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The phase diagram of 2D antiferromagnets

The magnetic phase diagram of thin-layered antiferromagnets is revealed experimentally by investigating the tunnelling conductance as a function of magnetic field. A rich magnetic behaviour in $CrCl_3$ is uncovered, from which relevant magnetic information is extracted that is not easily available with other approaches.

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The recent discovery of the emerging family of (2D) magnetic van der Waals (vdW) heterostructures, such as CrI_3, $CrBr_3$, $CrCl_3$, $Cr_2Ge_2Te_6$, Fe_3GeTe_2, VS_2 and VSe_2 makes it possible to explore magnetic properties down to the two-dimensional limit [1, 2]. It further enables to engineer all-vdW ultra-compact spintronic devices and architectures by shuffling those magnetic layers with selected 2D materials (graphene, hBN, transition metal dichalcogenides, etc). Following the first report of unprecedented tunnelling magnetoresistance in vertical magnetic tunnel junctions [3], considerable excitement and flourishing new findings are paving the way towards the fabrication of heterostructures with potentially disruptive impact in next generation non-volatile memory technologies, such as spin torques-driven MRAM [4,5].

The magnetic phenomenology of 2D magnetic insulators with strong Ising anisotropy is well understood, reflecting that individual layers just flip their magnetization under the presence of a magnetic field. However, in weakly anisotropic 2D antiferromagnets, such as $CrCl_3$, the competition amongst energy scales with comparable magnitude (non-dominant magnetic anisotropy, the interlayer exchange coupling and the Zeeman energy) is expected to result in a much richer phase diagram. Writing in *Nature Nanotechnology*, Zhe Wang and co-workers succeed in building up the phase diagram of $CrCl_3$ by fabricating and probing, by magnetoconductance measurements, the in-depth magnetic response of high-quality encapsulated hBN/graphite/$CrCl_3$/graphite tunnel junctions [6].

Their study offers a comprehensive and detailed analysis of the magnetization evolution of multilayer $CrCl_3$ upon varying an external control magnetic field, temperature and thickness of the material. Atomically-thin $CrCl_3$ is a model layered antiferromagnetic insulator with an easy plane normal to the c-axis, so that the polarization of each layer is in-plane without a preferred orientation. The inherent magnetic states present universal trends when varying from odd to even number of layers, evidencing the impact and relative magnitudes of interlayer exchange coupling, individual layer magnetization and the shape anisotropy (Fig. 1).

The transition from the antiferromagnetic to a ferromagnetic state (occurring at a characteristic critical spin flip field H_2) is captured by a simple antiferromagnetic linear chain model and mean-field theory considerations, with a dependence (for applied in-plane external field) given by $H_2 = 4J/(\mu_0 M_s) \cos^2(\pi/2N)$, with N the number of layers). The only free parameter of the model, adjusted to get a nearly perfect fit to all experimental data, provides an interlayer exchange coupling of J = 86 μeV. This remarkable result, together with the rest of the analysis, provides new insights into the magnetic phase diagram of the $CrCl_3$-based vdW materials.

These findings constitute a milestone for the field of 2D magnetic materials. In combination with an increasing number of related contributions, some of them published simultaneously [7-10] will serve as pathfinder for further characterization studies as well as to design suitable material combinations to boost applications in the field of vdW heterostructures-based spintronics. Furthermore, experimental realizations and control of a strictly 2D magnetic insulator with in-plane magnetization could provide a novel testbed of the 2D XY-model, hence allowing the study of the Berezinskii–Kosterlitz–Thouless transition [11], while providing model analogies to thin film superfluids, superconductors and liquid crystals [12,13].

There are still some remaining features in the magnetoconductance that are not understood and require further scrutiny, such as a dip around zero magnetic field in odd-N multilayers. In addition, an outstanding challenge to impact future applications is to overcome the low working temperature, limited by the Curie or Néel temperatures of the 2D magnetic structures (18 K for $CrCl_3$). Indeed, room-temperature magnetic materials need to be sought after. Possible candidates include VSe_2 and electrostatic doped Fe_3GeTe_2, showing Curie temperatures above 300 K [14, 15]. In this context, the careful fundamental study presented by Wang et al. [6] should inspire further systematic analysis of the magnetic phase diagram of all these different types of materials with larger working temperature.

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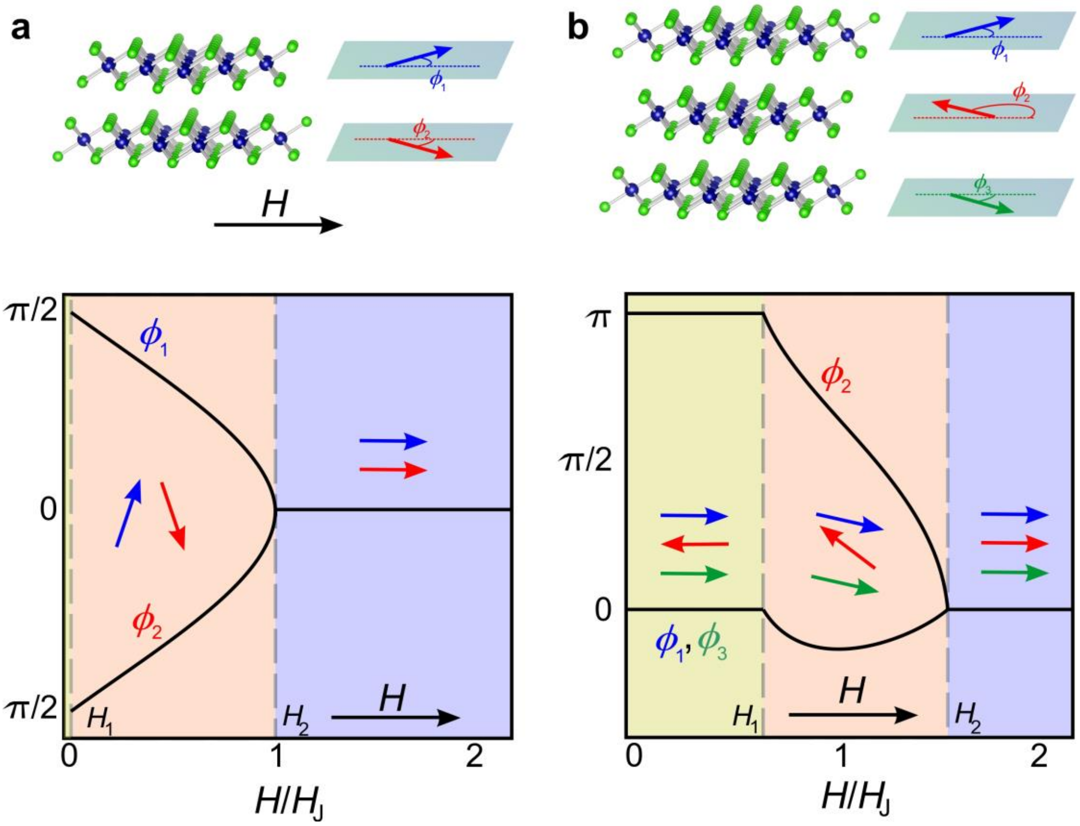

Fig. 1| Magnetic state of $CrCl_3$, as a function of applied in-plane magnetic field ***H***. **a** and **b** show respectively the cases for even and odd number of layers N, represented by $N = 2$ and $N = 3$. For even N, the layers magnetizations become nearly perpendicular to ***H*** at $H = H_1 \approx 0$, they tilt as H increases, until $H = H_2$ where the spin flip transition occurs. For odd N, the magnetizations orient along ***H*** direction for $H < H_1$ (either parallel or antiparallel to ***H***), they start rotating at $H > H_1$ and eventually align with ***H*** at $H = H_2$.