Surface-Bulk Difference in van der Waals Magnets

L. Zhao[1,+]

[1] *Department of Physics, University of Michigan, Ann Arbor, MI, USA*

The successful isolation of monolayer to few-layer magnetic atomic crystals from van der Waals (vdW) magnets have opened a new pathway of researching two-dimensional (2D) magnetism [1,2,3]. Over the past half a decade, the vdW and 2D magnet library has been greatly expanded, and new magnetic phenomena have been discovered in the 2D limit. Yet, one key question has been brought up: what is the distinction amongst bulk, surface and 2D magnetism for a vdW magnet? This question is well motivated by the observations of 2D behaviors in 3D vdW magnets, as well as the contrasts between 2D layers and 3D bulk, for systems such as CrI_3, CrSBr, $NiPS_3$, etc.

In this talk, we will show the surface-bulk difference in two archetype vdW magnets, CrI_3 [4] and CrSBr [5]. In CrI_3, it has been thought that the 3D bulk hosts the ferromagnetic (FM) state below T_c = 61K whereas the 2D films realizes the layered antiferromagnetic (AFM) order below T_N = 45K. We will show from our optical magneto-Raman spectroscopy measurements that even in a 3D bulk CrI_3, we capture clear signatures of layered AFM, in addition to the known bulk FM. We attribute the layered AFM signature here to the surface magnetism, which is the same as that of the 2D layers but distinct from that deep in the 3D bulk (Figure 1a). In CrSBr, it has the same layered AFM order in both 3D bulk and 2D layers, but surprisingly with a higher critical temperature in the 2D case. We will show with our nonlinear optical measurements that multiple characteristic temperature scales appear in the 3D bulk CrSBr, including a surface ($T_{surface}$) and a bulk (T_{bulk}) onset temperature between which the surface one is unexpectedly higher than the bulk one (Figure 1b). Our results on these two systems demonstrate that the surface of vdW magnets can well be distinct from their bulk.

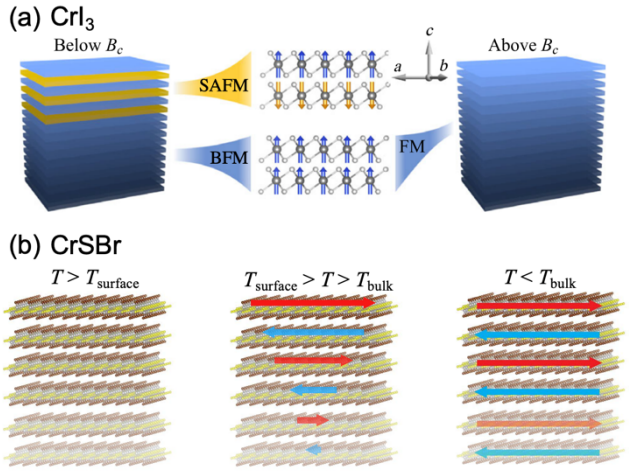

Figure 1 (a) Diagram to show the surface layered antiferromagnetism (SAFM) and bulk ferromagnetism (BFM) in the 3D bulk CrI_3; (b) Diagram to show the separation of surface and bulk magnetic critical temperatures in the 3D bulk CrSBr.

[1] Cheng *et al* Nature 546, 265 (2017)
[2] Huang *et al* Nature 546, 270 (2017)
[3] Wang *et al* ACS Nano, 16, 6960 (2022)
[4] Li *et al* Phys. Rev. X, 10, 011075 (2020)
[5] Guo *et al* arXiv 2309.01047 (2023)

[+] Author for correspondence: lyzhao@umich.edu