Chromium chalcohalide Janus monolayer ferromagnets with perpendicular magnetic anisotropy and high Curie temperature

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Using density functional theory (DFT), we revisit magnetic properties of a recently proposed family of noncentrosymmetric 2D van der Waals materials, chromium chalcohalide monolayers (MLs), CrXY (X = S, Se, Te; Y = Cl, Br, I). They comprise three atomic planes stacked in the X–Cr–Y sequence, which breaks inversion symmetry, giving rise to their designation as Janus MLs. Our total-energy calculations reveal that all dynamically stable CrXY MLs exhibit ferromagnetic (FM) coupling. However, robust perpendicular magnetic anisotropy (PMA) is found only in the CrSI and CrSeI compositions, for both 1T and 1H structures. The PMA results from a constructive interplay between single-ion and anisotropic exchange contributions that overcome the dipole–dipole interaction. By contrast, all other CrXY MLs display easy-plane-like behavior. Quantification of the Dzyaloshinskii-Moriya interaction (DMI) in CrSI and CrSeI for both polymorphs reveals weak-to-moderate strengths relative to isotropic exchange. Atomistic spin dynamics simulations, performed using DFT-derived parameters, predict Curie temperatures of at least 210 K for 1T-CrSI, 235–260 K for 1H-CrSeI, and 370–410 K for 1H-CrSI. In contrast, the sizable DMI in 1T-CrSeI induces a worm-like domain ground state at zero field and stabilizes skyrmions under a perpendicular magnetic field.

References:

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