Electronic Supporting Information: Rational Design of Two-Dimensional High-Temperature Ferromagnet from HCP Cobalt

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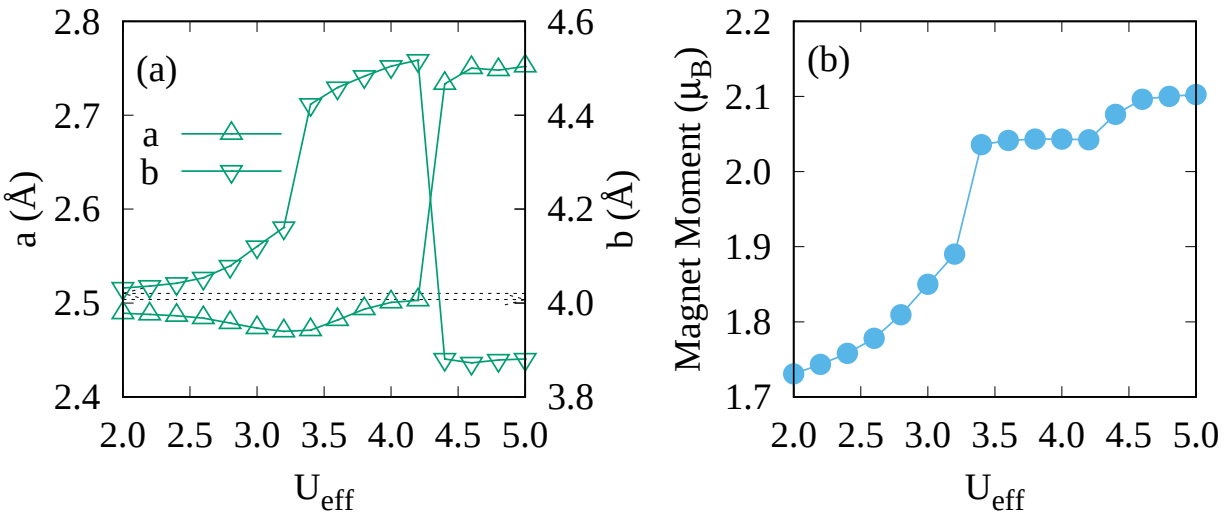

FIG. S1: (Color online). (a) The lattice constants ($\vec{a}$ and $\vec{c}$) and (b) the magnetic moment (μ_B) per Co atom as a function of U_{eff} for HCP Co.

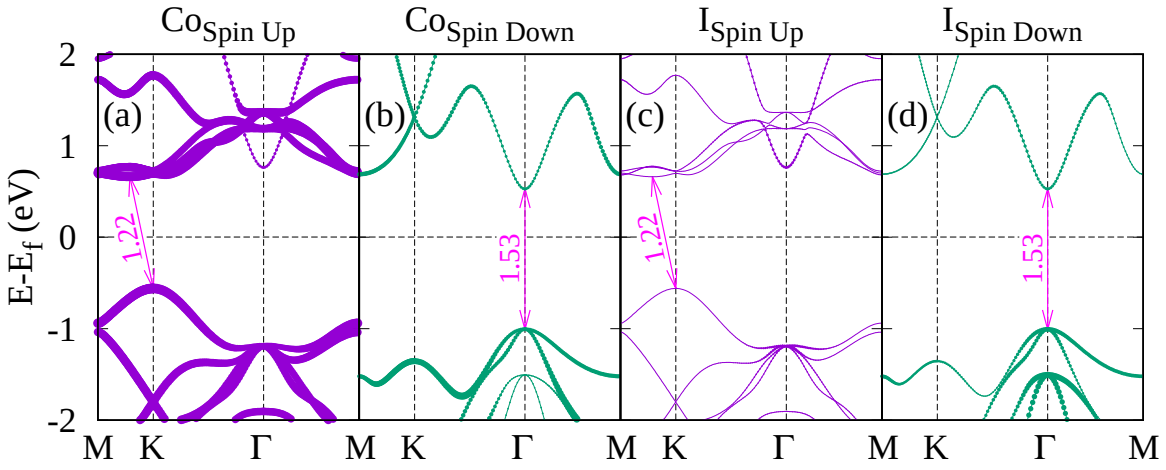

FIG. S2: (Color online). The spin/atom-projected band structures of two-dimensional Co_2I_2 at the PBE+U (U_{eff}=3.2 eV) level of theory. (a) Co for spin up; (b) Co for spin down; (c) I for spin up; (d) I for spin down.

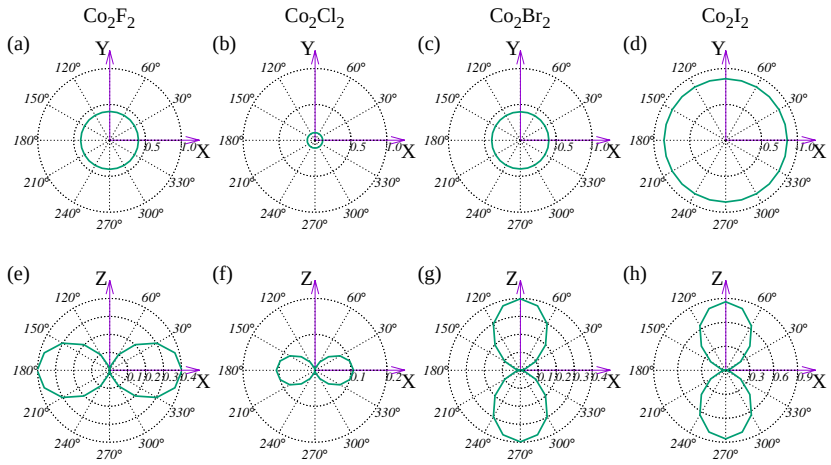

FIG. S3: (Color online). The magnetocrystalline anisotropy energy per Co atom (E_{MAE}, in meV) as a function of the magnetization direction in the XY (a-d) and XZ (e-h) plane at the PBE+U (U_{eff}=3.2 eV) level of theory, respectively. The energy of the configuration with the magnetic moment pointing to (001) for the XY plane or (100) for the XZ plane is set as 0 eV. (a, e) Co_2F_2, (b, f) Co_2Cl_2, (c, g) Co_2Br_2 and (d, h) Co_2I_2.

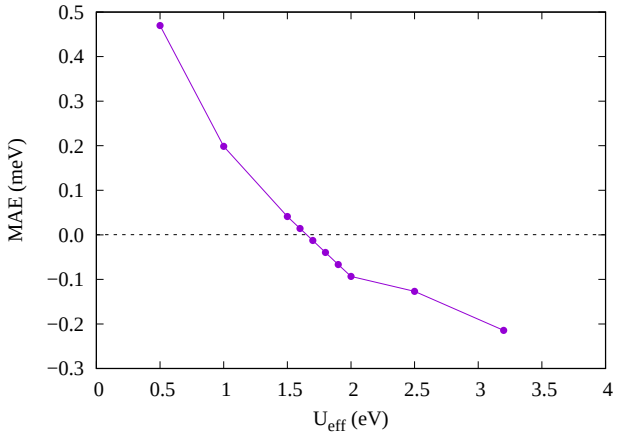

FIG. S4: (Color online). The magnetocrystalline anisotropy energy per Co atom ($E_{MAE} = E_{(001)} - E_{(100)}$, in meV) as a function of the effective on-site Coulomb interaction (U_{eff}) for Co_2Cl_2 at the PBE+U level of theory.

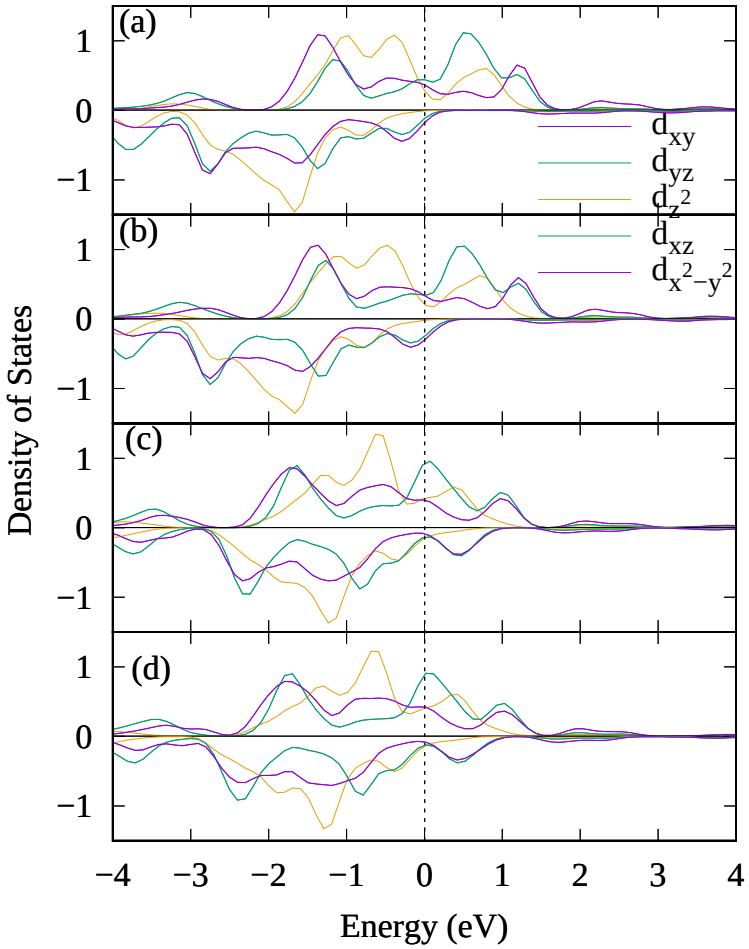

FIG. S5: (Color online). The orbital-projected density of states of Co_2I_2 as a function of the in-plane biaxial strain. (a) -2%, (b) -4%, (c) -6% and (d) -8%.

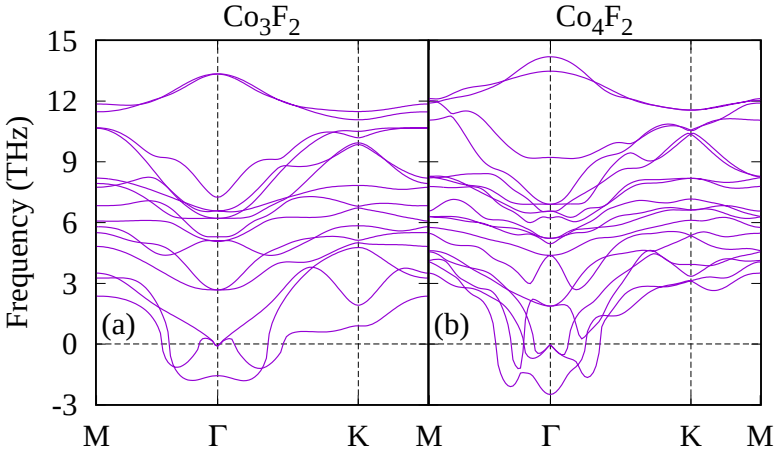

FIG. S6: (Color online). Phonon spectra of free-standing (a) Co_3F_2 and (b) Co_4F_2 calculated at the PBE level of theory.

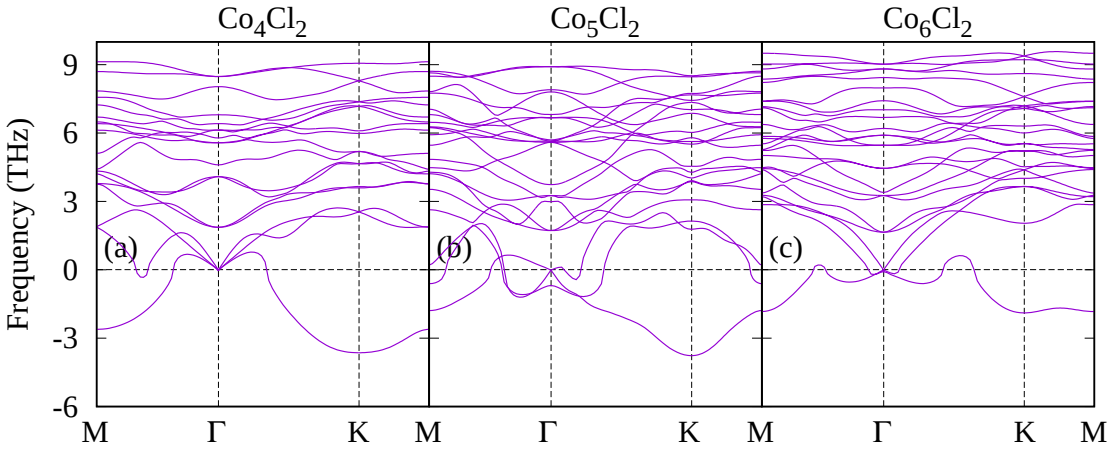

FIG. S7: (Color online). Phonon spectra of free-standing $Co_{2n}Cl_2$ calculated at the PBE level of theory. (a) Co_4Cl_2, (b) Co_5Cl_2 and (c) Co_6Cl_2.

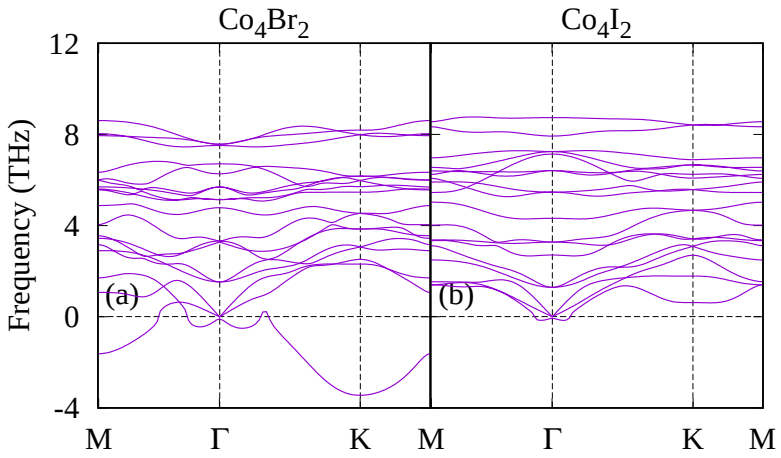

FIG. S8: (Color online). Phonon spectra of free-standing (a) Co_4Br_2 and (b) Co_4I_2 calculated at the PBE level of theory.