[VO_4]_2$ ($P\bar{3}$) and $SrAg_2Mg[VO_4]_2$ ($C2/c$), respectively.





S.7 DSC measurements for $AAg_2Fe[VO_4]_2$ (A = K, Rb, Ag).