Highlights

RESEARCH AREA 2 – Functional and Complex Materials for Innovative Electronics and Sensing - 2024

Delving into the anisotropic interlayer exchange in bilayer CrI₃

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Bilayer CrI_3 attracted much attention due to stacking-induced switching between the layered ferromagnetic and antiferromagnetic order. This discovery brought under the spotlight the interlayer Cr–Cr exchange interaction, which despite being much weaker than the intralayer exchange, plays an important role in shaping the magnetic properties of bilayer CrI_3. In this work we delve into the anisotropic part of the interlayer exchange with the aim to separate the contributions from the Dzyaloshinskii–Moriya (DMI) and the Kitaev interactions (KI). We leverage the density functional theory calculations with spin Hamiltonian modeling and develop an energy mapping procedure to assess these anisotropic interactions with μeV accuracy. After inspecting the rhombohedral and monoclinic stacking sequences of bilayer CrI_3, representative of low- (LT) and high-temperature (HT) structures respectively, we reveal a considerable DMI and a weak interlayer KI between the sublattices of a monoclinic bilayer. We explain the dependence of DMI and KI on the interlayer distance, stacking sequence, and the spin–orbit coupling (SOC) strength, and we suggest the dominant super-exchange processes at play. In addition, we demonstrate that the single-ion anisotropy in bilayer CrI_3 is highly stacking-dependent, increasing by 50% from monoclinic to rhombohedral bilayer. Remarkably, our findings prove that iodines are highly efficient in mediating the DMI across the van der Waals gap, much owing to spatially extended 5p orbitals which feature strong SOC. Our study gives promise that the interlayer chiral control of spin textures, demonstrated in thin metallic films where the DMI is with a much longer range, can be achieved with similar efficiency in semiconducting two-dimensional van der Waals magnets.

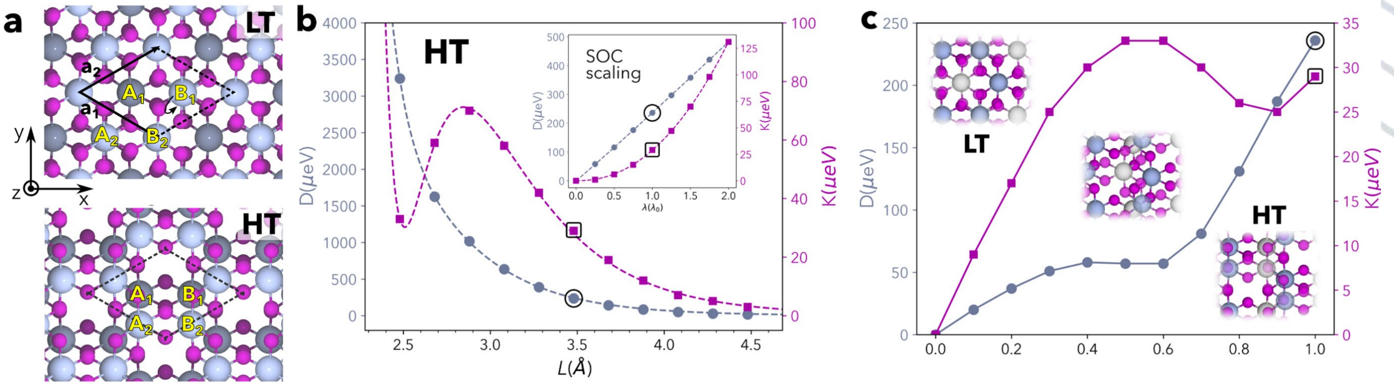

Figure a Top view of bilayer CrI3 with rhombohedral (LT) and monoclinic (HT) stacking. Atoms from the upper layer are colored by brighter nuances. The HT structure is obtained from LT when the upper layer is translated by $\mathbf{a}_2/3$. **b** Evolution of the magnitude of DMI and KI coupling strengths (D and K, respectively) with interlayer distance in the HT stacking; dashed lines are obtained by fitting on a general functional form describing super-super-exchange processes mediated by 5p iodine states. The D and K dependence on SOC constant is shown in the inset. **c** Evolution of the DMI and KI coupling strengths along an adiabatic path connecting the LT and HT stacking sequences. To make navigation through panels easier, the common points of the plots are denoted with ◯ (DMI) and □ (KI).