Supplementary Information

Magnetic structure and properties of centrosymmetric twisted-melilite $K_2CoP_2O_7$

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Table S1. Experimental and theoretical cell parameters and atomic coordinates. Thermal factors were included in the refinement in an overall isotropic form, B_{iso}, which converged to the value of 0.30(4) Å^2 for both 3 K and 20 K data sets.

	3K			20K			DFT		
a, Å	7.8855(2)			7.8849(3)			8.0043		
c, Å	11.2378(4)			11.2377(4)			11.4784		
Volume	698.78(4)			698.67(4)			735.43		
	x			y			z		
	NPD 3K	NPD 20K	DFT	NPD 3K	NPD 20K	DFT	NPD 3K	NPD 20K	DFT
K1	0.3140(5)	0.3135(6)	0.315241	0.3140(5)	0.3135(6)	0.315241	0	0	0
K2	0.8534(6)	0.8530(6)	0.84881	-0.8534(6)	-0.8530(6)	-0.84881	0	0	0
Co1	0.5	0.5	0.5	0	0	0	0.25	0.25	0.25
P1	0.1369(3)	0.1369(3)	0.136094	0.1369(3)	0.1369(3)	0.136094	0.2834(3)	0.2835(3)	0.282128
O1	0	0	0	0	0	0	0.3351(5)	0.3350(5)	0.332604
O2	0.1366(2)	0.1364(3)	0.135858	0.1366(2)	0.1364(3)	0.135859	0.1518(3)	0.1517(3)	0.150717
O3	0.3021(2)	0.3023(3)	0.30099150	0.0791(2)	0.0782(2)	0.07827450	0.3434(2)	0.3431(2)	0.34034750

Table S2. Interatomic distances and angles for each super-super exchange pathway for the low temperature experimentally measured structure and DFT relaxed structure of anti-melilite $K_2CoP_2O_7$ and melilite $Sr_2CoGe_2O_7$. J_1 corresponds to the intralayer exchange. J_{2a} and J_{2b} correspond to interlayer via inequivalent K1 and K2 sites, respectively.

	$K_2CoP_2O_7$ Experimental 3K			$K_2CoP_2O_7$ DFT Relaxed (U=4.5)			$Sr_2CoGe_2O_7$ Experimental 2.5K		
Pathway	J_1	J_{2a}	J_{2b}	J_1	J_{2a}	J_{2b}	J_1	J_{2a}	J_{2b}
Multiplicity (per unit cell)	4	4	4	4	4	4	4	4	4
Multiplicity (per J pathway)	1	2	2	1	2	2	1	2	2
Atom1	Co	Co	Co	Co	Co	Co	Co	Co	Co
Atom2	O	O	O	O	O	O	O	O	O
Atom3	P	K1	K2	P	K1	K2	Ge	Sr	Sr
Atom4	O	O	O	O	O	O	O	O	O
Atom5	Co	Co	Co	Co	Co	Co	Co	Co	Co
Interatomic Distances									
1-2	1.981	1.981	1.981	2.001	2.001	2.001	1.955	1.955	1.955
2-3	1.536	2.882	2.818	1.550	2.942	2.863	1.754	2.857	2.559
3-4	1.536	2.882	2.818	1.550	2.942	2.863	1.754	2.559	2.857
4-5	1.981	1.981	1.981	2.001	2.001	2.001	1.955	1.955	1.955
1-3	3.083	4.022	4.123	3.132	4.097	4.183	3.153	4.066	4.017
2-4	2.487	3.520	3.520	2.521	3.665	3.665	2.760	3.833	3.833
3-5	3.083	4.022	4.123	3.132	4.097	4.183	3.153	4.017	4.066
Angles									
1-2-3	121.926	110.202	117.351	123.216	110.512	117.524	116.342	113.985	125.198
2-3-4	108.096	75.274	77.302	108.819	77.049	79.612	103.775	89.908	89.908
3-4-5	121.926	110.202	117.351	123.216	110.512	117.524	116.342	125.198	113.985
1-3-5	129.465	88.622	85.920	129.288	88.909	86.640	132.467	82.133	82.133

Table S3. Unit cell parameters of DFT relaxed structures of $K_2CoP_2O_7$ with U of 4.0, 4.2 and 4.5eV

Structure type	U (eV)	a (Å)	b (Å)	c (Å)	α	β	γ	Volume (Å)	Total Energy (K/FU)	Energy difference (K/FU)
Experimental twisted-melilite-type structure	4.0	8.0016	8.0018	11.4802	90	90	90	735.055	-907189.1	0
	4.3	8.0032	8.0034	11.4791	90	90	90	735.279	-906494.6	0
	4.5	8.0043	8.0044	11.4784	90	90	90	735.426	-906043.6	0
Hypothetical melilite-type structure	4.0	8.0585	8.0587	11.4081	90	90	90	740.852	-906801.9	387.2
	4.3	8.0603	8.0604	11.4064	90	90	90	741.064	-906108.7	386.0
	4.5	8.0614	8.0615	11.4055	90	90	90	741.210	-905659.6	384.0

Table S4. Results of ionic relaxations of different configurations of a hypothetical $K_2CoP_2O_7$ with a melilite structure ($P\bar{4}2_1m$) – relaxed unit cell parameters and relative final energies for U = 4.5 eV.

Magnetic configuration	G-type	C-type
A (Å)	8.0614	8.06141
B	8.06152	8.06153
C	11.40546	11.40546
Volume Å^3	741.2091	741.21
Energy (K/FU)	0.0134	0

Table S5. Final relaxed positions of modified $K_2CoP_2O_7$ with melilite structure relaxed in C- and G-type magnetic configurations.

	C-type			G-type		
Atom	x	y	z	x	y	z
Co1	0.5	0	0.250007	0.5	0	0.250007
Co2	0	0.5	0.249993	0	0.5	0.249993
Co3	0.5	0	0.750007	0.5	0	0.750007
Co4	0	0.5	0.749993	0	0.5	0.749993
K1	0.664985	0.664989	0.005494	0.664986	0.66499	0.005494
K2	0.664985	0.664989	0.505494	0.664986	0.664989	0.505494
K3	0.835015	0.164989	0.994506	0.835014	0.164989	0.994506
K4	0.835015	0.164989	0.494506	0.835014	0.164989	0.494506
K5	0.164985	0.835011	0.994506	0.164986	0.835011	0.994506
K6	0.164985	0.835011	0.494506	0.164986	0.835011	0.494506
K7	0.335015	0.335011	0.005494	0.335014	0.335011	0.005494
K8	0.335015	0.335011	0.505494	0.335014	0.335011	0.505494
O1	0.5	0.5	0.321608	0.5	0.5	0.321608
O2	0.5	0.5	0.821608	0.5	0.5	0.821608
O3	0.577491	0.202275	0.33475	0.577491	0.202275	0.33475
O4	0.577491	0.202275	0.83475	0.577491	0.202275	0.83475
O5	0.638894	0.361118	0.141649	0.638893	0.361119	0.141649
O6	0.638894	0.361118	0.641649	0.638893	0.361119	0.641649
O7	0.702269	0.922514	0.165246	0.702269	0.922514	0.165246
O8	0.702269	0.922515	0.665246	0.702269	0.922514	0.665246
O9	0.797731	0.422515	0.334754	0.797731	0.422514	0.334754
O10	0.797731	0.422515	0.834755	0.797731	0.422515	0.834754
O11	0.861106	0.861118	0.358351	0.861107	0.861119	0.358351
O12	0.861106	0.861118	0.858351	0.861107	0.861119	0.858351
O13	0.922509	0.702274	0.16525	0.922509	0.702275	0.16525
O14	0.922509	0.702274	0.66525	0.922509	0.702275	0.66525
O15	0	0	0.178392	0	0	0.178392
O16	0	0	0.678392	0	0	0.678392
O17	0.077491	0.297725	0.16525	0.077491	0.297725	0.16525
O18	0.077491	0.297725	0.66525	0.077491	0.297725	0.66525
O19	0.138894	0.138882	0.358351	0.138893	0.138881	0.358351
O20	0.138894	0.138882	0.858351	0.138893	0.138881	0.858351
O21	0.202269	0.577485	0.334754	0.202269	0.577485	0.334754
O22	0.202269	0.577486	0.834754	0.202269	0.577485	0.834754
O23	0.297731	0.077485	0.165245	0.297731	0.077485	0.165246
O24	0.297731	0.077486	0.665246	0.297731	0.077485	0.665246
O25	0.361106	0.638882	0.141649	0.361107	0.638881	0.141649
O26	0.361106	0.638882	0.641649	0.361107	0.638881	0.641649
O27	0.422509	0.797726	0.334751	0.422509	0.797725	0.33475
O28	0.422509	0.797726	0.83475	0.422509	0.797725	0.83475
P1	0.635942	0.364064	0.273851	0.635942	0.364065	0.27385
P2	0.635942	0.364064	0.773851	0.635942	0.364065	0.77385
P3	0.864058	0.864064	0.226149	0.864058	0.864065	0.22615

P4	0.864058	0.864064	0.726149	0.864058	0.864065	0.72615
P5	0.135942	0.135936	0.226149	0.135942	0.135935	0.22615
P6	0.135942	0.135936	0.726149	0.135942	0.135935	0.72615
P7	0.364058	0.635936	0.273851	0.364058	0.635935	0.27385
P8	0.364058	0.635936	0.773851	0.364058	0.635935	0.77385

Table S6. Non-null components of the basis vectors (BVs) for the 4d(1/2,0,1/4) site of the $P4_2/mnm$ (#136) space group and the propagation vector k=(0,0,0). The irreducible representation providing the best agreement with experimental NPD data is highlighted in bold font.

		Co1(1/2,0,1/4)			Co2(0,1/2/1/4)			Co3(0,1/2,-1/4)			Co4(1/2,0,3/4)		
IR	BV	x	y	z	x	y	z	x	y	z	x	y	z
Г1	1			1			-1			-1			1
Г4	1			1			-1			1			-1
Г6	1			1			1			-1			-1
Г7	1			1			1			1			1
Г8	**1**	**1**			**-1**			**1**			**-1**		
	2		**1**			**1**			**-1**			**-1**	
	3		**1**			**-1**			**1**			**-1**	
	4	**-1**			**-1**			**1**			**1**		
Г9	1	1			-1			-1			1		
	2		1			1			1			1	
	3		-1			1			1			-1	
	4	1			1			1			1		

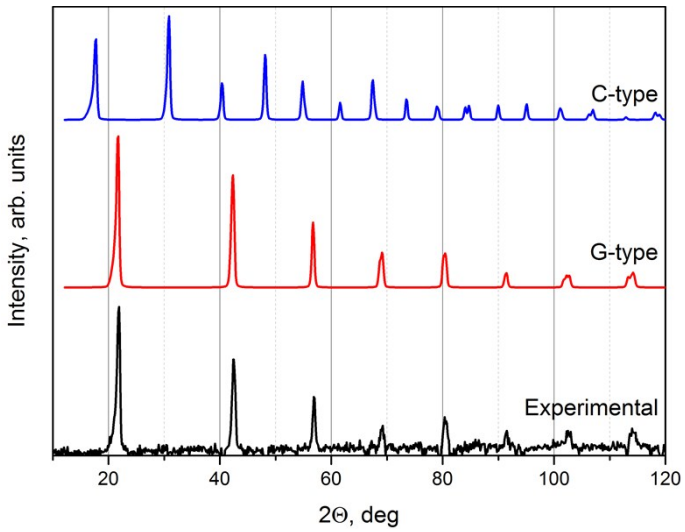

Figure S1. Bottom: purely magnetic diffraction signal (difference between NPD data collected at 20 K and 3 K); middle and top: simulated magnetic diffraction patterns for G-type and C-type models, respectively.

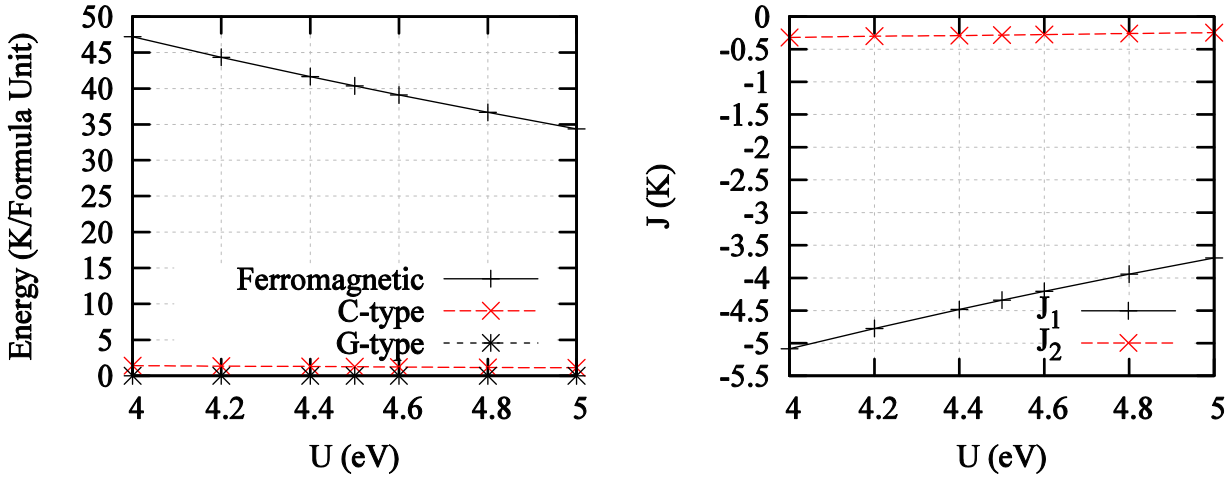

Figure S2. Total energy of considered magnetic configurations (left) and exchange parameters J_1 and J_2 (right) as a function of U.

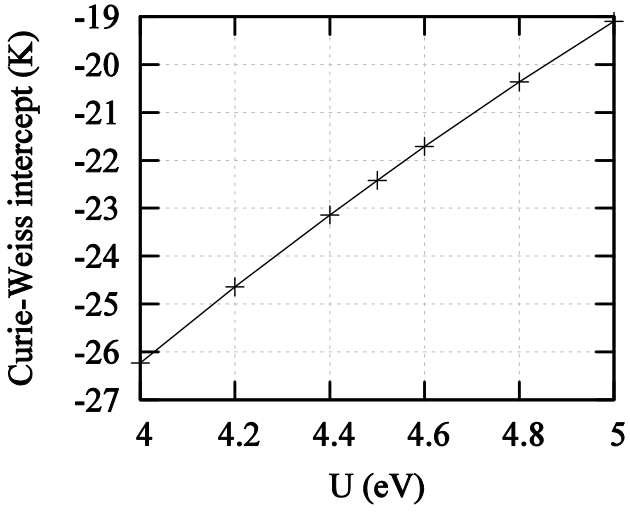

Figure S3. Calculated Curie-Weiss temperature as a function of U value.