

## Supplementary Information

Synthesis, structure and magnetic ordering of the mullite-type  $\text{Bi}_2\text{Fe}_{4-x}\text{Cr}_x\text{O}_9$  solid solutions with frustrated pentagonal Cairo lattice.

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Table S1. 298K and 78K  $^{57}\text{Fe}$  Mössbauer hyperfine parameters for  $\text{Bi}_2\text{Fe}_3\text{CrO}_9$ . (IS, isomer shift relative to  $\alpha\text{-Fe}$  at ambient temperature;  $\Delta E_Q$ , apparent quadrupole shift;  $H_{\text{Hf}}$ , magnetic hyperfine field; G, line width; A, relative spectral area)

T, K	Component	IS, mm/s $\pm 0.01$	$\Delta E_Q$ , mm/s $\pm 0.01$	$H_{\text{Hf}}$ , T $\pm 0.1$	A % $\pm 1$	G, mm/s $\pm 0.01$	Assignment
298	S1	0.37	-0.21	50.7	8	0.44	$\text{Fe}_2\text{O}_3$ imp
	D1	0.35	0.38	-	31	0.26	$\text{Fe}^{3+}$ (oct)
	D2	0.24	0.99	-	61	0.31	$\text{Fe}^{3+}$ (tet)
78	S11	0.46	-0.14	53.1	4	0.36	$\text{Fe}_2\text{O}_3$ imp
	S21	0.45	0	47.2	31	0.57	$\text{Fe}^{3+}$ (oct)
	S22	0.32	0.38	43.0	60	0.53	$\text{Fe}^{3+}$ (tet)
	S31	0.36	0.1	7.6	5	0.52	$\text{Fe}^{3+}$ (avr)

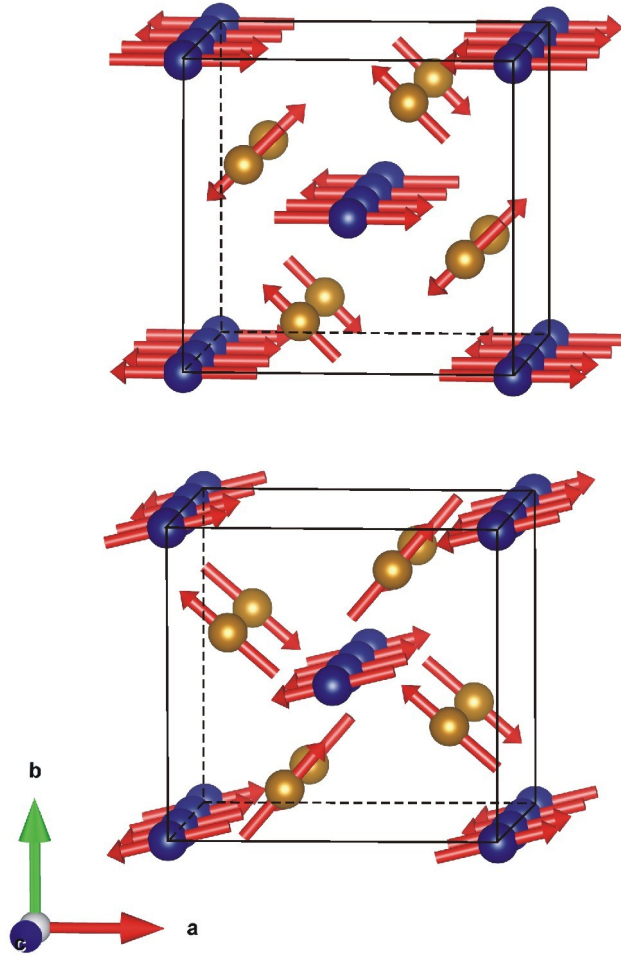


Figure S1. Variants of the  $\text{Bi}_2\text{Fe}_3\text{CrO}_9$  magnetic structure with the magnetic moments confined to  $ab$  plane. The Fe1 and Fe2 sublattices are marked in yellow and blue, respectively. The structure is shown in the  $a, b, 2c$  unit cell, where  $a, b, c$  are the lattice parameters of the nuclear structure.

Table S2. Symmetry operators of the  $F_a\bar{1}$  magnetic space group (S.G.  $P_S\bar{1}$  (#2.7) in Belov-Neronova-Smirnova notations).

Seitz symbol	Symmetry operator	Seitz symbol	Symmetry operator
$(\bar{1} 0,0,0)$	$x, y, z, m$	$(\bar{1} 0,0,\frac{1}{2})'$	$x, y, z+1/2, -m$
$(\bar{1} 0,0,0)$	$-x, -y, -z, m$	$(\bar{1} 0,0,\frac{1}{2})'$	$-x, -y, -z+1/2, -m$
$+(\bar{1} 0,\frac{1}{2},\frac{1}{2}), (\bar{1} \frac{1}{2},0,\frac{1}{2}), (\bar{1} \frac{1}{2},\frac{1}{2},0)$			

Table S3. Fractional coordinates of the magnetic atoms in the magnetic supercell (magn. S.G.  $F_a\bar{1}$ ,  $a = 15.8982(3)\text{\AA}$ ,  $b = 16.7673(3)\text{\AA}$ ,  $c = 11.9520(2)\text{\AA}$ ,  $R_{\text{magn}} = 0.014$ ,  $R_{\text{p}} = 0.013$ ,  $R_{\text{wp}} = 0.014$ ) and magnetic moments in spherical coordinates ( $\mu, \theta, \varphi$ ) at  $T = 30\text{K}$ .

Atom	$x/a$	$y/b$	$z/c$	$\mu$ , $\mu\text{B}$	$\theta$ , deg.	$\varphi$ , deg.
Fe1 <sub>1</sub>	-0.8239	0.1677	1/4	3.40(3)	51(1)	140(2)
Fe1 <sub>2</sub>	-0.5739	0.0823	1/4	$\mu(\text{Fe1}_1)$	$\theta(\text{Fe1}_1)$	$-270 + \varphi(\text{Fe1}_1)$
Fe2 <sub>1</sub>	0	1/4	0.3746	$\mu(\text{Fe1}_1)$	$180 - \theta(\text{Fe1}_1)$	-111(2)
Fe2 <sub>2</sub>	3/4	0	0.6254	$\mu(\text{Fe1}_1)$	$180 - \theta(\text{Fe1}_1)$	$180 + \varphi(\text{Fe2}_1)$

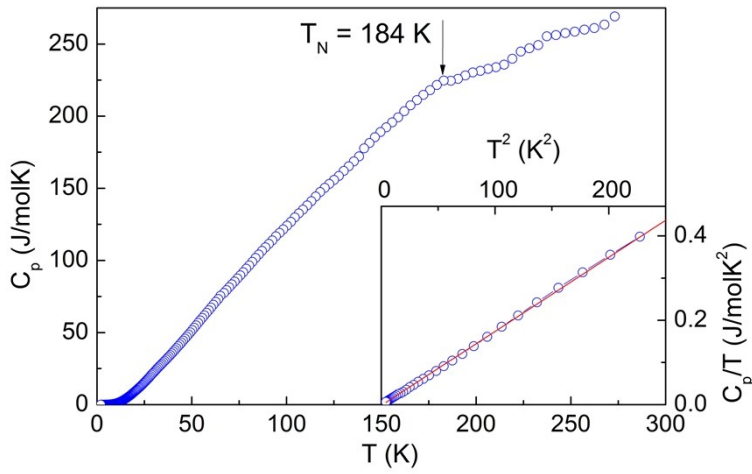


Figure S2. Temperature dependence of heat capacity for  $\text{Bi}_2\text{Fe}_3\text{CrO}_9$ . The insert shows fitting the low temperature part with the  $C_p/T - \alpha T^2$  Debye dependence.