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Supporting Information for

Direction control of the easy magnetization axis in the magnetic GdN and GdNX (X=F, Cl) monolayers

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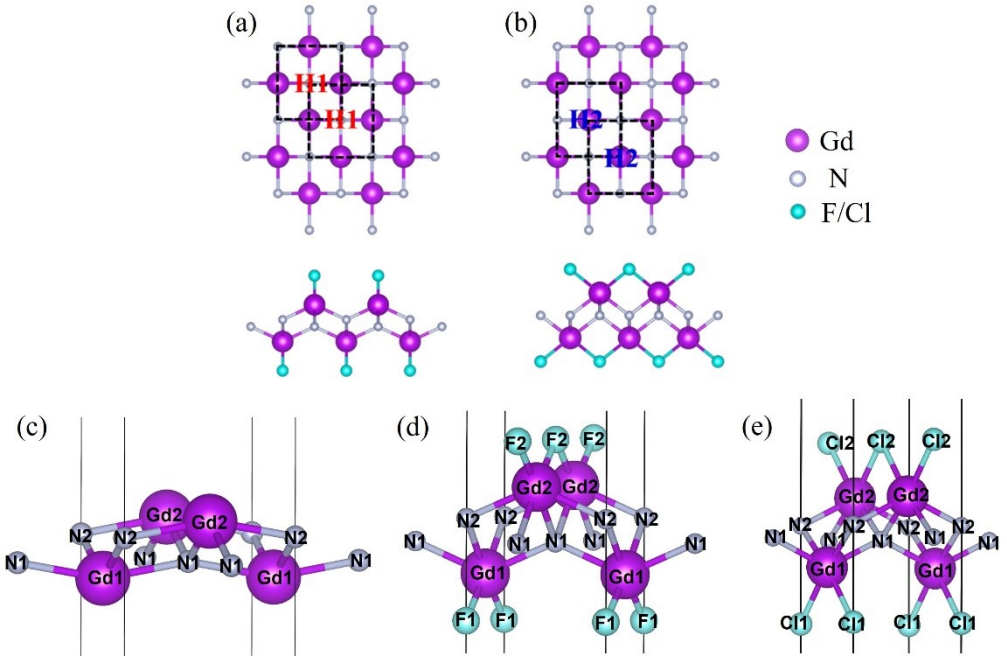

Fig. S1 Top and side views showing two adsorption sites for X (X= F, Cl) atom on the surface of monolayer (ML) GdN, the hollow site in the rectangle consisting of (a) N atoms, named as H1 site, and (b) Gd atoms, named as H2 site. Side views of MLs (c) GdN, (d) H1-GdNF, (e) H1-GdNCl.

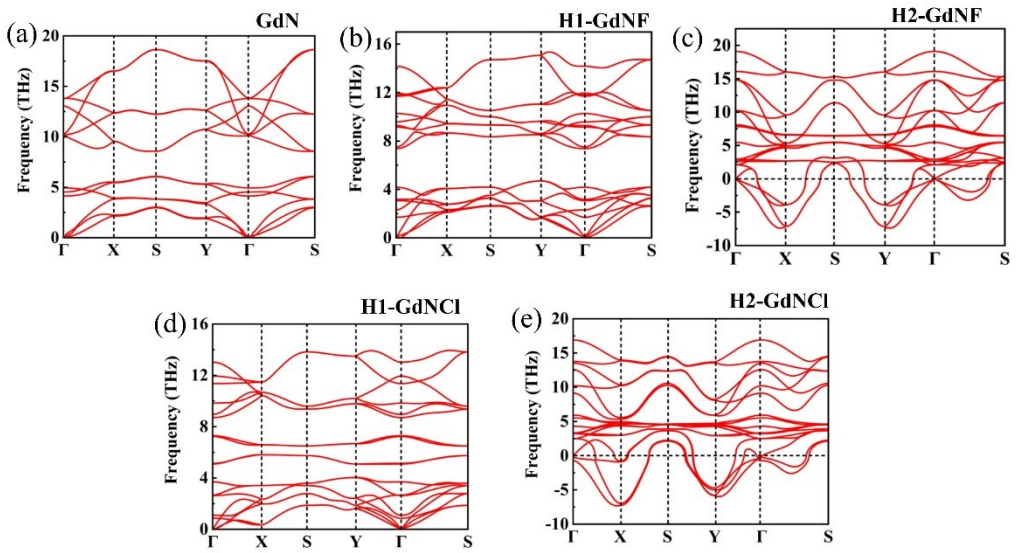

Fig. S2 The phonon dispersion spectra of monolayers (MLs) (a) GdN, (b) H1-GdNF, (c) H2-GdNF, (d) H1-GdNCl, and (e) H2-GdNCl along the high-symmetry direction in the first Brillion Zone.

Table S1 The calculated total energies (meV) for the ferromagnetic (FM) and three antiferromagnetic (AFM) AFM$_i$ (i=1, 2, 3) configurations, elastic constants C_{ij} (N/m) for the most stable magnetic configuration of monolayers (MLs) GdN and H1-GdNX (X= F, Cl). The total energy E of the most stable magnetic configuration is set to 0.

	C_{11}	C_{12}	C_{22}	C_{66}	E_{FM}	E_{AFM1}	E_{AFM2}	E_{AFM3}
GdN	91.53	32.00	128.88	41.78	69.72	0	23.44	26.86
GdNF	150.40	23.80	116.09	12.10	0	158.41	281.74	100.88
GdNCl	72.58	12.03	62.90	13.92	0	63.28	317.96	175.82

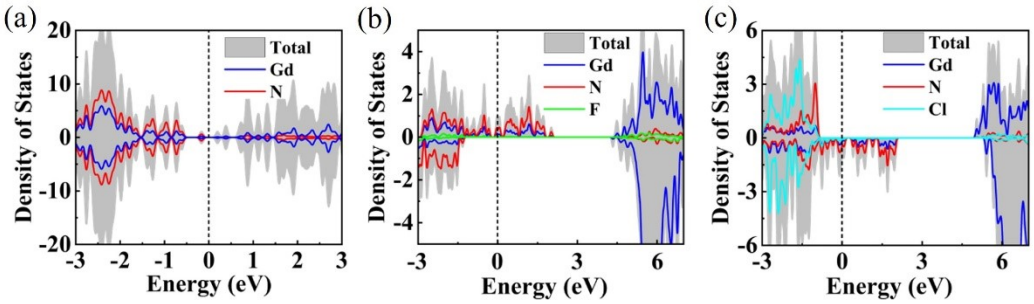

Fig. S3 Density of states of monolayers (MLs) (a) GdN, (b) GdNF and (c) GdNCl.

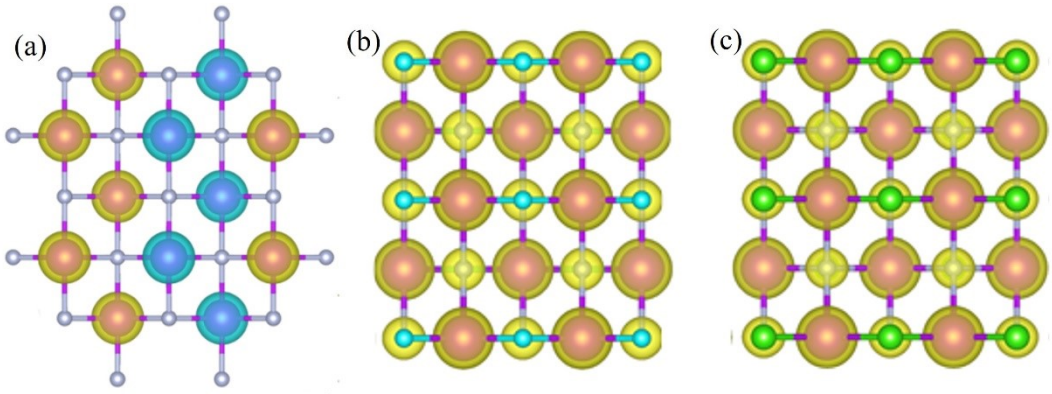

Fig. S4 Spin-resolved charge density of monolayers (MLs) (a) GdN, (b) GdNF and (c) GdNCl.

Table S2 The Mulliken bond overlap population *n* (electrons) of monolayers (MLs) GdN, GdNF, and GdNCl.

Monolayer	Bonds	*n*
GdN	Gd-Gd	-0.29
	Gd-N	0.73
GdNF	Gd-Gd	-0.28
	Gd-N	0.50
	Gd-F	0.39
GdNCl	Gd-Gd	-0.23
	Gd-N	0.51
	Gd-Cl	0.58

Table S3 The lattice constants (Å), bond lengths (Å) and bond angles (°) of monolayers (MLs) GdN, GdNF and GdNCl.

Monolayer	lattice parameters	Bond length	Bond angle
GdN	*a*=3.98 *b*=4.63	*L*(Gd1-N1)=2.35	∠Gd1-N1-Gd1=159.45 ∠Gd2-N1-Gd2=122.03 ∠Gd1-N1-Gd2= 94.96
GdNF	*a*=3.67 *b*=4.24	*L*(Gd1-N1)=2.31 *L*(Gd1-N2)=2.45 *L*(Gd1-F1)=2.28	∠Gd1-N1-Gd1=133.34 ∠Gd2-N1-Gd2= 96.95 ∠Gd1-N1-Gd2=105.22 ∠Gd2-F2-Gd2=107.26

GdNCl	a=3.87 b=4.20	L(Gd1-N1)=2.32 L(Gd1-N2)=2.41 L(Gd1-Cl1)=2.73	∠Gd1-N1-Gd1=129.85 ∠Gd2-N1-Gd2=106.62 ∠Gd1-N1-Gd2=104.67 ∠Gd2-Cl2-Gd2= 90.40

Table S4 The lattice constants (Å) of ferroelastic (FE-I and FE-II) and paraelastic (PE) phases, ferroelastic energy barriers (meV/f.u.), and reversible ferroelastic strain for monolayers (MLs) GdN, GdNF and GdNCl.

Monolayer	Ferroelastic phases				Paraelastic phase		Barrier	Strain
	a_{FE-I}	b_{FE-I}	a_{FE-II}	b_{FE-II}	a_{PE}	b_{PE}		
GdN	3.98	4.63	4.63	3.98	4.38	4.38	49.8	16.3%
GdNF	3.67	4.24	4.24	3.67	3.96	3.96	51.3	15.5%
GdNCl	3.87	4.20	4.20	3.87	4.05	4.05	50.2	8.5%