Electronic Supplementary Information (ESI) for

Enhancement of the coercivity in Co-Ni layered double hydroxides by increasing basal spacing

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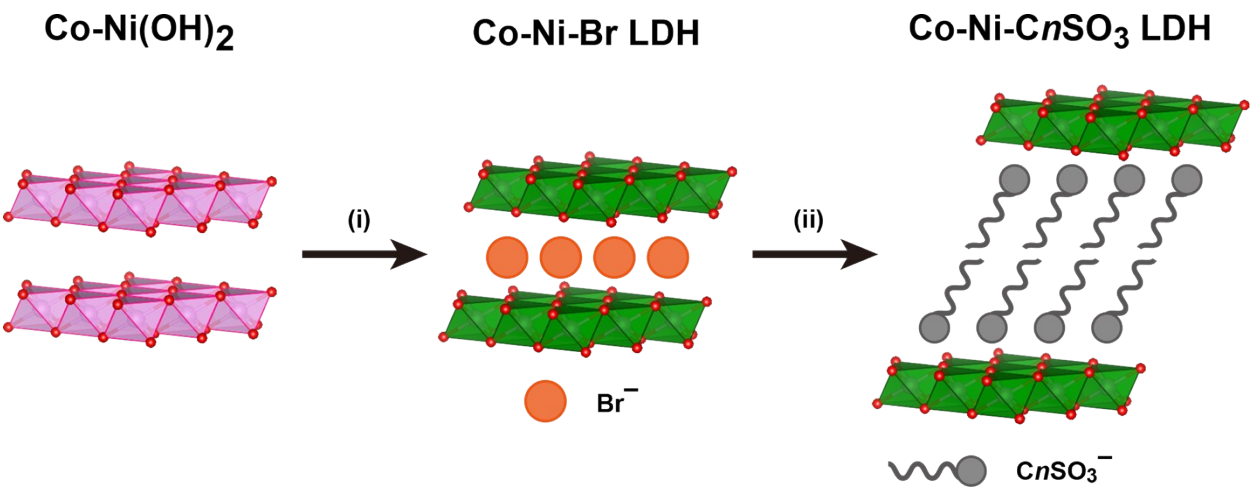
Contents.

Scheme 1. Synthesis of Co-Ni-C*n*SO_3 LDH[a]



[a]Illustrations drawn by using the software VESTA 3: K. Momma and F. Izumi, VESTA 3 for Three-Dimensional Visualization of Crystal, Volumetric and Morphology Data. *J. Appl. Crystallogr.*, 2011, **44**, 1272–1276.

Synthesis of the precursor, Co-Ni$(OH)_2$, was carried out as follows. Cobalt chloride hexahydrate ($CoCl_2 \cdot 6H_2O$, 1.19 g, 5.00 mmol), nickel chloride hexahydrate ($NiCl_2 \cdot 6H_2O$, 0.594 g, 2.50 mmol), and hexamethylenetetramine (6.31 g, 45.0 mmol) were dissolved into water (1000 mL), and refluxed for 5 h under an argon atmosphere. The precipitate was filtered, washed with water and ethanol, and then air-dried.

Synthesis of Co-Ni-Br LDH was carried out with an oxidative intercalation approach. Bromine (1.12 g, 12.1 mmol) was dissolved into acetonitrile (600 mL). Co-Ni$(OH)_2$ (1.07 g) was dispersed into the solution after purging with argon gas, followed by stirring for 24 h at room temperature under an argon atmosphere. The precipitate was collected by centrifugation, washed with ethanol, and air-dried.

Synthesis of Co-Ni-NO_3 LDH was carried out with an ethanol-assisted anion-exchange approach. Sodium nitrate (25.5 g, 3.00 mmol) was dissolved into ethanol/water solution (200 mL, 1:1 v/v). Co-Ni-Br LDH (0.250 g) was dispersed into the solution after purging with argon gas, followed by stirring for 24 h at room temperature under an argon atmosphere. The precipitate was filtered, washed with water and ethanol, and then air-dried.

Synthesis of Co-Ni-C*n*SO_3 LDH was carried out with an ethanol-assisted anion-exchange approach. Added amount of *n*-alkylsulfonates, Co-Ni-Br LDH, and volume of ethanol/water solution were listed as Table S1. The dispersion was purged with argon gas, followed by stirring for 24 h at room temperature (n = 4) or refluxing 24 h (n = 6, 8, 10, and 12) under an argon atmosphere. The product was filtered, washed with ethanol, and air-dried.

Table S1. Added amount of Co-Ni-Br LDH, *n*-alkylsulfonates, and volume of ethanol/water solution.

Sample	Co-Ni-Br LDH (g)	*n*-Alkylsulfonate (g; mmol)	Ethanol/Water (mL)
Co-Ni-C*4*SO_3 LDH	0.134	26.2; 164	120
Co-Ni-C*6*SO_3 LDH	0.188	40.1; 220	160
Co-Ni-C*8*SO_3 LDH	0.182	47.5; 220	160
Co-Ni-C*10*SO_3 LDH	0.185	49.1; 201	160
Co-Ni-C*12*SO_3 LDH	0.155	50.6; 186	240

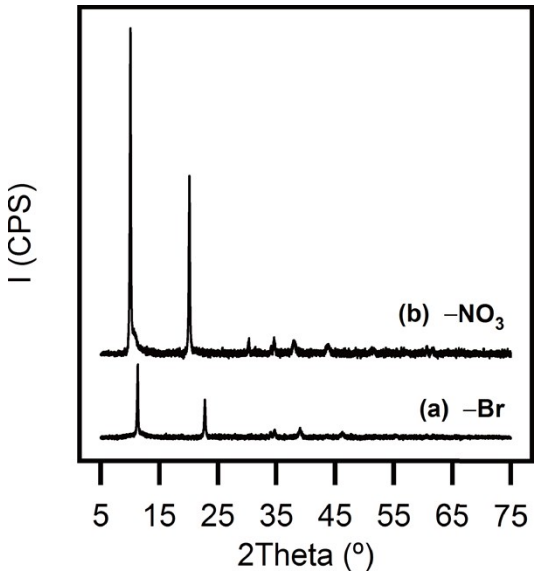


Fig S1. XRD patterns: Co-Ni-Br (a), and -NO_3 (b) LDH.

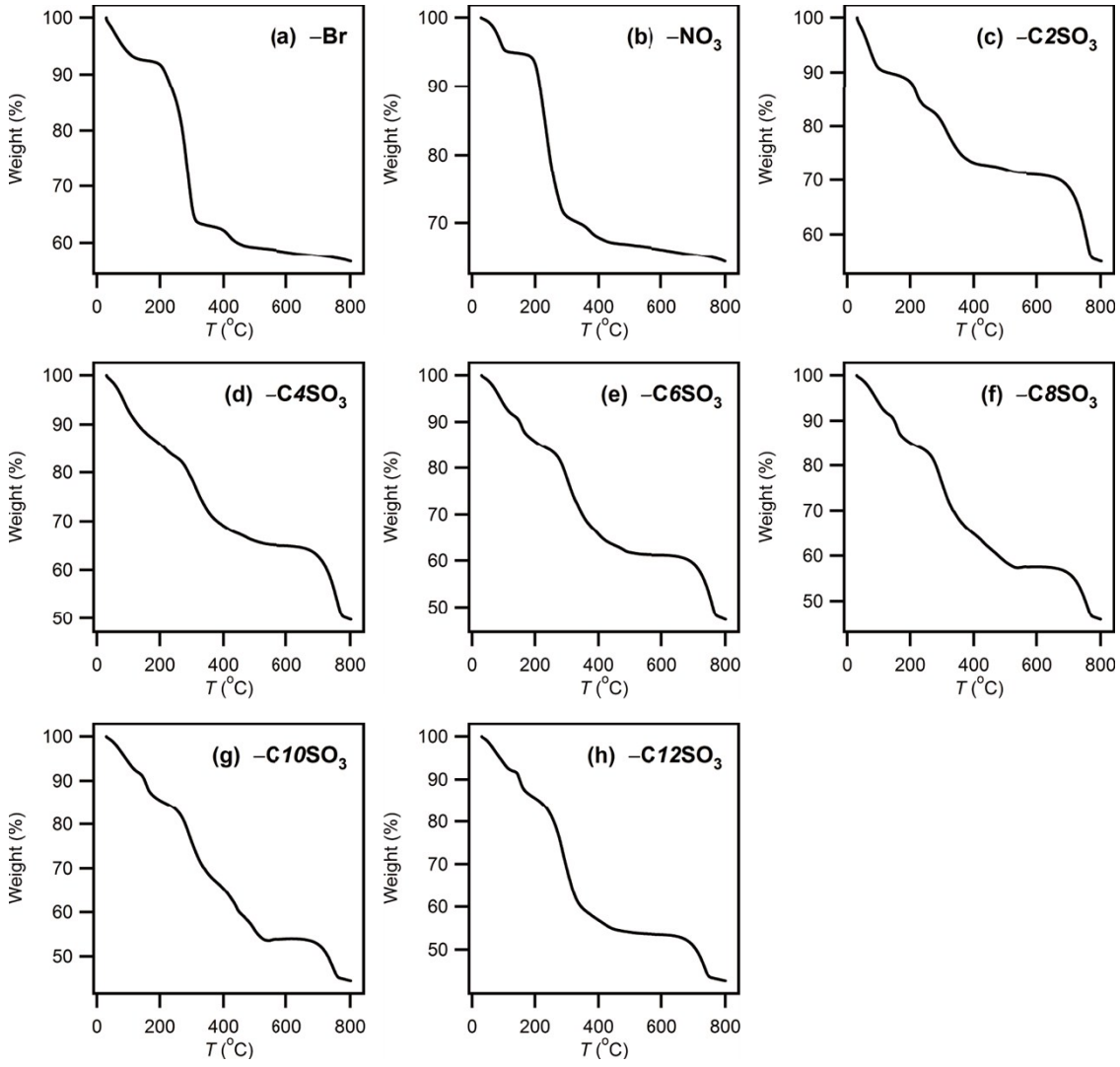


Fig S2. TG curves: Co-Ni-Br (a), -NO_3 (b), -C*2*SO_3 (c), -C*4*SO_3 (d), -C*6*SO_3 (e), -C*8*SO_3 (f), -C*10*SO_3 (g), -C*12*SO_3 (h) LDH.

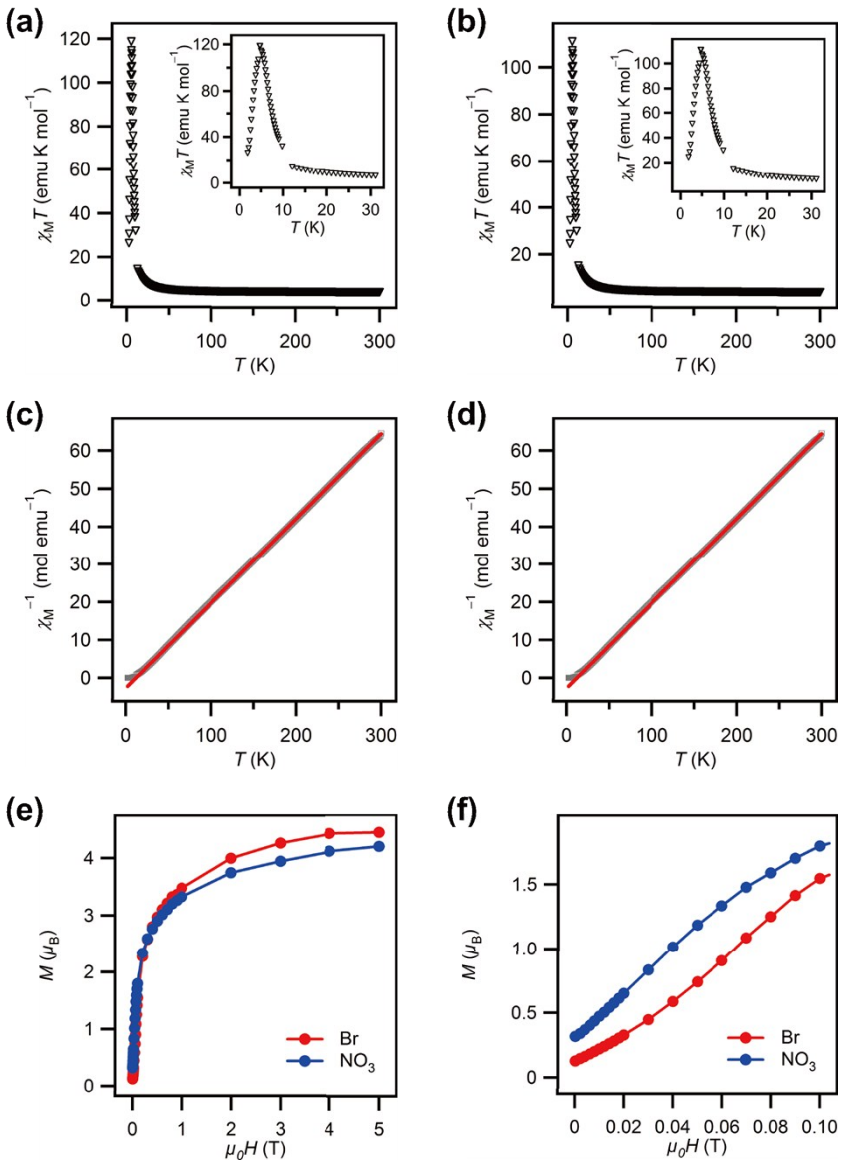


Fig S3. Plots of $\chi_M T$ against T between 2 and 300 K for Co-Ni-Br (a) and -NO_3 (b) LDH. Inset shows the plots of $\chi_M T$ against T in the low-temperature region (2−30 K). Plots of χ_M^{-1} against T between 2 and 300 K for Co-Ni-Br (c) and -NO_3 (d) LDH. The red solid line is fit to the Curie−Weiss law in the high temperature region (150−300 K). Field-dependent magnetization at 2 K: (e) up to 5 T and (f) the low-field region up to 0.1 T. The solid lines are guided to eyes.

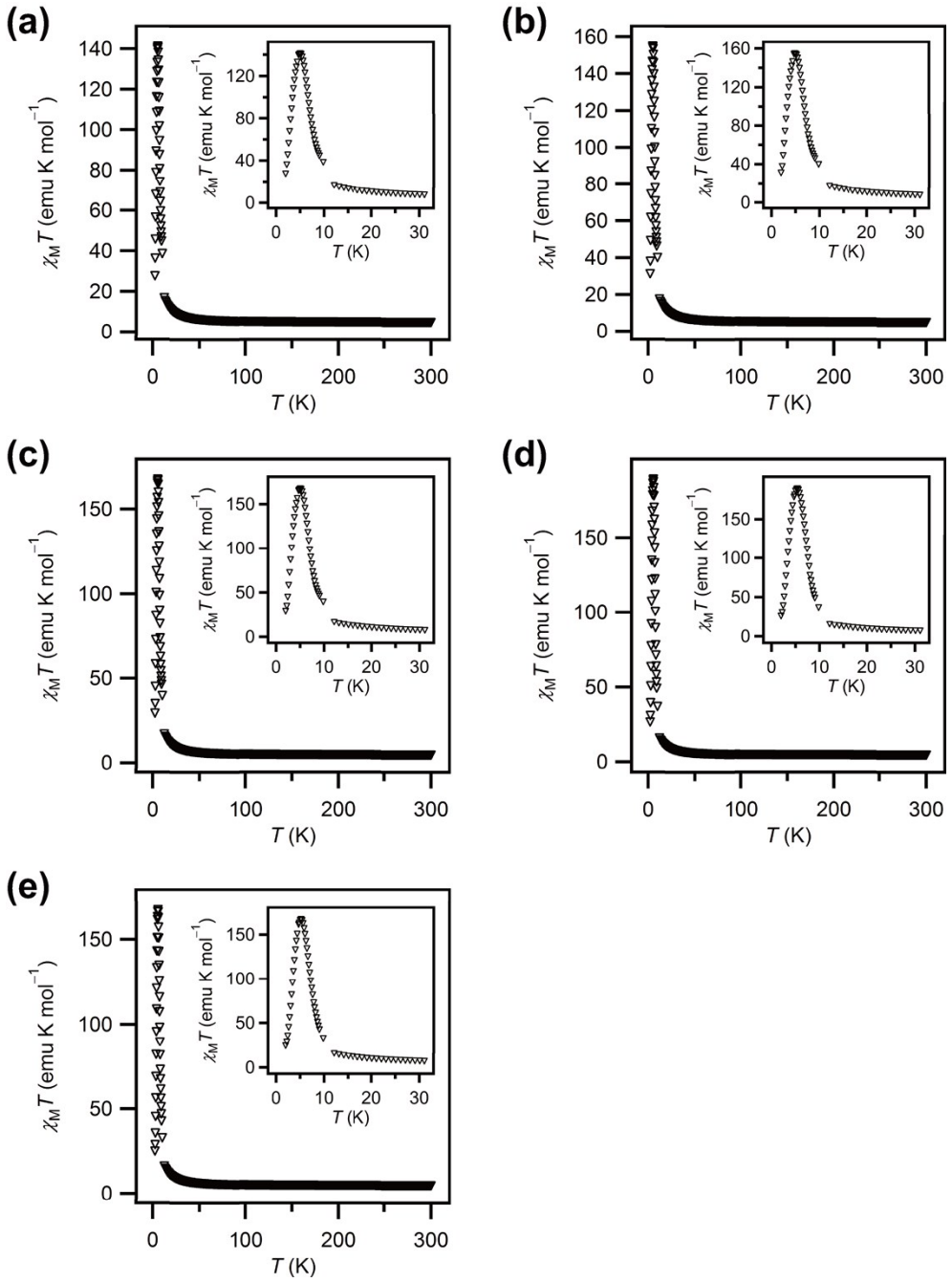


Fig S4. Plots of $\chi_M T$ against T between 2 and 300 K under 5000 Oe: Co-Ni-C*4*SO_3 (a), -C*6*SO_3 (b), -C*8*SO_3 (c), -C*10*SO_3 (d), -C*12*SO_3 (e) LDH. Inset shows the plots of $\chi_M T$ against T in the low-temperature region (2−30 K).

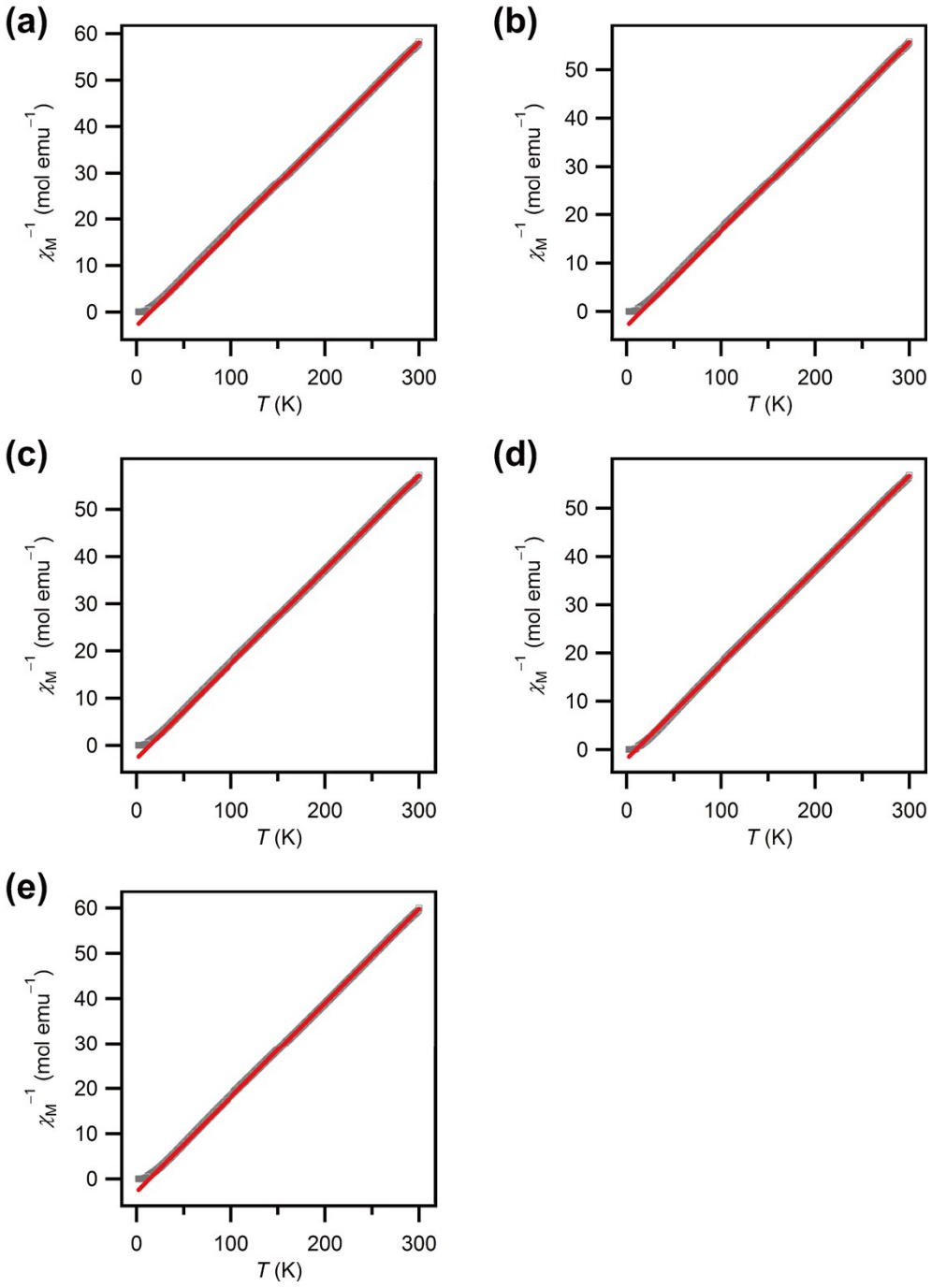


Fig S5. Plots of χ_M^{-1} against T between 2 and 300 K under 5000 Oe: Co-Ni-C*4*SO_3 (a), -C*6*SO_3 (b), -C*8*SO_3 (c), -C*10*SO_3 (d), -C*12*SO_3 (e) LDH. The red solid line is fit to the Curie−Weiss law in the high temperature region (150−300 K).

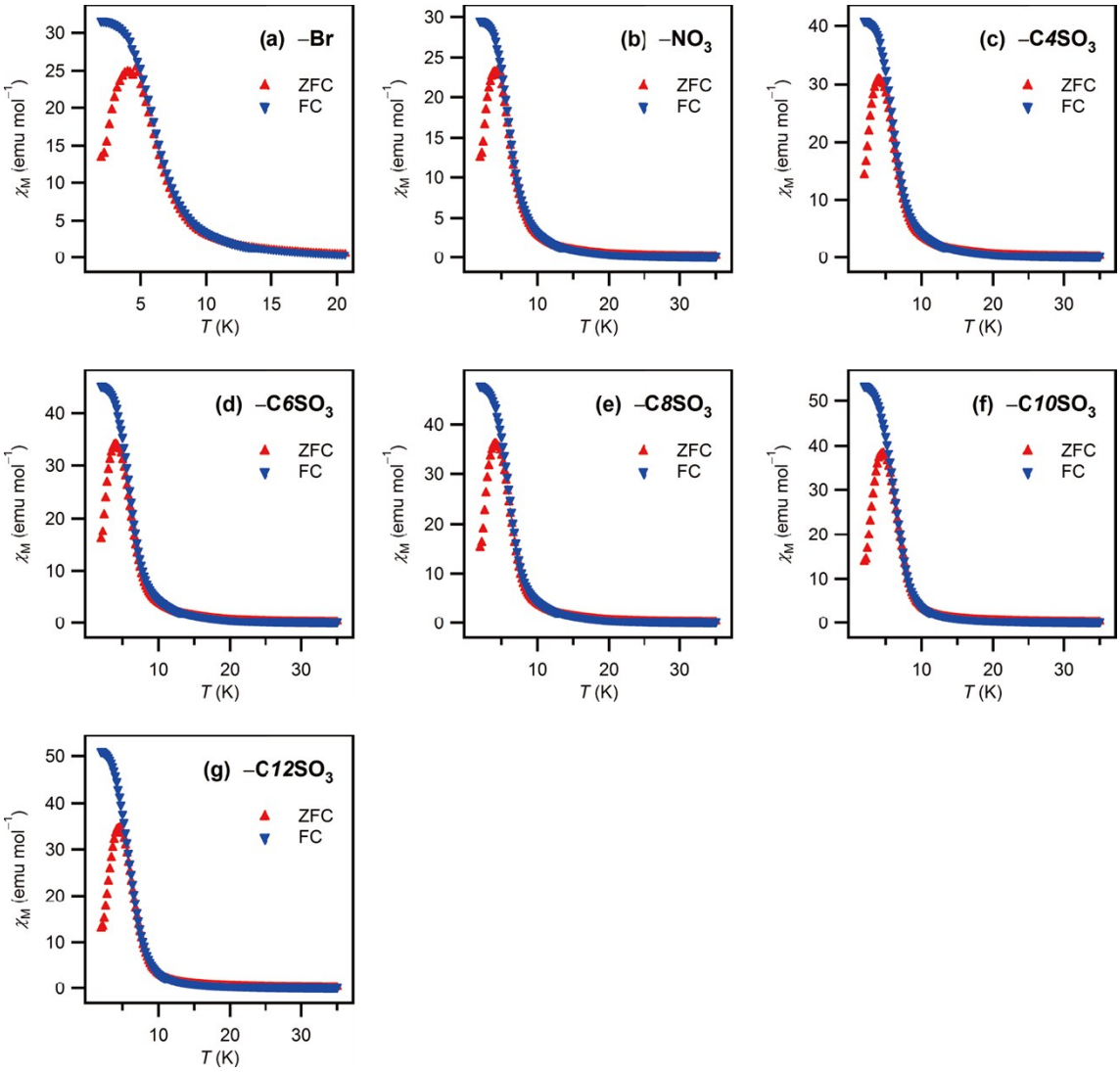


Fig S6. ZFC-FC magnetizations under 100 Oe: Co-Ni-Br (a), -NO_3 (b), -C*4*SO_3 (c), -C*6*SO_3 (d), -C*8*SO_3 (e), -C*10*SO_3 (f), -C*12*SO_3 (g) LDH.

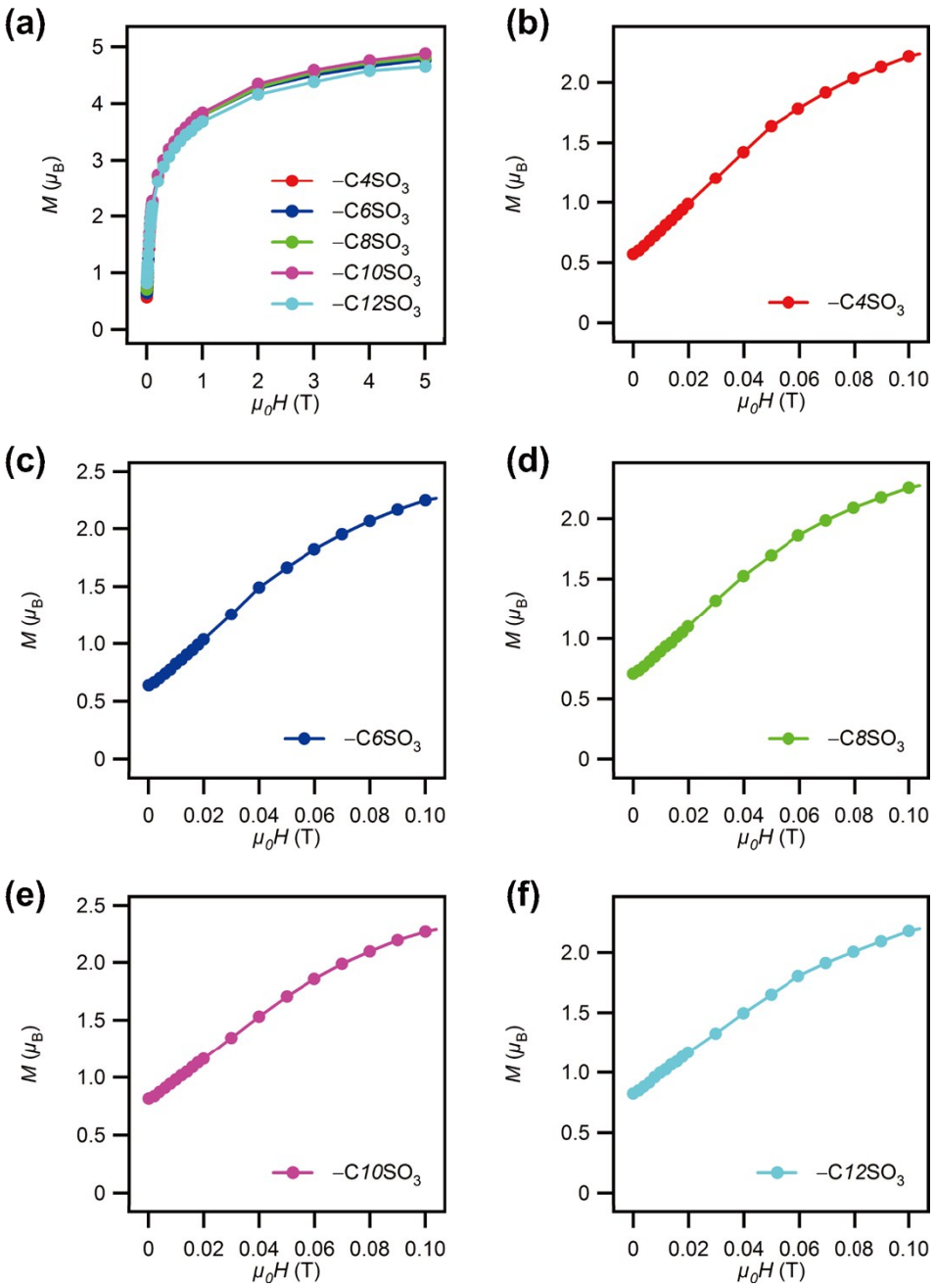


Fig S7. Field-dependent magnetizations at 2 K: (a) up to 5 T for Co-Ni-C*n*SO_3 (*n* = 4, 6, 8, 10, and 12), and the low-field region up to 0.1 T for (b) Co-Ni-C*4*SO_3, (c) -C*6*SO_3, (d) -C*8*SO_3, (e) -C*10*SO_3, (f) -C*12*SO_3 LDH. The solid lines are guided to eyes.

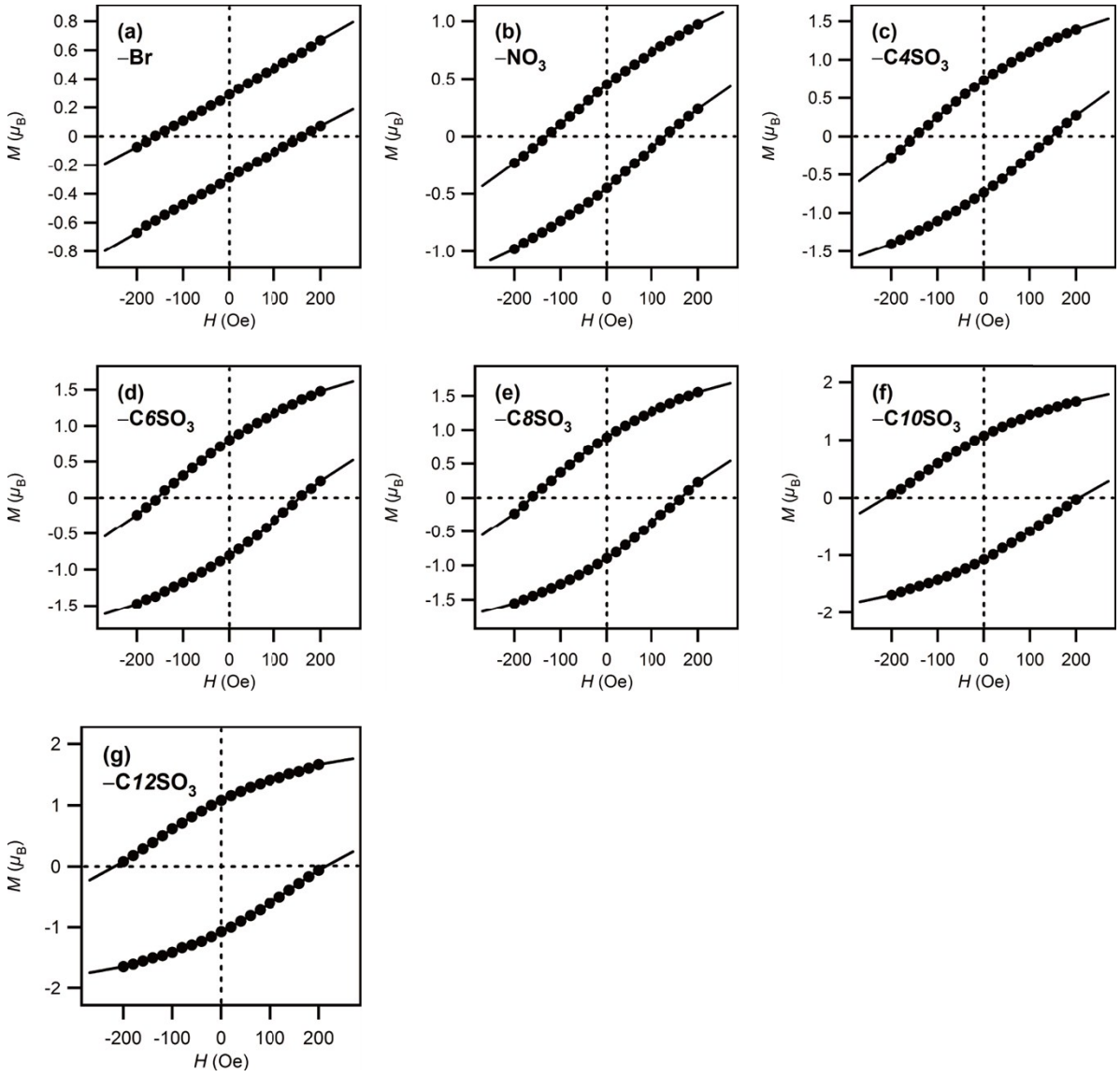


Fig S8. Hysteresis loops at 2 K: Co-Ni-Br (a), -NO_3 (b), -C*4*SO_3 (c), -C*6*SO_3 (d), -C*8*SO_3 (e), -C*10*SO_3 (f), -C*12*SO_3 (g) LDH. The solid lines are guided to eyes.