

Supporting Information

Unconventional magnetism in ThCr₂Si₂-type phosphides,

La_{1-x}Nd_xCo₂P₂

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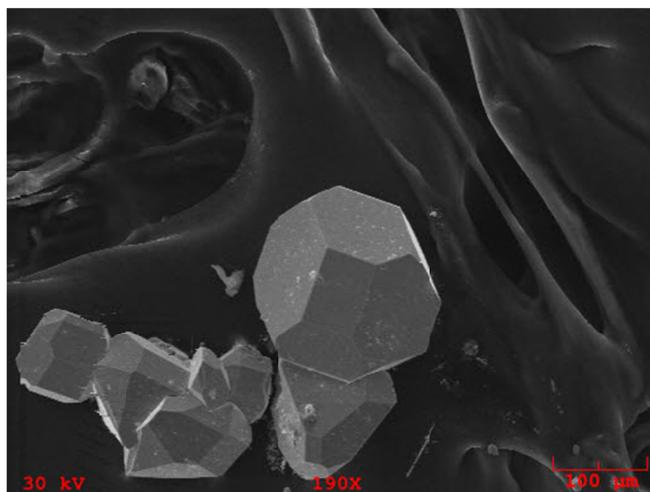


Fig. S1 Typical SEM image of a $\text{La}_{1-x}\text{Nd}_x\text{Co}_2\text{P}_2$ crystal.

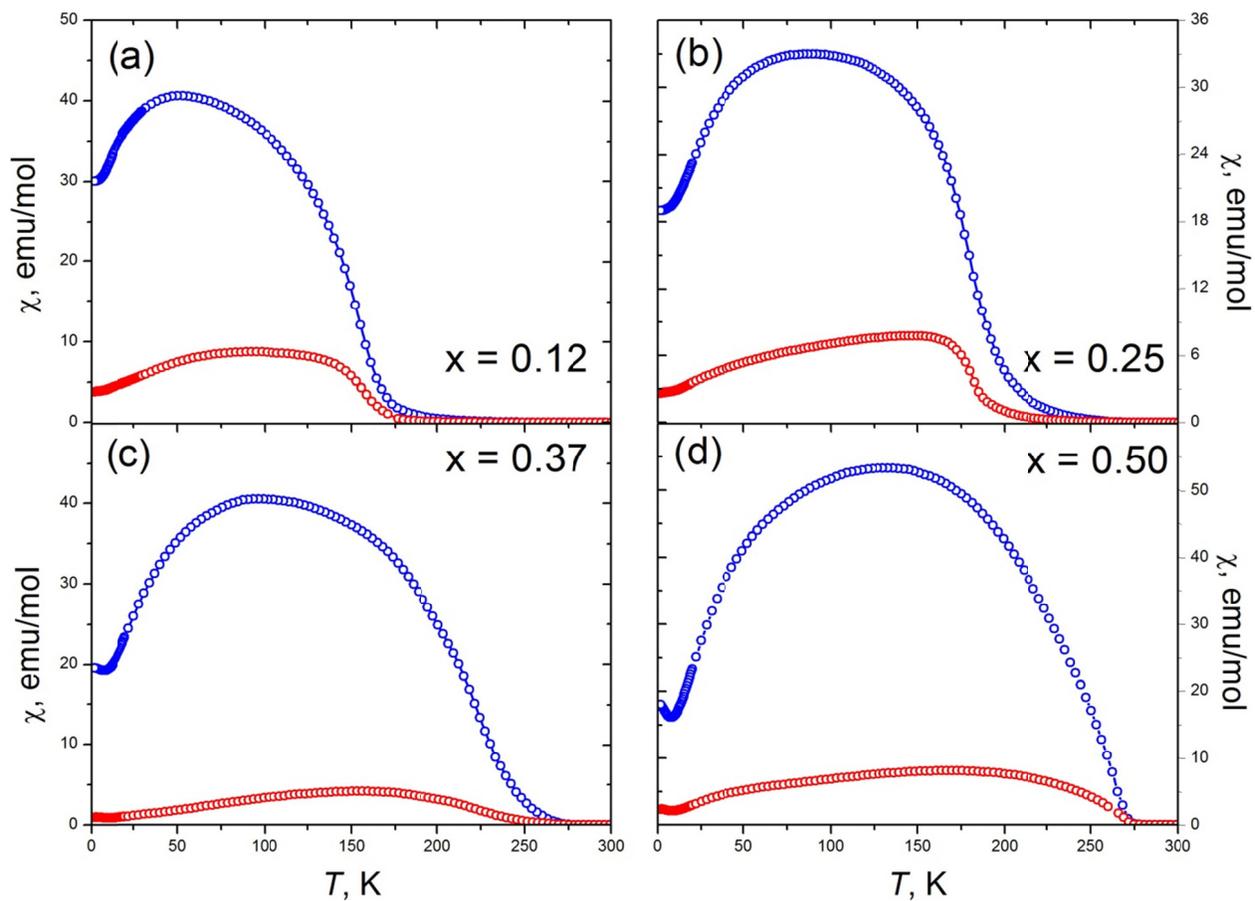


Fig. S2 Temperature dependence of zero-field cooled (red) and field cooled (blue) magnetic susceptibilities at 10 Oe for: a) $\text{La}_{0.88}\text{Nd}_{0.12}\text{Co}_2\text{P}_2$; b) $\text{La}_{0.75}\text{Nd}_{0.25}\text{Co}_2\text{P}_2$; c) $\text{La}_{0.63}\text{Nd}_{0.37}\text{Co}_2\text{P}_2$; and d) $\text{La}_{0.50}\text{Nd}_{0.50}\text{Co}_2\text{P}_2$.

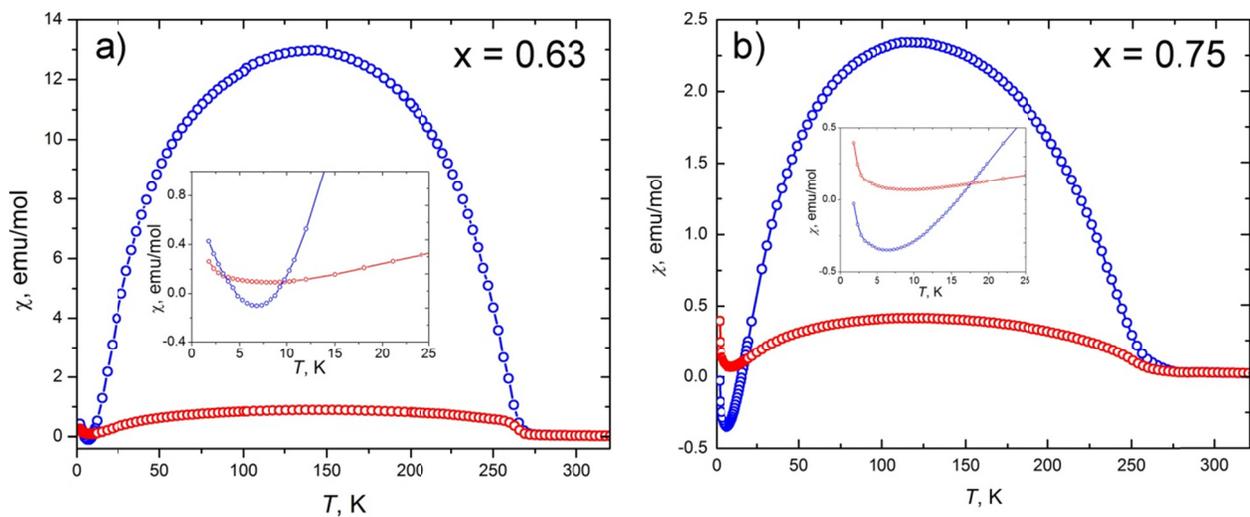


Fig. S3 Temperature dependence of zero-field cooled (red) and field cooled (blue) magnetic susceptibilities at 10 Oe for: a) $\text{La}_{0.37}\text{Nd}_{0.63}\text{Co}_2\text{P}_2$ and b) $\text{La}_{0.25}\text{Nd}_{0.75}\text{Co}_2\text{P}_2$.

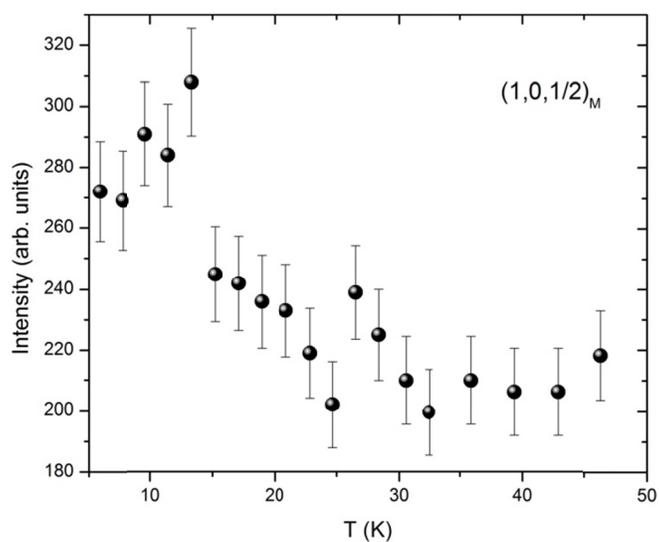


Fig. S4 Temperature dependence of the intensity of the neutron powder diffraction magnetic reflection $(1,0,1/2)_M$ of $\text{La}_{0.25}\text{Nd}_{0.75}\text{Co}_2\text{P}_2$.

Table S1 Results of the magnetic structure refinement for $\text{La}_{0.50}\text{Nd}_{0.50}\text{Co}_2\text{P}_2$ from neutron powder diffraction data collected at various temperatures with the 2.41 Å wavelength. The results of refinements obtained with the 1.54 Å wavelength are also provided, below the 2.41 Å data at 4 K. The results of the simultaneous refinement of both wavelengths are shown in brackets.

Temp. (K)	Nd position [‡] Model	R_{mag}^* (%)	Nd moment Co moment	
			μ_{B}	μ_{B}
4	(0,0,0) & (1/2,1/2,1/2) ^a	42.0	2.2(1)	0.85(5)
		41.5	2.2(1)	0.74(6)
		[68, 35.2]	[2.2(1)]	[0.80(6)]
	(1/2,1/2,1/2) ^b	61.5	3.1(2)	0.73(5)
		41.2	3.1(2)	0.67(6)
		[66.5, 34.4]	[3.1(2)]	[0.73(6)]
50	(0,0,0) & (1/2,1/2,1/2) ^a	68.6	0.7(2)	0.74(5)
	(1/2,1/2,1/2) ^b	65.0	0.7(2)	0.75(5)
125	(0,0,0) & (1/2,1/2,1/2) ^a	54.2	0.2(3)	0.69(6)
	(1/2,1/2,1/2) ^b	54.2	0.2(3)	0.69(6)

[‡]In both magnetic structure models the order of the cobalt moments along the *c*-axis is the same with the sequence +++-,++--.

^aThis magnetic structure model corresponds to the order of the Nd^{3+} moments +++-,++--.

^bEvery other Nd layer has disordered moments and the sequence is 0-0+,0-0+.

Table S2 Results of the magnetic structure refinement for $\text{La}_{0.25}\text{Nd}_{0.75}\text{Co}_2\text{P}_2$ from neutron powder diffraction data collected at various temperatures with the 2.41 Å wavelength. The results of refinements obtained with the 1.54 Å wavelength are also provided, below the 2.41 Å data at 4 K. The results of the simultaneous refinement of both wavelengths are shown in brackets.

Temp. (K)	Nd position [‡] Model	R_{mag}^* (%)	Nd moment Co moment	
			μ_{B}	μ_{B}
4	(0,0,0) & (1/2,1/2,1/2) ^a	59.0	2.03(9)	0.94(7)
		44.8	2.07(8)	0.69(7)
		[59.6, 49.2]	[2.04(9)]	[0.77(7)]
	(1/2,1/2,1/2) ^b	56.9	2.9(1)	0.82(6)
		41.6	2.9(1)	0.60(6)
		[55.7, 45.9]	[2.9(1)]	[0.67(6)]
15	(0,0,0) & (1/2,1/2,1/2) ^a	62.6	1.6(1)	0.88(8)
	(1/2,1/2,1/2) ^b	57.9	2.2(2)	0.81(9)
50	(0,0,0) & (1/2,1/2,1/2) ^a	60.9	0.9(2)	0.8(1)
	(1/2,1/2,1/2) ^b	56.5	1.2(3)	0.7(1)
100	(0,0,0) & (1/2,1/2,1/2) ^a	70.4	0.5(3)	0.7(1)
	(1/2,1/2,1/2) ^b	69.6	0.6(3)	0.7(1)
125	(0,0,0) & (1/2,1/2,1/2) ^a	62.3	0.3(2)	0.88(7)
	(1/2,1/2,1/2) ^b	61.4	0.4(2)	0.88(7)

* Note that the residual factors for the magnetic phase (R-mag) are quite large due to the small intensities of the magnetic reflections as compared to the nuclear Bragg peaks.

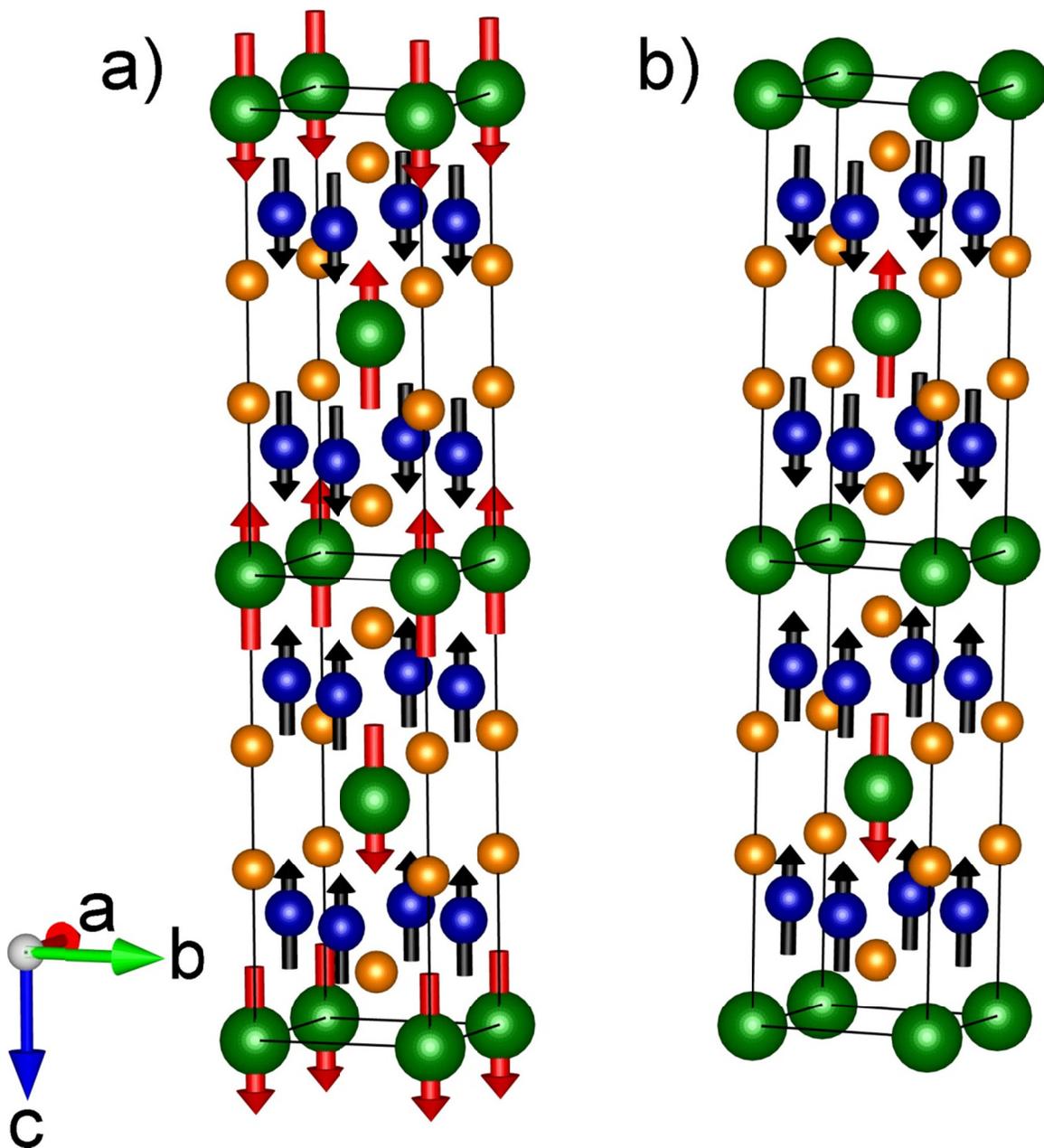


Fig. S5 The two possible magnetic structure models of $\text{La}_{1-x}\text{Nd}_x\text{Co}_2\text{P}_2$. In both magnetic structure models the order of the cobalt moments along the c -axis is the same with the sequence $++-,++-$. a) This magnetic structure model corresponds to the order of the Nd^{3+} moments $++-,++-$ (Nd positions $(0,0,0)$ & $(\frac{1}{2},\frac{1}{2},\frac{1}{2})$). b) Every other Nd layer has disordered moments and the sequence is $0-0+,0-0+$ (Nd position $(\frac{1}{2},\frac{1}{2},\frac{1}{2})$).