**Fig. S1** Powder X ray diffraction pattern for crystals of 1 (blue) and 3 (green) and simulated one (red) from the structure of 1 at 120 K.
Fig. S2 Two possible configurations of [Fe^{II}(TPMA)(en)]^{2+} complex in 1 at 120 K with 0.85 (a) and 0.15 (b) occupancies. (C (black), N (blue), and Fe (brown)). Hydrogen atoms have been omitted for clarity.
**Fig. S3** Unit cell parameters of 2 at different temperatures calculated from single crystal X-ray diffraction data.
Fig. S4 Views of the helices of [Fe^{II}(TPMA)(2-pic)]^{2+} complexes of opposite helicity in the structure of 2 in crystals solved as $P_{4_1}$ at 90 K (a) and as $P_{4_3}$ at 120 K (b).
**Fig. S5** Powder X ray diffraction pattern for crystals of 2 (blue) and simulated one (red) from the structure of 2 at 300 K.