

Imaging and Writing Chiral Antiferromagnetic Domains in the 2D Triangular Antiferromagnet $\text{Co}_{1/3}\text{NbS}_2$

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The family of intercalated niobium and tantalum dichalcogenides, $\text{M}_{1/3}\text{NbS}_2$ and $\text{M}_{1/3}\text{TaS}_2$ (where the $3d$ magnetic atom $\text{M} = \text{V}, \text{Cr}, \text{Mn}, \text{Fe}, \text{Co}, \text{Ni}$), are van der Waals materials that host a wide range of fascinating magnetic properties. $\text{Co}_{1/3}\text{NbS}_2$ is an antiferromagnet (AFM) that features layers of Co spins on a 2D triangular lattice (see Fig.1) -- an archetypal frustrated network that can lead to complex magnetic topologies. Despite its vanishing net magnetization, $\text{Co}_{1/3}\text{NbS}_2$ was recently shown to exhibit a giant anomalous Hall effect [1], suggesting a nontrivial AFM order and potential for AFM electronic and spintronic devices. Recent neutron diffraction studies point to a non-coplanar 3Q AFM order with scalar spin chirality [2]. In contrast to conventional collinear AFM order, this (and certain other) complex AFM spin configurations can allow for off-diagonal elements the conductivity, σ_{xy} , which in turn generates anomalous and topological Hall effects in transport studies.

Crucially, $\sigma_{xy}(\omega)$ are frequency-dependent, and at optical frequencies they generate Kerr rotation and magnetic circular dichroism (MCD). Thus, the full power of optical methods, including spectroscopy and spatially-resolved imaging, can now be applied to investigate the complex AFM order in $\text{Co}_{1/3}\text{NbS}_2$. Here we show [3], using light spanning infrared to ultraviolet (1-3 eV), that MCD is a powerful and incisive probe of chiral 3Q AFM order in these van der Waals magnets. Measurements at different photon energies are compared with DFT calculations. Scanning MCD microscopy is used to directly image chiral AFM domains, and also, to demonstrate optical writing of chiral 3Q domains. These studies suggest routes to AFM spintronic devices based on $\text{Co}_{1/3}\text{NbS}_2$ and related 2D magnets.

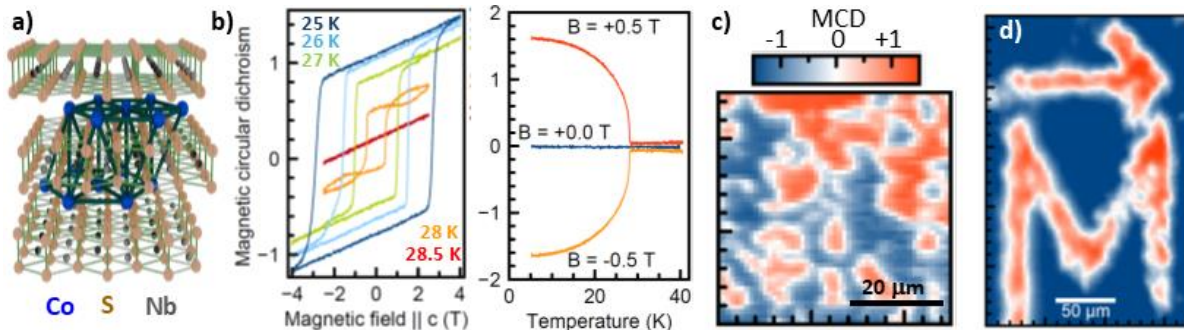


Fig. 1 a) Structure of $\text{Co}_{1/3}\text{NbS}_2$, with 2D triangular lattices of Co spins. b) MCD vs. B, T shows onset of giant MCD and hysteretic AFM switching below the AFM ordering temperature $T_{\text{Neel}} \approx 28.5$ K. c) Direct optical imaging of chiral AFM domains. d) Optical writing of chiral AFM domains.

[1] N.J. Ghimire *et al.*, Nat. Comm. **9**, 3280 (2018); G. Tenasini *et al.*, Phys. Rev. Res. **2**, 023051 (2020).

[2] H. Takagi *et al.*, Nat. Phys. **19**, 961 (2023).

[3] E. Kirstein *et al.*, submitted.

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