Manipulating the Spin Orientation of VdW Materials

The spin orientation of $CrPS_4$, a candidate magnetic 2D van der Waals (vdW) material, can be manipulated through changes in temperature, magnetic field, and hydrostatic pressure.

Research on atomically thin materials (ATMs) has emerged after the discovery of graphene, the first ATM produced in a laboratory; its discoverers were awarded the 2010 Nobel Prize in Physics. ATMs exhibit unique physical properties, and their atomically thin sheet geometry is favored for use in the semiconductor industry. Thus, experts regard ATMs as promising candidate materials for next-generation semiconductors, spintronic devices, rechargeable batteries, and quantum and optoelectronic devices. Transition metal chalcogenides (TMCs), such as MoS_2, WSe_2, $MoTe_2$, and SnS, are ATMs with well-researched electrical and optical properties. However, due to their nonmagnetic properties, these materials lack spintronic applications. Conversely, many of the ternary transition metal chalcogenides (TTMCs), such as $CrSiTe_3$, $FePS_3$, and $CrPS_4$, are magnetic, and they undergo a magnetic phase transition at low temperatures. Therefore, TTMCs are well-suited for spintronic applications.

Due to the weak van der Waals (vdW) gaps between sulfur layers in $CrPS_4$, the traditional method of mechanical cleavage can be employed to easily isolate thin flakes. Researchers have prepared and studied single-layer and multiple-layer $CrPS_4$ samples. Wenyun Yang (Peking University, China) and collaborators investigated the physical properties of the TTMC $CrPS_4$. Yang and his team manipulated the spin orientation of $CrPS_4$ with externally controllable parameters, such as temperature, magnetic field, and hydrostatic pressure using neutron powder diffraction (NPD) at **ECHIDNA**, X-ray powder diffraction, magnetometry, and magnetic torque measurements.[1,2] Without a magnetic field, $CrPS_4$ exhibits an antiferromagnetic (AFM) structure in which the ferromagnetic layers are coupled antiferromagnetically along the *c*-axis. The magnetic anisotropy is temperature-dependent; it shifts from in-plane anisotropy (//b) in the vicinity of the T_N (Néel temperature) to the ground state and then to out-of-plane anisotropy (in the *ac*-plane and mainly along the *c*-axis) upon cooling. The magnetic field can be used to tune the magnetic structure, as illustrated in **Fig. 1**. After the magnetic field is applied, the AFM magnetic structure first undergoes a spin-flop transition to a canted AFM (cAFM) state with a net magnetization in the *ab*-plane. A further increase in the magnetic field causes the cAFM to undergo a spin-flip transition to the ferromagnetic state, where the spin orientation flips again to be along the *c*-axis.

Existing experimental evidence also indicates that the transition of spin reorientation from the *ac*-plane to the *b*-axis is first-order in nature. T_{SR}, the characteristic reorientation temperature, can be tuned using hydrostatic pressure, which provides an effective method of controlling the Néel vector in $CrPS_4$. As presented in **Fig. 2** (see next page), high hydrostatic pressure favors the in-plane anisotropy, and increasing pressure reduces T_{SR} with an increasing temperature range of phase coexistence. The authors claim that the tunable magnetic anisotropy between the easy axis and easy plane provides an ideal platform for producing and investigating 2D Ising-type magnetism and the Berezinskii–Kosterlitz–Thouless regime of 2D magnetism. Specifically, $CrPS_4$ opens a pathway to the evolution of the universality classes of magnetism through the use of ATMs. Yang and his collaborators experimentally demonstrated the control of spin orientation and metamagnetic transition

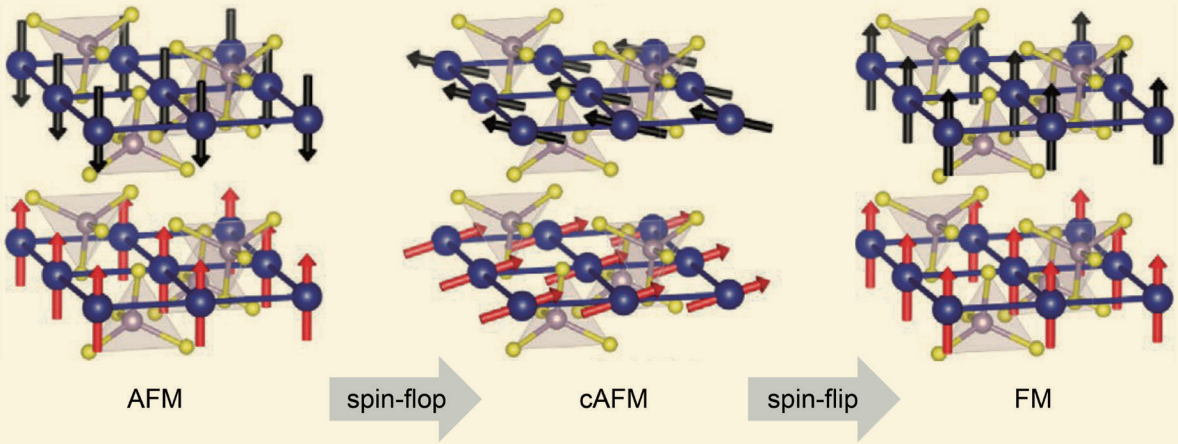

Fig. 1: The two successive field-induced transitions of $CrPS_4$. The antiferromagnetic magnetic structure first undergoes a spin-flop transition to a canted AFM state and then a spin-flip transition to the ferromagnetic state. [Reproduced from Ref. 1]

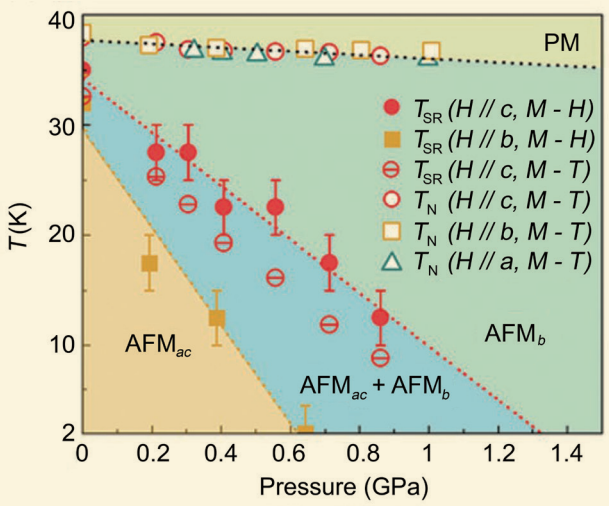

Fig. 2: Temperature-pressure magnetic phase diagram of $CrPS_4$ determined from magnetometry data. The blue region between the AFM_{ac} and AFM_b phases represents the coexistence of both phases (AFM_{ac}+ AFM_b), reflecting the first-order nature of the magnetic phase transition. [Reproduced from Ref. 2]

in a vdW AFM structure; this experiment may contribute to the understanding and tunability of 2D magnet materials and stimulate future developments in 2D AFM spintronics. (Reported by Chin-Wei Wang)

This report features the work of Wenyun Yang and his collaborators published in Adv. Mater. ***32****, 2001200 (2020) and Adv. Funct. Mater.* ***32****, 2106592 (2022).*

ANSTO ECHIDNA – High-resolution Powder Diffractometer

- NPD
- Materials Science, Condensed-matter Physics

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Field-Induced Magnetic Ordering in a Tetrahedral Sublattice with Strong Magnetic Anisotropy

Neutron powder diffraction is a powerful tool for studying magnetic properties in extreme sample environments.

Geometric frustration in condensed matter systems is a phenomenon in which competing forces act on atoms reside on a regular lattice. Because of the frustration in the geometry, such a system exhibits degenerate ground states, preventing entrance into the long-range order phase. An example is geometrically frustrated antiferromagnets, where antiferromagnetic couplings with neighboring spins cannot be satisfied simultaneously. Pyrochlore compounds, with corner-sharing tetrahedral sublattices, are the most well-known and studied geometrically frustrated systems and undertake various magnetic phases. On the basis of degenerate ground states, a perturbation, such as a dipolar interaction, stabilizes spin ice behavior in $Ho_2Ti_2O_7$ and $Dy_2Ti_2O_7$. Likewise, the conduction electron-mediated coupling, namely the Ruderman–Kittel–Kasuya–Yosida (RKKY) interaction, can serve as perturbation, causing novel magnetic phases in intermetallic compounds composed of geometrically frustrated sublattices. $Ho_5Co_6Sn_{18}$ is one example. Chin-Wei Wang (NSRRC) and co-workers reported comprehensive neutron scattering results for this compound.

Two distinct crystallographic Ho sites, Ho(1) and Ho(2), exist in $Ho_5Co_6Sn_{18}$. Both sublattices contain tetrahedral arrangements and can be magnetically frustrated. Each Ho(1) atom has 12 adjacent Ho(1) atoms, forming edge-sharing tetrahedrons. Conversely, the $Ho(2)_4$ tetrahedra are somewhat isolated; $Ho(2)_4$ units are separated by Ho(1) atoms and together form a rock salt structure. No significantmagnetic order exists in their magnetometry, while their heat capacity exhibit a peak at ~3.4 K, that is closed to the superconducting gap of tin metal. The temperature dependent magnetic peak intensity is measured on **SIKA,** that indicates the 3.4 K peak in heat capacity is associated to the long-range magnetic order. The data for neutron powder diffraction (NPD) performed at 1.5 K on **ECHIDNA** indicate a ferromagnetic structure on the Ho(1) sublattice and no ordered moment on the Ho(2) sublattice. A further investigation at the ultralow temperature of 60 mK demonstrated the magnetic order of Ho(2) spins; the Ho(2) spins on a $Ho(2)_4$ tetrahedron were (Mx, My, 0), (–Mx, –My, 0), (My, –Mx, 0), and (–My, Mx, 0). A 2-in-2-out configuration that is analogous to the magnetic