Electronic Supplementary Material (ESI) for Physical Chemistry Chemical Physics.

Supporting Information for

Monolayers gadolinium halides GdX_2 (X = F, Cl, Br): intrinsic ferrovalley materials with spontaneous spin and valley polarizations

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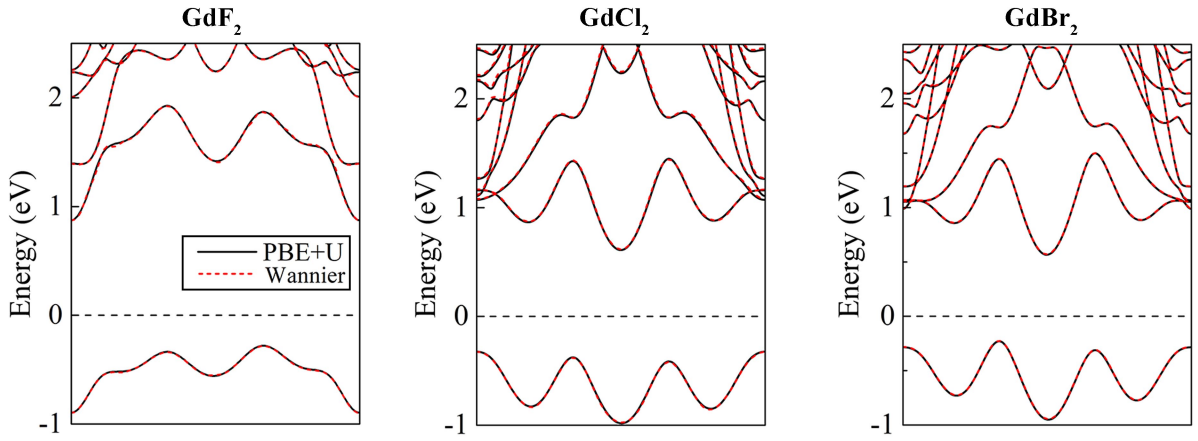

Figure S1: Comparison of the band structures of MLs GdX_2 obtained by means of the PBE+U and Wannier methods in the presence of the SOC effect.

Table S1: Calculated Elastic Constants in N/m for MLs GdX_2.

Monolayer	C_{11}	C_{12}	C_{22}	C_{44}
GdF_2	70.55	18.64	70.55	26.07
$GdCl_2$	46.07	14.20	46.07	15.93
$GdBr_2$	40.90	12.56	40.90	14.17

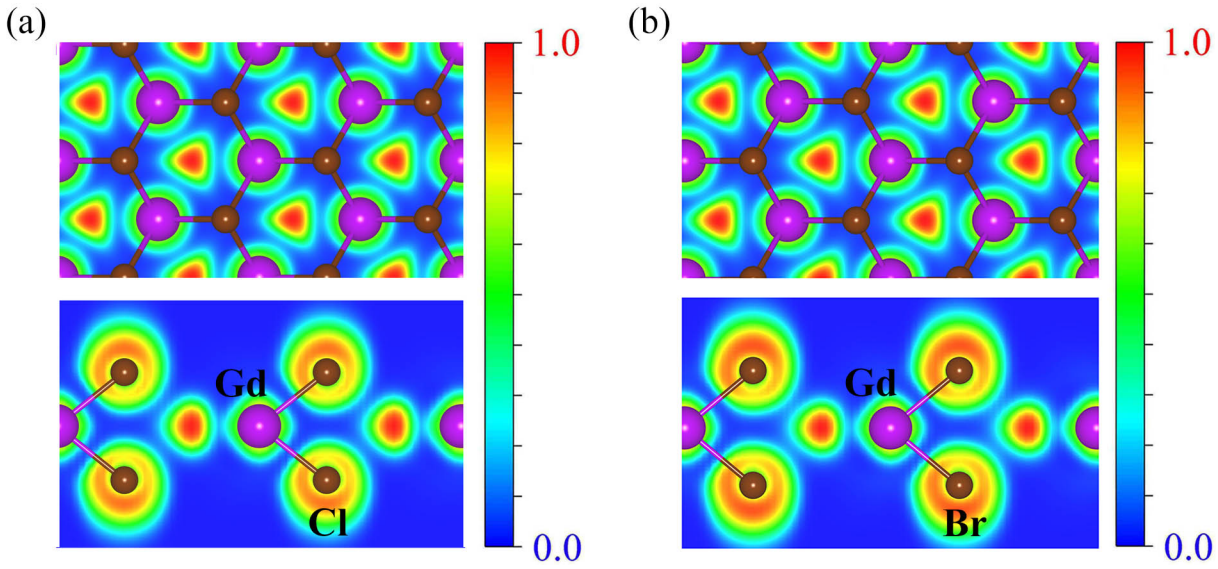

Figure S2: Top and side views of the ELF for MLs (a) $GdCl_2$ and (b) $GdBr_2$. The values of 1.0 and 0.0 indicate the electron completely localized and vanishing regions, respectively.

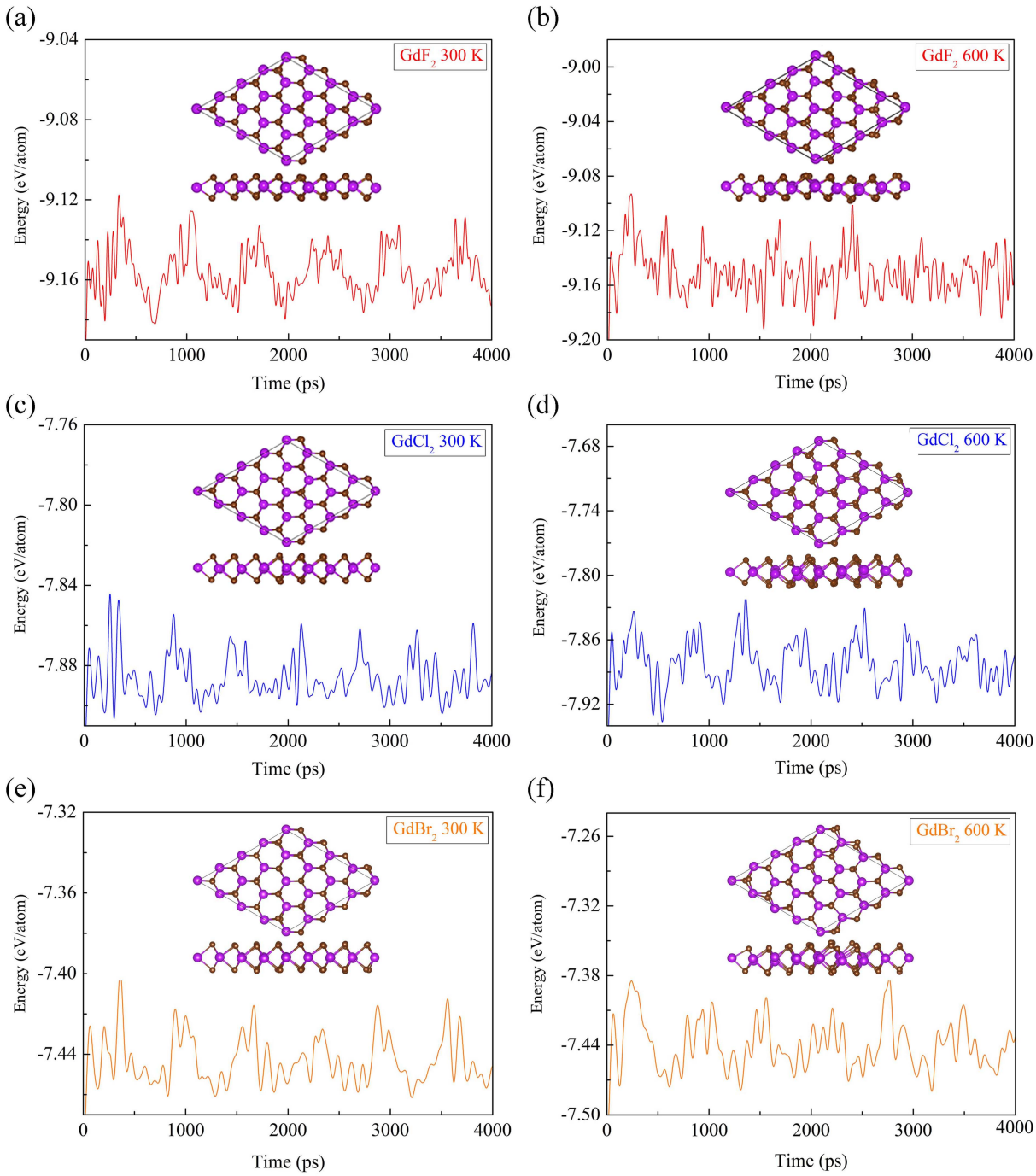

Figure S3: Evolution of potential energy in eV/atom for MLs GdX_2 during the AIMD simulations at 300 and 600 K for 4 ps. The insets in all cases refer to the snapshot of final configuration at the end of the AIMD simulations.

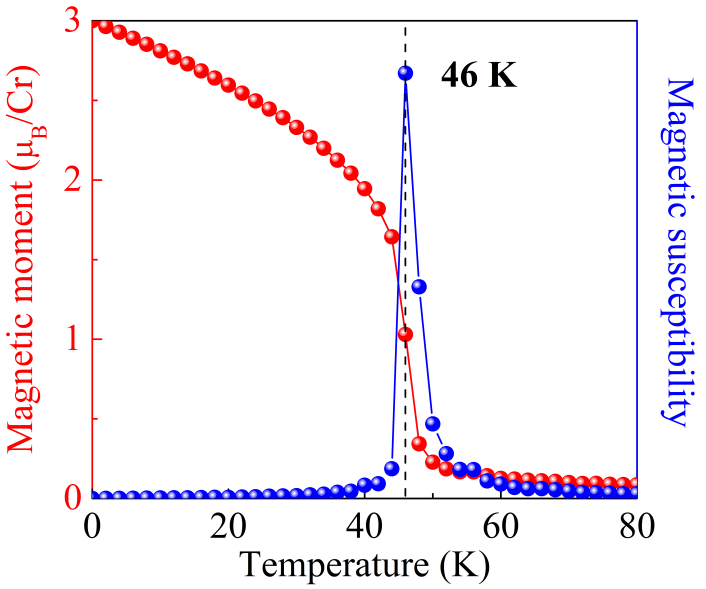

Figure S4: Average magnetic moment per Cr atom and magnetic susceptibility as a function of temperature for ML CrI_3 from MC simulations (J =3.90 meV, A =0.35 meV, and $|\boldsymbol{S}|$ =3/2). Our estimated Curie temperature is 46 K, in excellent agreement with the experimental measurement of 45 K.

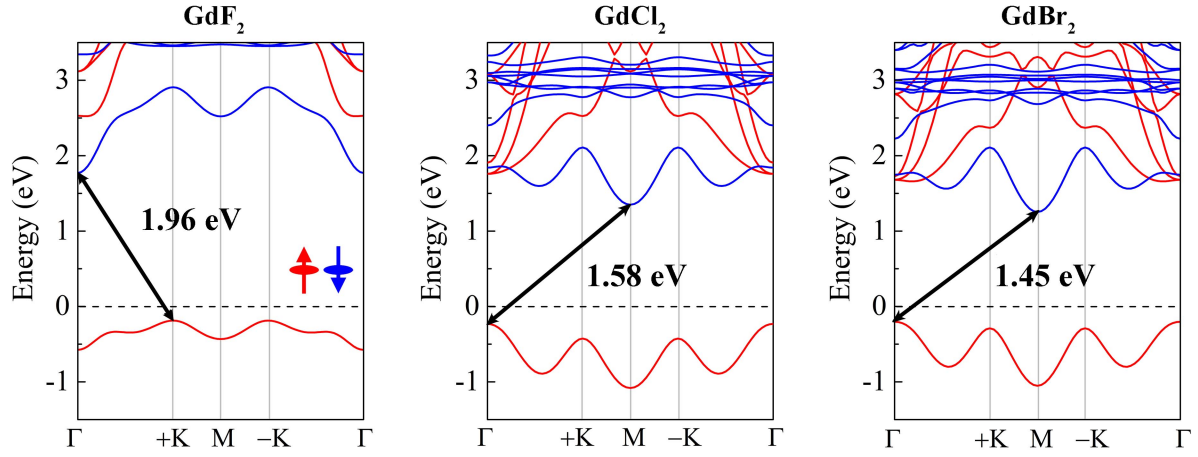

Figure S5: Spin-resolved band structures of MLs GdX_2 in the absence of the SOC effect in the FM ground state at the HSE06 level. The Fermi level is set to zero in all cases.

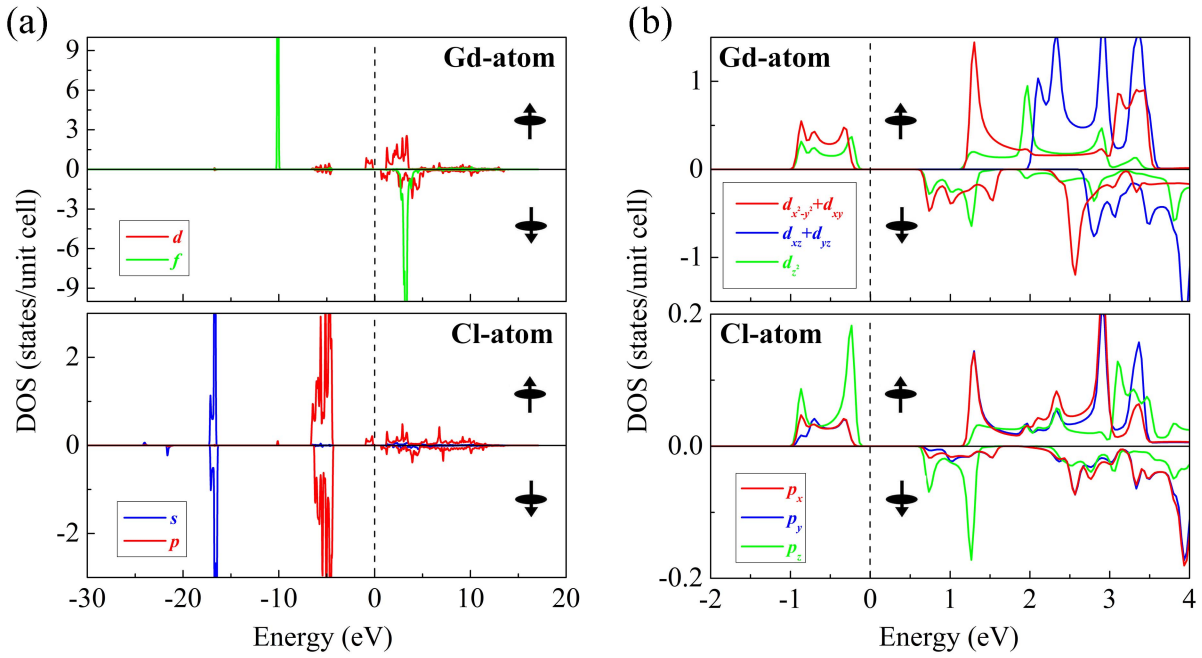

Figure S6: Orbital-resolved DOSs in the presence of spin polarization for ML $GdCl_2$ in the FM ground state at the PBE+U level. The Fermi level is set to zero in all panels.

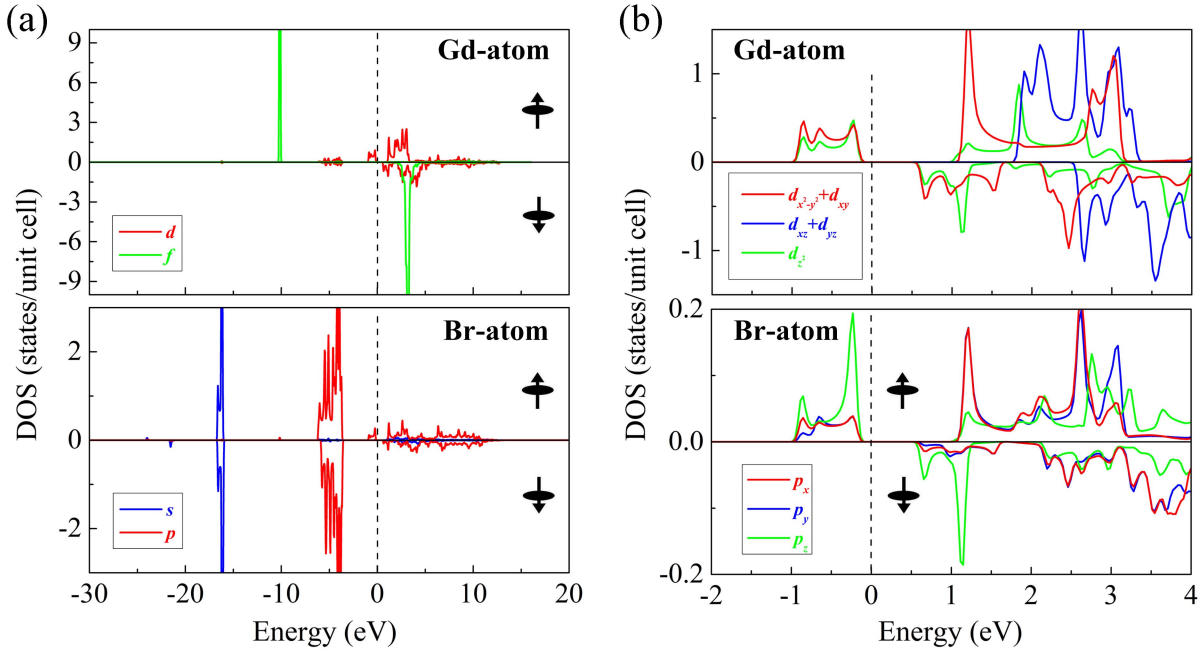

Figure S7: Orbital-resolved DOSs in the presence of spin polarization for ML $GdBr_2$ in the FM ground state at the PBE+U level. The Fermi level is set to zero in all panels.

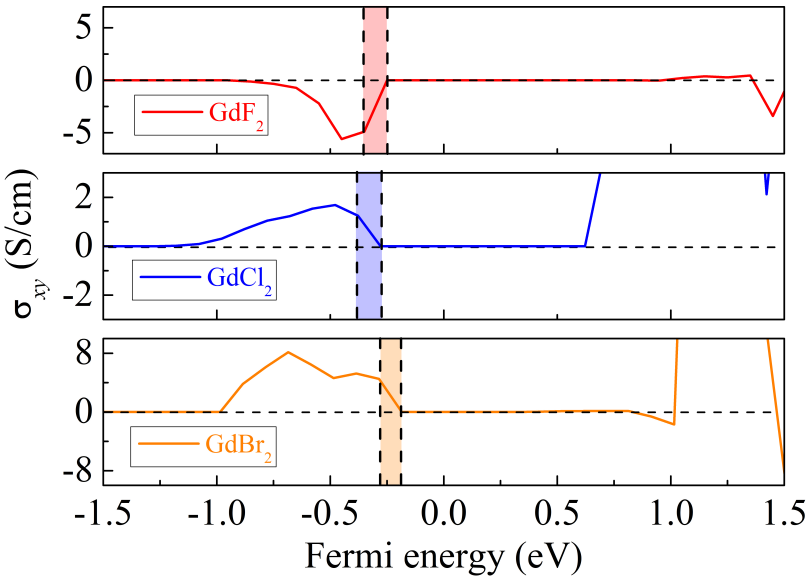

Figure S8: Calculated anomalous Hall conductivities of MLs GdX_2 with respect to the Fermi energy. The two vertical dashed lines in each panel refer to the energy extrema of two valleys, respectively.