Electronic Supplementary Material (ESI) for Journal of Materials Chemistry C.

Supporting Information

High-Temperature Quantum Anomalous Hall Effect in Electride Gadolinium Monohalides

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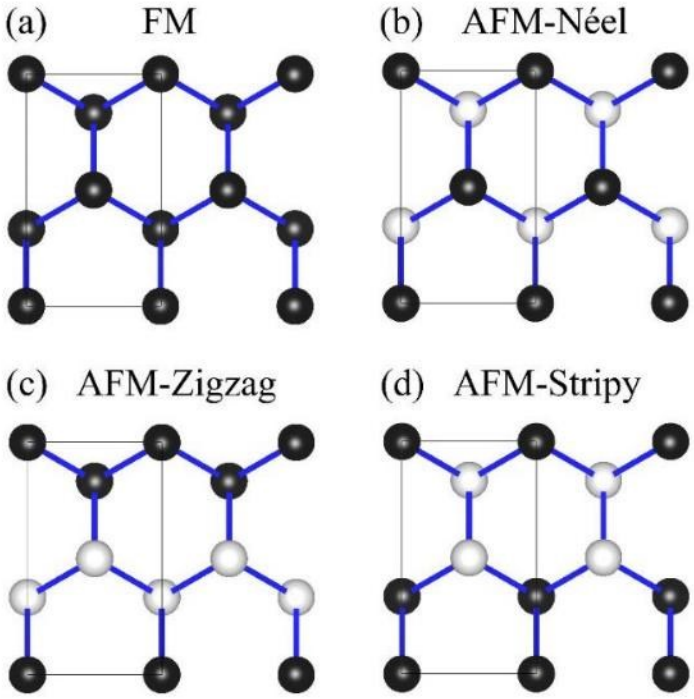


Figure S1. The four types of magnetic configurations for monolayer GdX (X = F, Cl, Br, and I), (a) ferromagnetic (FM), (b) Néel, (c) Zigzag, and (d) Stripy anti-ferromagnetic (AFM). Only the magnetic element Gd is shown, the black and white atoms indicate spin-up and spin-down, respectively.

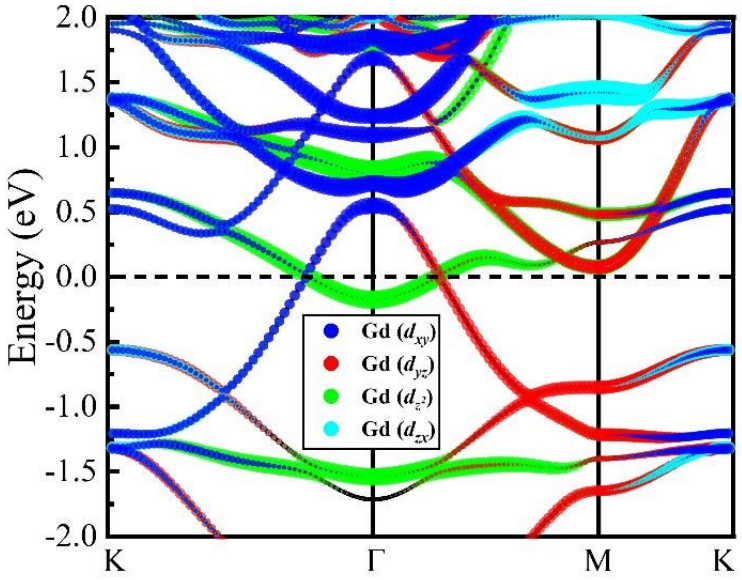


Figure S2. Projected band structure of monolayer GdCl. The blue, red, green, and cyan circles represent the d_{xy}, d_{yz}, d_{z2} and d_{zx} orbitals of Gd atom, the black dashed line at 0 eV indicates the Fermi energy.

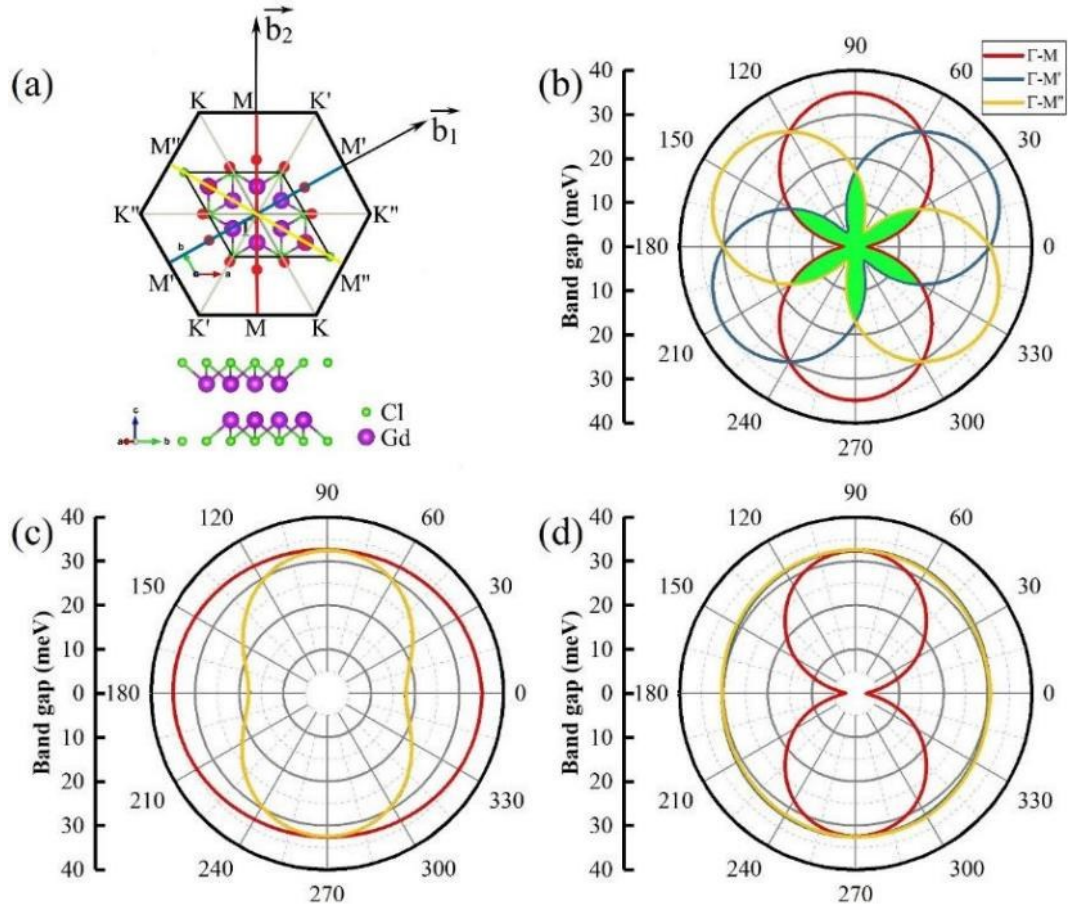


Figure S3. (a) The 2D Brillouin zone of monolayer GdCl structure, where the red dots indicate the position of the Berry curvature peaks in the Brillouin zone for in-plane magnetization. Manipulation of the band gaps by rotating the spin direction within (b) *xoy*, (c) *yoz*, and (d) *zox* planes. The red, green, and yellow lines represent the SOC band gaps opened on the Γ-M, Γ-M', and Γ-M'' paths, respectively. All band gaps are PBE results.

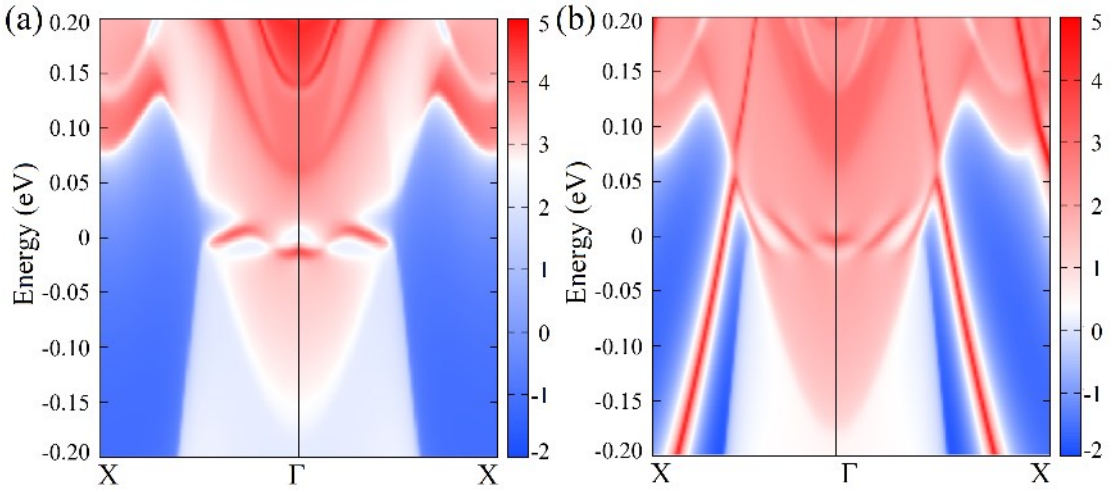


Figure S4. The edge state of monolayer GdCl. (a) and (b) represent the left and right viewing angles of the edge state, respectively.

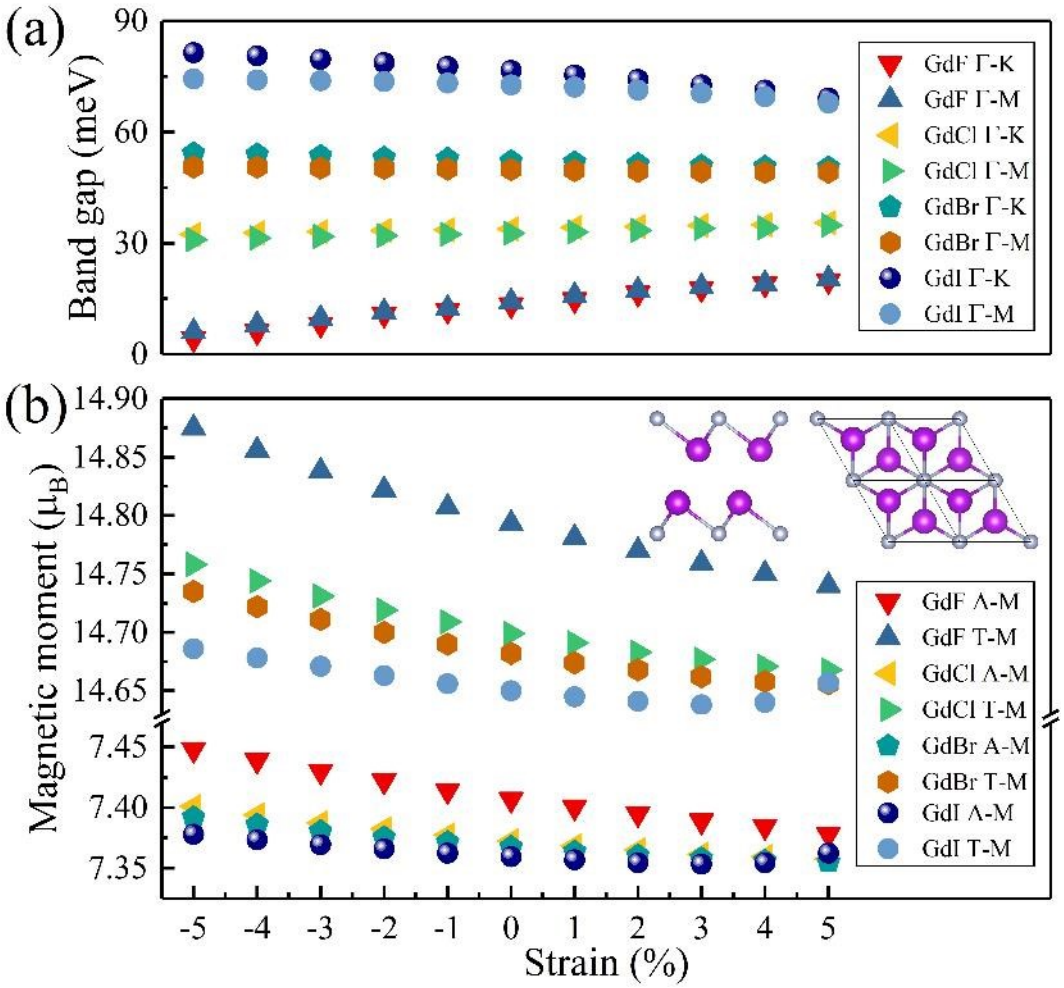


Figure S5. The calculated (a) band gaps and (b) magnetic moments of monolayer GdX under biaxial strain along *x* and *y* direction from -5% to 5%.

Table S1. The lattice constant a and c (Å), magnetic moments of interstitial electrons (μ_B) and total magnetic moment (μ_B), global band gap (meV) for in-plane and out-of-plane magnetizations of bulk GdX. All band gaps are corrected by the mBJ method.

System	Lattice constant a (Å)	Lattice constant c (Å)	Magnetic (μ_B)		Band gap (meV)	
			Interstitial electrons	Total	In-plane	Out-of-plane
GdF	3.718	19.686	0.961	7.443	7.0	7.6
GdCl	3.752	27.629	0.960	7.421	25.6	31.5
GdBr	3.803	29.330	0.956	7.412	36.3	37.5
GdI	3.937	31.212	0.347	7.395	38.5	45.1

Table S2. Total energy of monolayer GdX (X = F, Cl, Br, and I) with different magnetic configurations. The total energy of monolayer GdF, GdCl, GdBr, and GdI for in-plane FM magnetization is lower by about 0.47 meV/f.u., 0.10 meV/f.u., 0.11 meV/f.u., 0.46 meV/f.u. than that for out-of-plane FM magnetization, respectively.

System	FM (eV/f.u.)	Néel (eV/f.u.)	Zigzag (eV/f.u.)	Stripy (eV/f.u.)
GdF	-21.0376	-20.9720	-20.9081	-20.8448
GdCl	-19.2973	-19.2349	-19.1785	-19.0925
GdBr	-18.6728	-18.6082	-18.5619	-18.4825
GdI	-17.9640	-17.8941	-17.8676	-17.7935

Table S3. Total energy of monolayer GdCl of four different magnetic configurations with different Hubbard U.

Hubbard U	FM (eV/f.u.)	Néel (eV/f.u.)	Zigzag (eV/f.u.)	Stripy (eV/f.u.)
0	-19.4683	-19.4193	-19.3781	-19.3234
2	-19.3650	-19.3135	-19.2600	-19.1965
4	-19.3169	-19.2597	-19.2041	-19.1311
6	-19.2973	-19.2349	-19.1785	-19.0925

Table S4. Total energy of monolayer GdCl of four different magnetic configurations under different biaxial strain.

Strain	FM (eV/f.u.)	Néel (eV/f.u.)	Zigzag (eV/f.u.)	Stripy (eV/f.u.)
-4%	-19.2287	-19.1853	-19.1096	-19.0232
-2%	-19.2809	-19.2282	-19.1563	-19.0662
0	-19.2973	-19.2349	-19.1785	-19.0925
2%	-19.2828	-19.2103	-19.1588	-19.0703
4%	-19.2418	-19.1589	-19.1347	-19.0347

Table S5. Total energy of bulk GdX (X = F, Cl, Br, and I) with different magnetic configurations. The total energy of bulk GdF, GdCl, GdBr, and GdI for in-plane FM magnetization is lower by about 0.11 meV/f.u., 0.86 meV/f.u., 0.20 meV/f.u., 0.22 meV/f.u. than that for out-of-plane FM magnetization , respectively.

System	FM (eV/f.u.)	Néel (eV/f.u.)	Zigzag (eV/f.u.)	Stripy (eV/f.u.)
GdF	-21.3061	-21.2440	-21.1929	-21.1059
GdCl	-19.3977	-19.3356	-19.2822	-19.1750
GdBr	-18.7812	-18.7221	-18.6717	-18.5625
GdI	-18.0454	-18.0113	-17.9473	-17.8697

Table S6. The exchange coupling parameters (J_1, J_2, and J_3) and Curie temperature (T_C) for monolayer GdX (X = F, Cl, Br, and I).

System	J_1 (meV)	J_2 (meV)	J_3 (meV)	T_C (K)
GdF	1.194	1.189	-0.384	650
GdCl	1.386	1.220	-0.609	510
GdBr	1.347	1.106	-0.542	530
GdI	1.350	0.924	-0.477	560

The calculations details of Currie Temperature

Full Hamiltonian

$$H = -2J_1\sum_{\langle i,j\rangle} S_i \cdot S_j - 2J_2\sum_{\langle\langle i,j\rangle\rangle} S_i \cdot S_j - 2J_3\sum_{\langle\langle\langle i,j\rangle\rangle\rangle} S_i \cdot S_j - D\sum_i \left|S_i^z\right|^2 - 2\lambda\sum_{\langle i,j\rangle} S_i^z \cdot S_j^z$$

$\langle i,j\rangle$, $\langle\langle i,j\rangle\rangle$, and $\langle\langle\langle i,j\rangle\rangle\rangle$ denotes the nearest, second, and third neighbor spins.

$J_{1,2,3}$ are the Heisenberg exchange constants between nearest, second, and third neighbor spins.

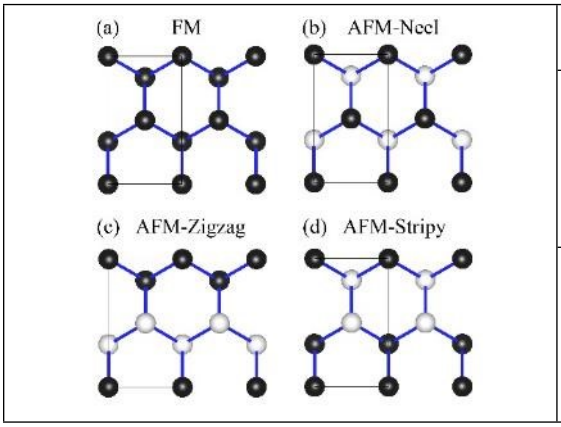
For Cubic lattice: $N_1 = N_2 = N_3 = 4$

For Hexagonal/Honeycomb lattice: $N_1 = 3, N_2 = 6, N_3 = 3$

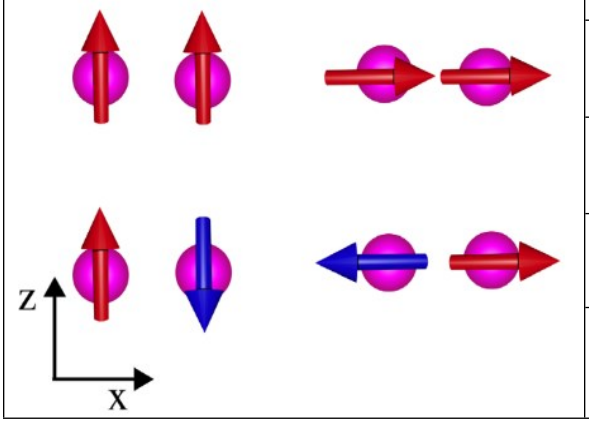
For Triangular lattice: $N_1 = N_2 = N_3 = 6$

$|\vec{S}|^2$: S is spin quantum number, 1/2 of the magnetic moment.

Heisenberg Exchange Constant

	Hexagonal example (if the summation of spins are isotropy)[1]
	$E_{FM/Néel} = -E_0 - (\pm 3J_1 + 6J_2 \pm 3J_3)\|\vec{S}\|^2$
	$E_{Zigzag/Stripy} = -E_0 - (\pm J_1 - 2J_2 \mp 3J_3)\|\vec{S}\|^2$

Single Ion Anisotropy + Anisotropy Symmetric Exchange[2]

	Hexagonal example (just considering the nearest neighbors)
	$E_{FM,z} = -E_0 - D\|\vec{S}\|^2 - 3(J+\lambda)\|\vec{S}\|^2$
	$E_{AFM,z} = -E_0 - D\|\vec{S}\|^2 + 3(J+\lambda)\|\vec{S}\|^2$
	$E_{FM,x} = -E_0 - 3J\|\vec{S}\|^2$
	$E_{AFM,x} = -E_0 + 3J\|\vec{S}\|^2$

Reference:

1. N. Sivadas, M. W. Daniels, R. H. Swendsen, S. Okamoto and D. Xiao, *Phys. Rev. B*, 2015, **91**.
2. J. L. Lado, Fernández-Rossier, Joaquín, *2D Materials*, 2017, **4**, 035002.