Electronic Supplementary Material (ESI) for Journal of Materials Chemistry C.
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Electronic Supplementary Information for Theoretical Prediction of the Carrier Mobilities for $M^{II}_2M^{III}$–Cl–Layered Double Hydroxides in the Three-Dimensional Directions

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Table S1. Band Gap Energies (E_g), Work Functions (W), Valence Band Maximum (E_{VBM}), and Conduction Band Minimum (E_{CBM}) of Mg_2Fe–Cl–LDH, Mg_2Ga–Cl–LDH, Ni_2Fe–Cl–LDH, Ni_2Ga–Cl–LDH, Zn_2Fe–Cl–LDH, and Zn_2Ga–Cl–LDH under Lattice Dilation and Compression in x and y Directions

model	direction	$\Delta l / l_0$	E_g (eV)	W (eV)	E_{VBM} (eV)	E_{CBM} (eV)
Mg_2Fe–Cl–LDH	x	–1%	1.819	4.685	–5.459	–3.637
		–0.5%	1.823	4.548	–5.529	–3.709
		0%	1.832	4.753	–5.695	–3.775
		0.5%	1.879	4.589	–5.769	–3.887
		1%	1.889	4.919	–5.864	–3.974
	y	–1%	1.791	4.672	–5.631	–3.819
		–0.5%	1.800	4.613	–5.567	–3.777
		0%	1.812	4.725	–5.537	–3.713
		0.5%	1.827	4.553	–5.513	–3.671
		1%	1.827	4.624	–5.466	–3.640
Mg_2Ga–Cl–LDH	x	–1%	4.169	4.753	–4.844	–0.885
		–0.5%	4.159	4.764	–4.838	–0.878
		0%	4.146	4.729	–4.802	–0.868
		0.5%	4.127	4.728	–4.792	–0.865
		1%	4.116	4.726	–4.784	–0.856
	y	–1%	4.167	4.759	–4.843	–0.876
		–0.5%	4.143	4.749	–4.820	–0.878
		0%	4.125	4.742	–4.804	–0.879
		0.5%	4.097	4.739	–4.788	–0.882
		1%	4.075	4.717	–4.755	–0.891
Ni_2Fe–Cl–LDH	x	–1%	1.910	4.660	–5.615	–3.421
		–0.5%	1.887	4.433	–5.499	–3.489
		0%	1.875	4.359	–5.407	–3.605
		0.5%	1.859	4.669	–5.377	–3.639
		1%	1.843	4.665	–5.297	–3.743
	y	–1%	1.695	4.684	–5.531	–3.837
		–0.5%	1.695	4.631	–5.478	–3.784
		0%	1.669	4.487	–5.421	–3.703
		0.5%	1.662	4.459	–5.361	–3.653
		1%	1.638	4.482	–5.290	–3.628
Ni_2Ga–Cl–LDH	x	–1%	2.554	4.746	–6.024	–4.169
		–0.5%	2.556	4.746	–5.973	–4.108
		0%	2.571	4.556	–5.941	–4.071
		0.5%	2.572	4.554	–5.860	–4.008

		1%	2.573	4.542	–5.829	–3.955
		–1%	2.531	4.598	–6.286	–4.048
		–0.5%	2.545	4.576	–6.273	–4.033
	y	0%	2.545	4.576	–6.261	–4.024
		0.5%	2.538	4.617	–6.259	–4.014
		1%	2.547	4.578	–6.248	–4.005
		–1%	2.187	4.724	–5.818	–3.630
		–0.5%	2.176	4.763	–5.839	–3.645
	x	0%	2.197	4.772	–5.851	–3.653
		0.5%	2.184	4.747	–5.861	–3.665
		1%	2.174	4.764	–5.871	–3.677
Zn_2Fe–Cl–LDH		–1%	1.485	4.696	–5.399	–4.003
		–0.5%	1.355	4.747	–5.424	–4.025
	y	0%	1.375	4.712	–5.439	–4.070
		0.5%	1.390	4.789	–5.466	–4.070
		1%	1.396	4.768	–5.484	–4.094
		–1%	3.131	4.813	–4.879	–1.518
		–0.5%	3.135	4.793	–4.861	–1.547
	x	0%	3.093	4.798	–4.852	–1.536
		0.5%	3.131	4.785	–4.850	–1.520
		1%	3.132	4.786	–4.844	–1.520
Zn_2Ga–Cl–LDH		–1%	3.109	4.803	–4.857	–1.549
		–0.5%	3.118	4.797	–4.856	–1.538
	y	0%	3.116	4.788	–4.846	–1.535
		0.5%	3.111	4.790	–4.845	–1.530
		1%	3.109	4.783	–4.838	–1.528

Table S2. Transfer Integral V, Reorganization Energy λ, Gibbs Free Energy Change ΔG, Transfer Rate k, and Carrier Mobility μ for $[Mg_2Ga(OH)_6(OH_2)_7]^+$ Using Different Exchange-Correlation Functionals

exchange-correlation functional	charge carrier	V (eV)	λ (eV)	ΔG (eV)	k (s^{-1})	μ ($cm^2 \cdot V^{-1} \cdot s^{-1}$)
B3LYP	e	0.049	2.20	6.54	4×10^{-133}	5×10^{-164}
	h					
M06	e	0.030	2.30	4.56	2×10^{-73}	2×10^{-104}
	h					
PBE	e	0.088	2.61	3.75	6×10^{-52}	9×10^{-83}
	h					

Table S3. Transfer Integrals V, Reorganization Energies λ, Gibbs Free Energy Changes ΔG, Transfer Rates k, and Carrier Mobilities μ for Carrier Transportation in Mg_2Ga-Cl-LDH Matrix Clusters with Different Sizes

chemical formula	charge carrier	V (eV)	λ (eV)	ΔG (eV)	k (s^{-1})	μ ($cm^2 \cdot V^{-1} \cdot s^{-1}$)
$[Mg_2Al(OH)_6(OH_2)_7]\cdot Cl$	e	0.049	2.20	6.54	4×10^{-133}	5×10^{-164}
	h					
$[Mg_4Al_2(OH)_{12}(OH_2)_{10}]\cdot 2Cl$	e	0.693	1.75	3.44	1×10^{-49}	2×10^{-80}
	h					
$[Mg_6Al_3(OH)_{18}(OH_2)_{12}]\cdot 3Cl$	e	0.715	3.27	3.18	2×10^{-38}	2×10^{-69}
	h					

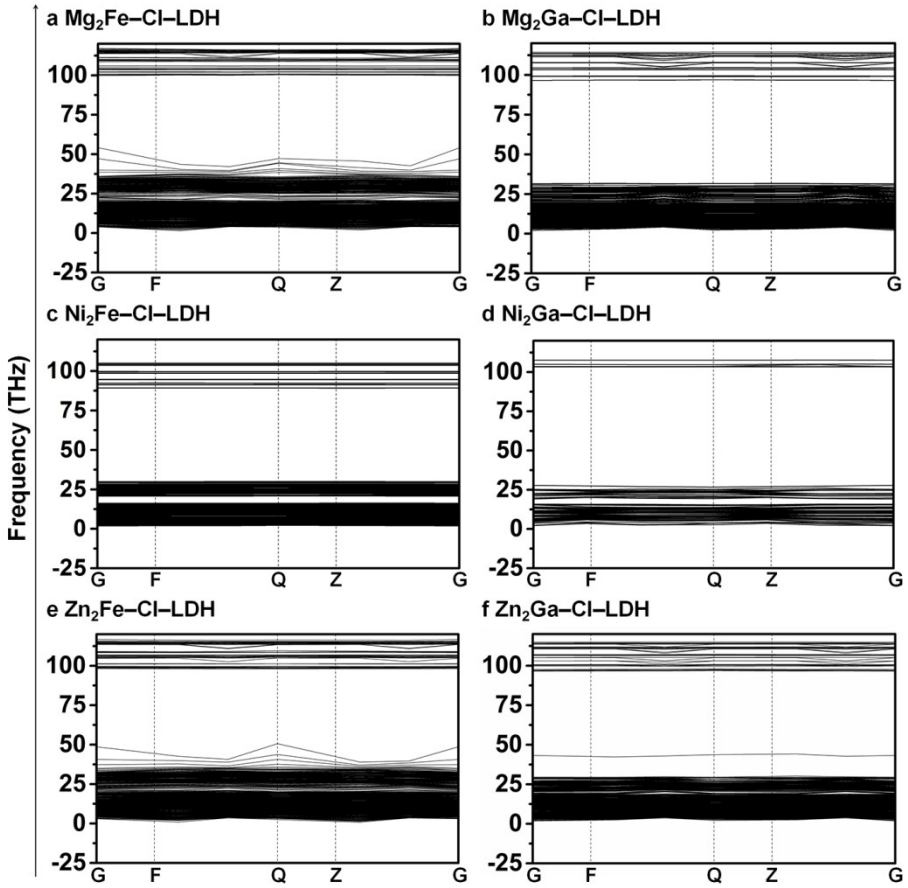

Figure S1. Phonon dispersion curves of (a) Mg_2Fe–Cl–LDH, (b) Mg_2Ga–Cl–LDH, (c) Ni_2Fe–Cl–LDH, (d) Ni_2Ga–Cl–LDH, (e) Zn_2Fe–Cl–LDH, and (f) Zn_2Ga–Cl–LDH, respectively.

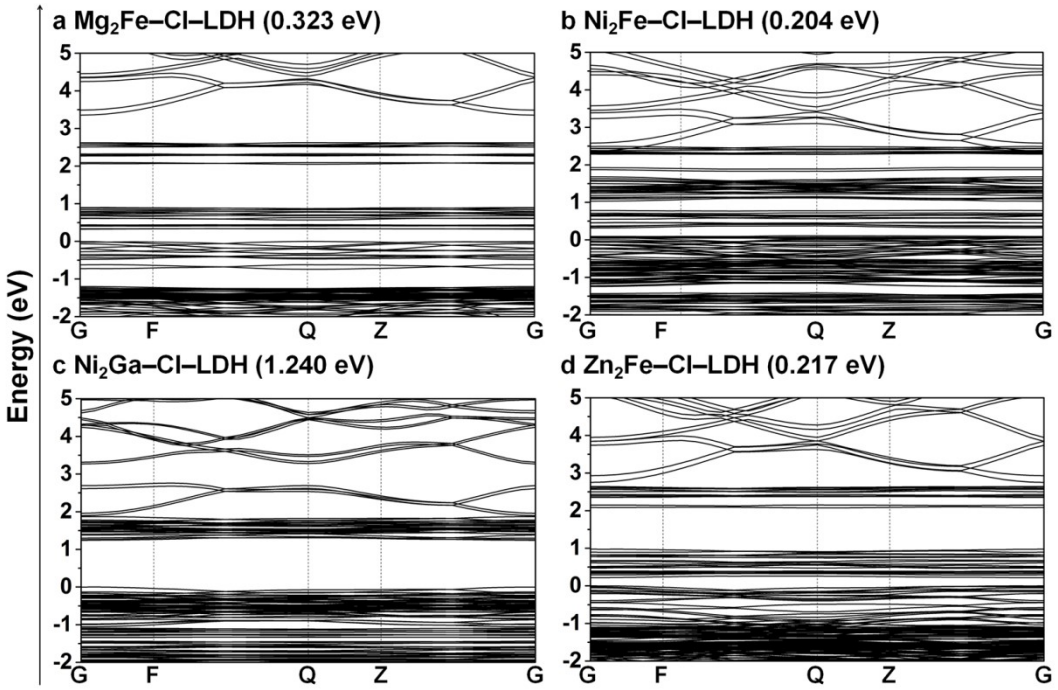

Figure S2. Band structures of Mg_2Fe–Cl–LDH, Ni_2Fe–Cl–LDH, Ni_2Ga–Cl–LDH, and Zn_2Fe–Cl–LDH without the Hubbard correction. The band gap energy of each LDH is listed in the bracket.

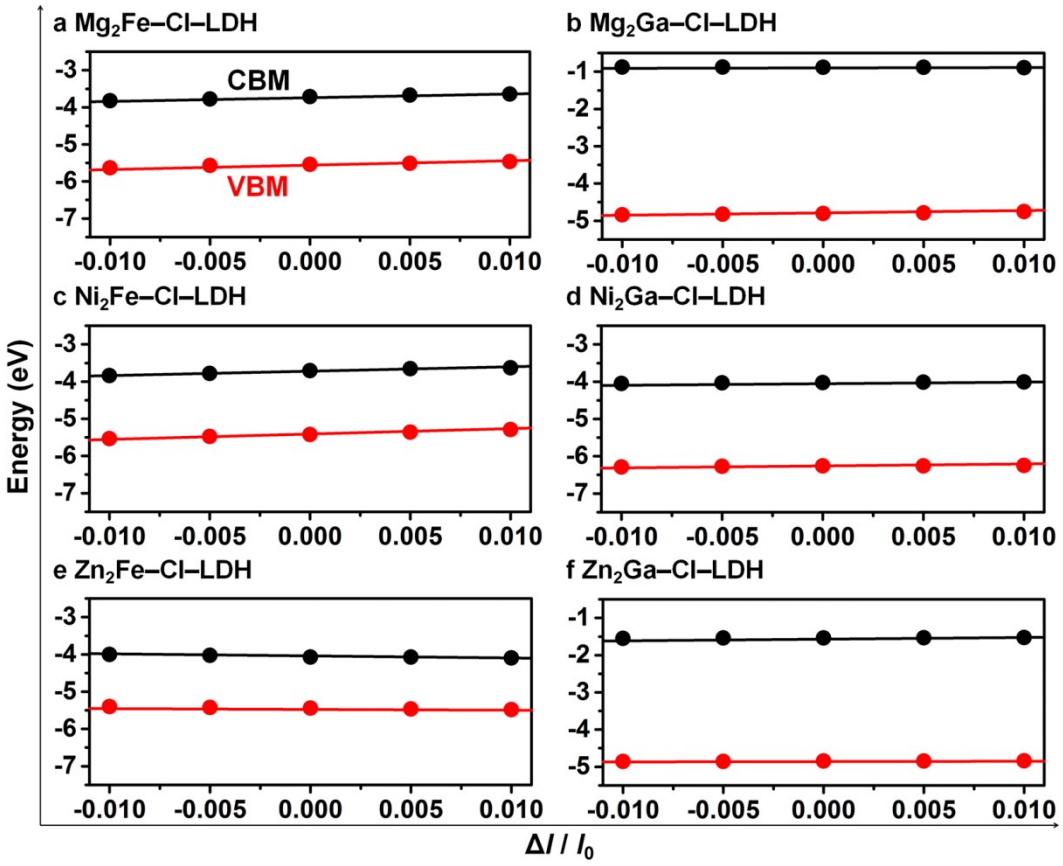

Figure S3. Linear fitting of the conduction band minimum and valence band maximum shift (Δv) with respect to the lattice dilation and compression ($\Delta l/l_0$) in the *y* direction for (a) Mg_2Fe–Cl–LDH, (b) Mg_2Ga–Cl–LDH, (c) Ni_2Fe–Cl–LDH, (d) Ni_2Ga–Cl–LDH, (e) Zn_2Fe–Cl–LDH, and (f) Zn_2Ga–Cl–LDH, respectively.