

## Supplementary materials

### A Two-step Field-induced Magnetic Transition in a Novel Layered Cobalt Diphosphonate

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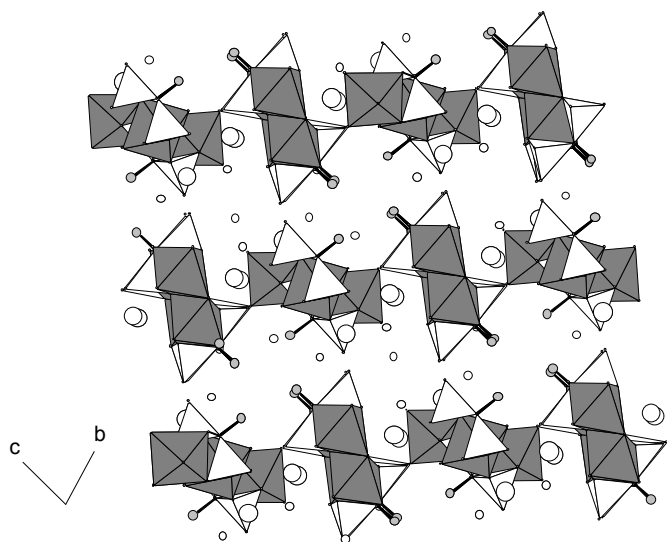
#### Experimental Section

##### *Ion exchanges:*

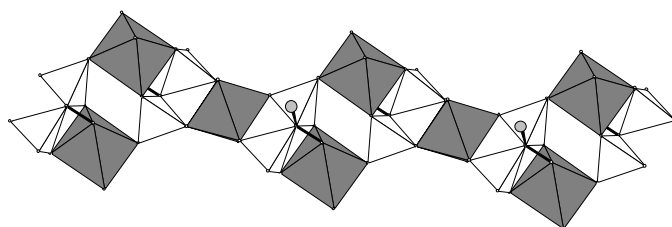
A mixture of compound **1** and an aqueous solution of 1M KCl (or NH<sub>4</sub>Cl, CaCl<sub>2</sub>) was stirred firstly at R.T. for 1h, then at 80°C for 4h. The resulted product was filtered, washed with water and dried in air. The same experiment was repeated once more. The atomic absorption spectra and elemental analysis of the final products show that 18.2%, 58.3% and 95.7% Na<sup>+</sup> ions in compound **1** were replaced by NH<sub>4</sub><sup>+</sup>, K<sup>+</sup> and Ca<sup>2+</sup> ions, respectively. The XRD measurements suggest that the structure of **1** is maintained after ion exchanges.

## Figure Captions:

- Figure S1** (a) Polyhedral representation of the structure of **1** viewed down [100]; CoO<sub>6</sub> octahedra dark shaded, CPO<sub>3</sub> tetrahedra not shaded, Na<sup>+</sup> ions and H<sub>2</sub>O molecules with large and small open circles, respectively. (b) The two types of chains (top: chain I, bottom: chain II) within the layer.
- Figure S2** Temperature dependent molar magnetic susceptibilities of compound **1** at 10 kOe. Inset:  $\chi_M T$  versus T curve for **1**.
- Figure S3**  $\chi_M^{-1}$  versus T curve for **1** at the field of 10 kOe.
- Figure S4** Temperature dependent *ac* magnetic susceptibilities for **1**.
- Figure S5**  $d(\chi_M T)/dT$  versus T curve for **1** at the field of 1 kOe.
- Figure S6** Field dependent *ac* magnetic susceptibilities of **1** at different temperatures.
- Figure S7** Field dependent magnetization of **1** at different temperatures.
- Figure S8** The magnetic phase diagram for **1**.
- Figure S9** Hysteresis loop for **1** at 1.8 K.
- Figure S10** Thermal analysis of **1** with a heating rate of 20°C/min at N<sub>2</sub>.



(a)



(b)

Fig. S1

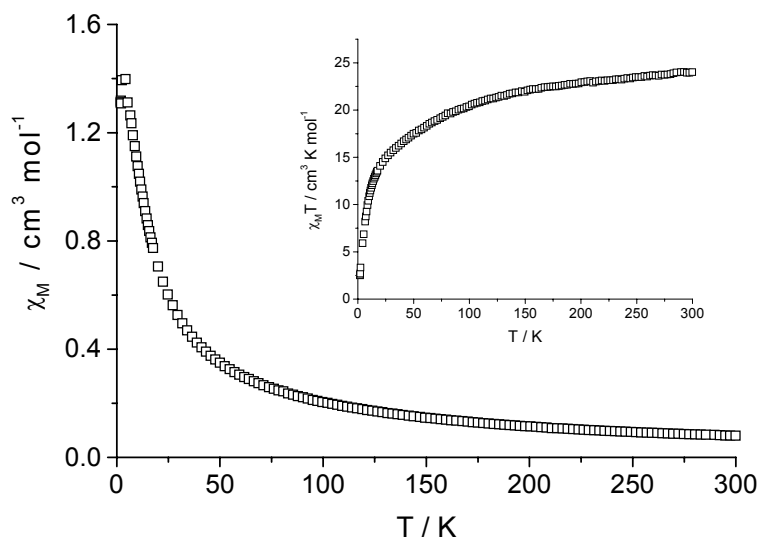


Fig. S2

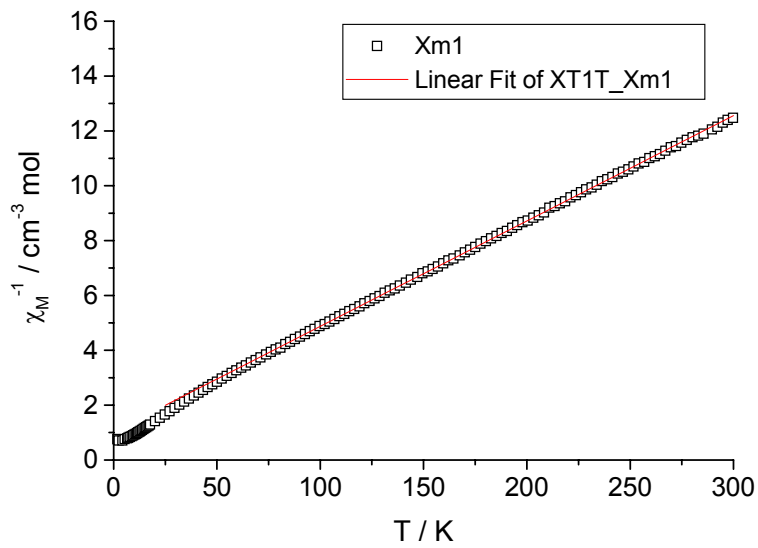


Fig. S3

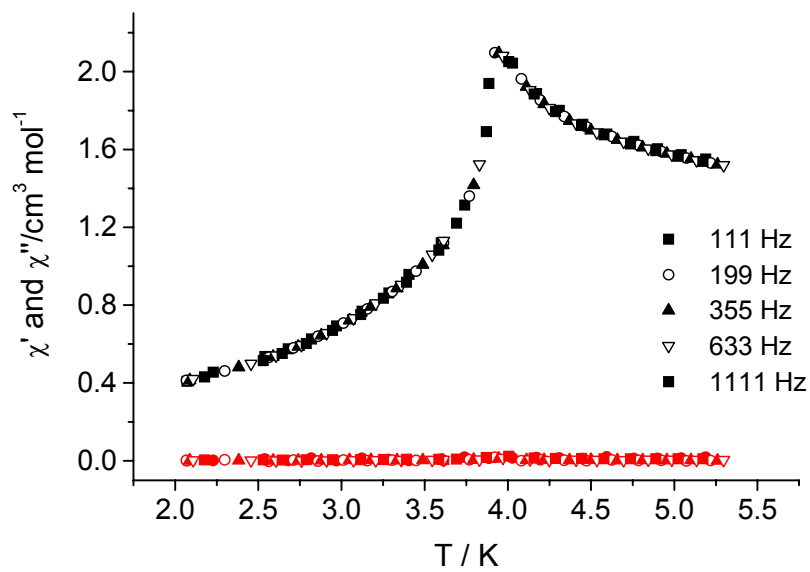


Fig. S4

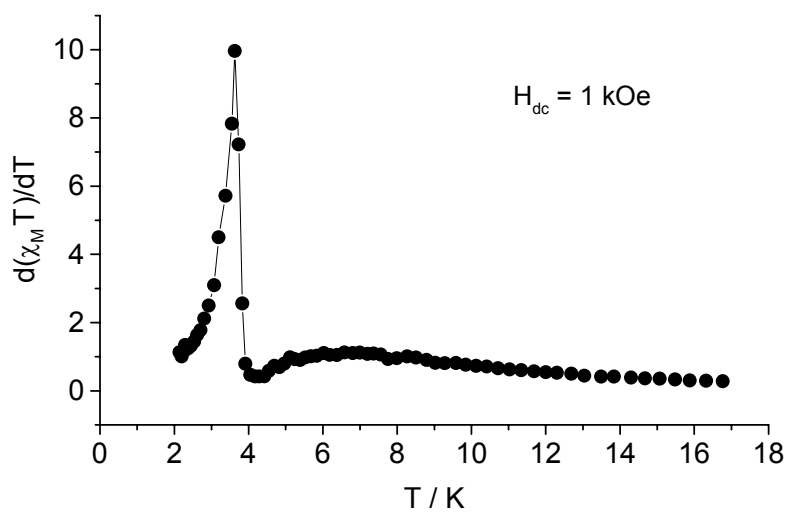


Fig. S5

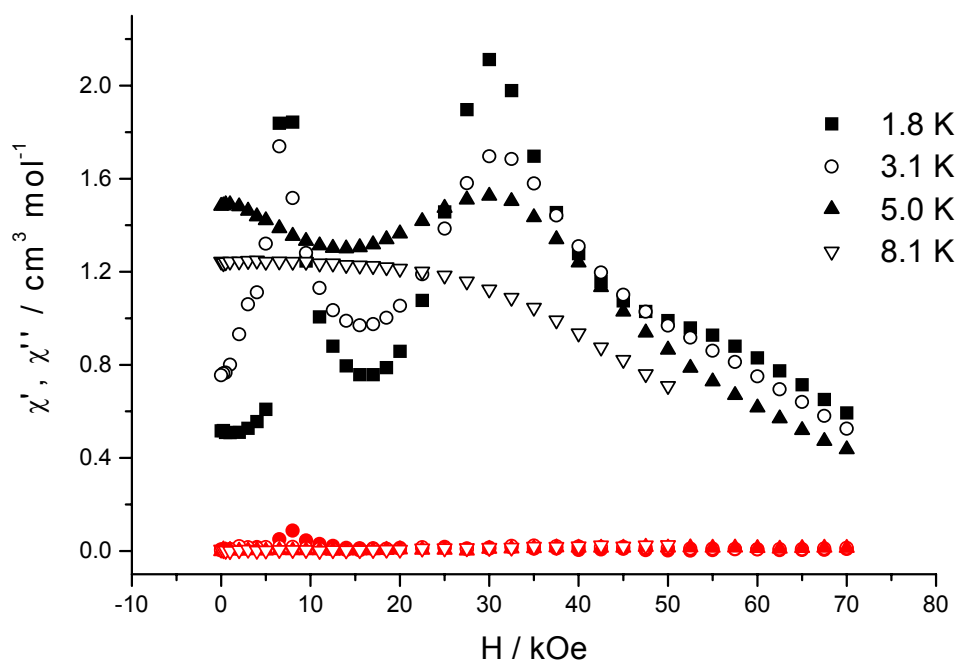


Fig. S6

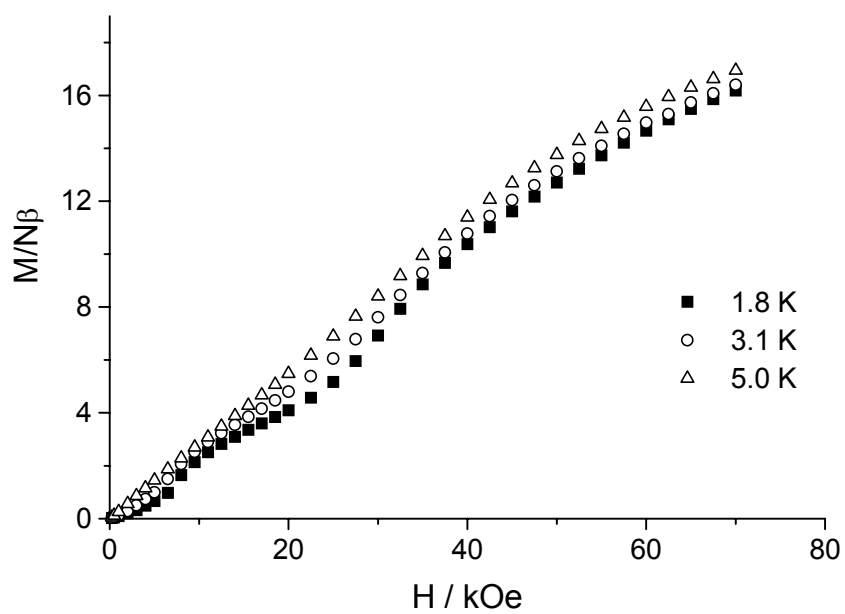


Fig. S7

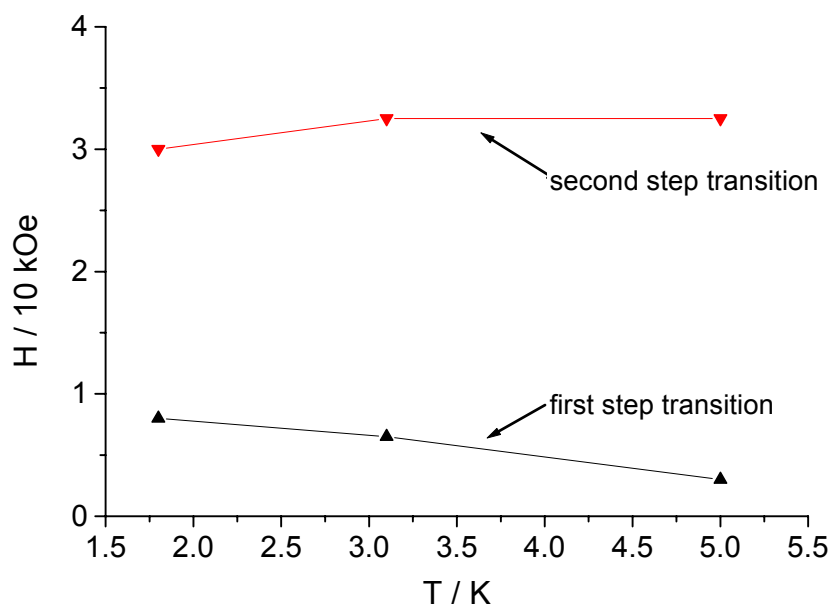


Fig. S8

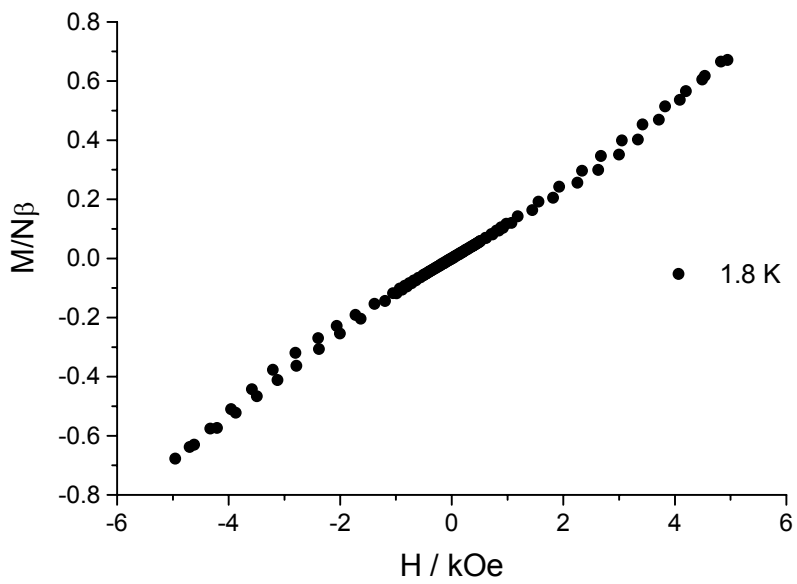


Fig. S9

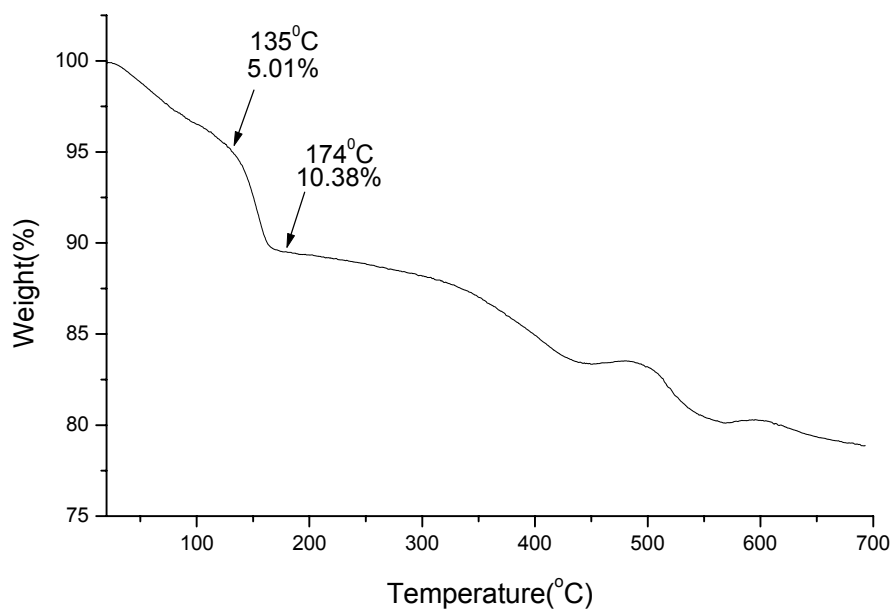


Fig. S10