Van der Waals Antiferromagnets $Co_{1/3}NbS_2$ and $Co_{1/3}TaS_2$: Topological Magneto-Optics & Tunable Chiral/Nematic Phases

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The family of intercalated niobium and tantalum dichalcogenides, $M_{1/3}NbS_2$ and $M_{1/3}TaS_2$ (where M= V, Cr, Mn, Fe, Co, Ni), are van der Waals materials hosting layers of spins on 2D triangular lattices -- an archetypal frustrated network that can lead to a rich variety of complex magnetic states. $Co_{1/3}NbS_2$ and $Co_{1/3}TaS_2$ are antiferromagnets (AFMs) that exhibit giant spontaneous Hall conductivity despite vanishing net magnetization [1], suggesting nontrivial AFM order and potential for AFM spintronics. Recent neutron diffraction studies point to a non-coplanar "tetrahedral" triple-**Q** AFM order with scalar spin chirality [1]. In contrast to conventional collinear AFM order, this (and certain other) complex AFM spin configurations can allow for off-diagonal Hall conductivity, σ_{xy}, which in turn generates anomalous and topological Hall effects in transport studies.

Crucially, $\sigma_{xy}(\omega)$ is frequency-dependent, and at optical frequencies it generates Kerr rotation and magnetic circular dichroism (MCD). Thus, the full power of optics, including imaging and spectroscopy, can be applied to study complex AFM orders in $Co_{1/3}NbS_2$ and $Co_{1/3}TaS_2$. Here we show [2,3], using light spanning infrared-ultraviolet (1-3 eV), that MCD is a powerful and incisive probe of chiral triple-**Q** AFM order. Measurements at different photon energies are compared with DFT calculations. Scanning MCD microscopy is used to directly image (and also write) chiral 3**Q** domains. In $Co_{1/3}TaS_2$, linear dichroism studies also reveal three-state (Z_3) nematicity arising from a single-**Q** stripe phase, as well as phases where chirality *and* nematicity coexist [3]. A theoretical analysis based on a *continuous multi-**Q** manifold* captures the emergence of these distinct magnetic phases, resulting from the interplay between four-spin interactions and weak magnetic anisotropy. Our findings establish $Co_{1/3}TaS_2$ as a rare platform hosting diverse multi-**Q** states with distinct combinations of spin chirality and nematicity).

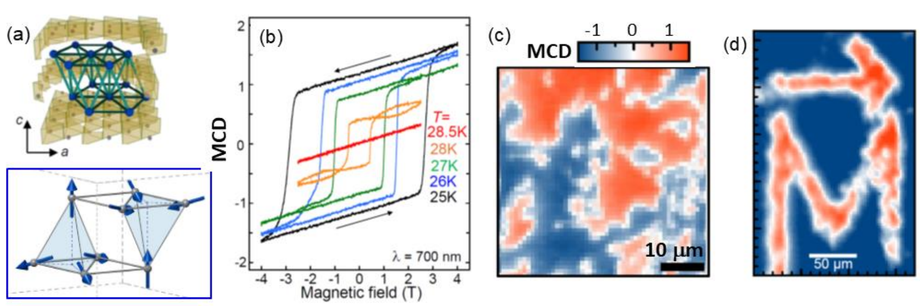

Fig. 1 a) Structure of $Co_{1/3}(Nb,Ta)S_2$, with triple-**Q** 'tetrahedral' AFM order. b) MCD vs. B shows hysteretic AFM switching below $T_N \approx 28.5$ K. c,d) Optical imaging & writing of chiral AFM domains.

[1] N.J. Ghimire *et al*, Nat. Comm. **9**, 3280 (2018); H. Takagi *et al*, Nat. Phys. **19**, 961 (2023).
[2] E. Kirstein *et al*, *arXiv:2507.18829 (in press, Phys Rev. Lett.);* E. Kirstein, *arXiv:2507.08148*

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