

This is the submitted version of the article:

Valenzuela S.O., Roche S.. The phase diagram of 2D antiferromagnets. *Nature Nanotechnology*, (2019). 14. : 1088 - . 10.1038/s41565-019-0592-x.

Available at: <https://dx.doi.org/10.1038/s41565-019-0592-x>

The phase diagram of atomically-thin layered antiferromagnets

Atomically thin layered antiferromagnets with weak magnetic anisotropy present a very rich magnetic behaviour. Their magnetic phase diagram is revealed experimentally by investigating the tunnelling conductance as a function of magnetic field.

Sergio O. Valenzuela and Stephan Roche

Institució Catalana de Recerca i Estudis Avançats (ICREA) and Catalan Institute of Nanoscience and Nanotechnology (ICN2), CSIC and The Barcelona Institute of Science and Technology (BIST), Barcelona, Spain

SOV@icrea.cat, stephan.roche@icn2.cat

The recent discovery of the emerging family of (2D) magnetic van der Waals (vdW) heterostructures, such as CrI_3 , CrBr_3 , CrCl_3 , $\text{Cr}_2\text{Ge}_2\text{Te}_6$, Fe_3GeTe_2 , VS_2 and VSe_2 makes it possible to explore magnetic properties down to the two-dimensional limit [Gibertini 2019, Gong 2019]. 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 [Song 2018], 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 [Alghamdi 2019, Wangx 2019].

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 [Wangz 2019].

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 magnetic material. Atomically-thin CrCl_3 is a paradigmatic 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).

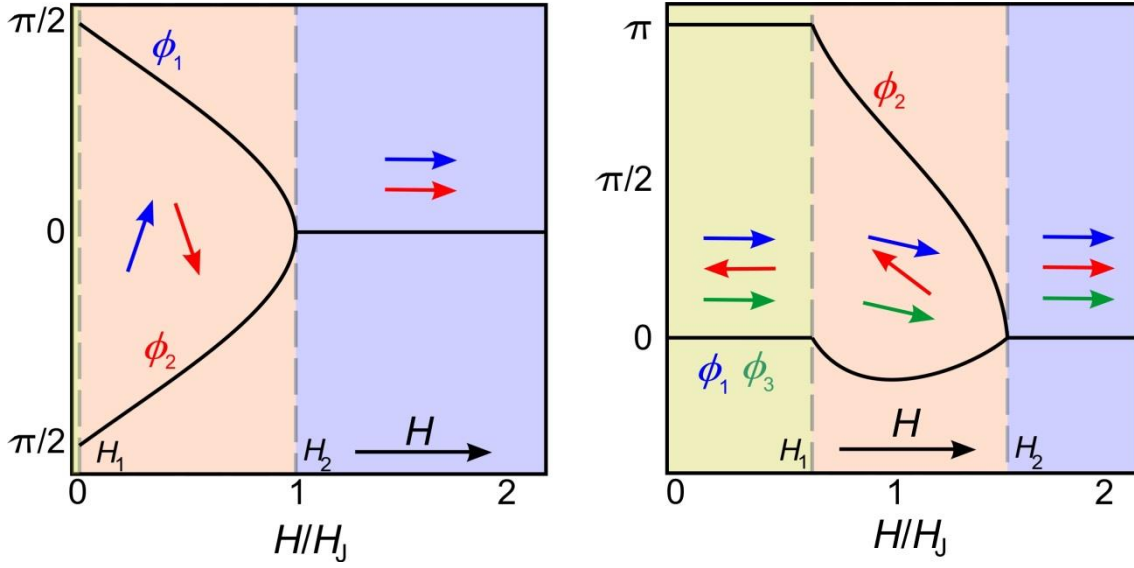


Fig. 1 | Magnetic state of CrCl_3 , as a function of applied in-plane magnetic field H , for even (a) and odd (b) number of layers N , represented by $N = 2$ and $N = 3$, respectively. For even N , the layers magnetizations become nearly perpendicular to H at $H = H_1 \approx 0$, they tilt as H increases, 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 $H = H_2$.

Quite spectacularly, 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 $J = 86 \mu\text{eV}$. 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 [Cao 2019, Klein, Kim1, Kim2] 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 [KT 1973], while providing model analogies to thin film superfluids, superconductors and liquid crystals [de Jong 1974, Mondal 2011].

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 [Deng 2018; Bonilla 2018]. In this context, the careful fundamental study presented by Wang et al. [Wangz 2019] should inspire further systematic analysis of the magnetic phase diagram of all these different types of materials with larger working temperature, leading the way towards revolutionary advances for data storage technologies.

References

[Gibertini 2019] M. Gibertini, M. Koperski, A. F. Morpurgo & K. S. Novoselov, *Magnetic 2D materials and heterostructures*, Nature Nanotechnology 14, 408–419 (2019).

[Gong 2019] C. Gong and X. Zhang, *Two-dimensional magnetic crystals and emergent heterostructure devices*, Science 363, Issue 6428, eaav4450 (2019).

[Song, 2018] T. Song, X. Cai, M. W.-Y. Tu, X. Zhang, B. Huang, N. P. Wilson, K. L. Seyler, L. Zhu, T. Taniguchi, K. Watanabe, M. A. McGuire, D. H. Cobden, D. Xiao, W. Yao, X. Xu, *Giant tunneling magnetoresistance in spin-filter van der Waals heterostructures*, Science 360, 1214 (2018).

[Alghamdi 2019] M. Alghamdi, M. Lohmann, J. Li, P. R. Jothi, Q. Shao, M. Aldosary, T. Su, B. P. T. Fokwa, J. Shi, *Highly efficient spin-orbit torque and switching of layered ferromagnet Fe_3GeTe_2* , Nano Lett. 19, 4400 (2019).

[Wangx 2019] X. Wang, J. Tang, X. Xia, C. He, J. Zhang, Y. Liu, C. Wan, C. Fang, C. Guo, W. Yang, Y. Guang, X. Zhang, H. Xu, J. Wei, M. u Liao, X. Lu, J. Feng, X. Li, Y. Peng, H. Wei, R. Yang, D. Shi, X. Zhang, Z. Han, Z. Zhang, G. Zhang, G. Yu, X. Han, *Current-driven magnetization switching in a van der Waals ferromagnet Fe_3GeTe_2* . Science Advances 5, eaaw8904 (2019).

[Wangz 2019] Z. Wang.... Nature Nanotechnology [2019]

[Cai 2019] Xinghan Cai, Tiancheng Song, Nathan P. Wilson, Genevieve Clark, Minhao He, Xiaouu Zhang, Takashi Taniguchi, Kenji Watanabe, Wang Yao, Di Xiao, Michael A. McGuire, David H. Cobden, and Xiaodong Xu, *Atomically Thin CrCl_3 : An In-Plane Layered Antiferromagnetic Insulator*, Nano Lett. 19, 63993-3998 (2019).

[Klein] Dahlia R. Klein, David MacNeill, Qian Song, Daniel T. Larson, Shiang Fang, Mingyu Xu, R. A. Ribeiro, P. C. Canfield, Efthimios Kaxiras, Riccardo Comin & Pablo Jarillo-Herrero, *Enhancement of interlayer exchange in an ultrathin two-dimensional magnet*, Nature Physics (2019), 10.1038/s41567-019-0651-0.

[Kim1] H.H. Kim et al. *Evolution of interlayer and intralayer magnetism in three atomically thin chromium trihalides*, PNAS 116 (23) 11131-11136 (2019).

[Kim2] Hyun Ho Kim* Bowen Yang Shangjie Tian Chenghe Li Guo-Xing Miao Hechang Lei Adam W. Tsen, *Tailored Tunnel Magnetoresistance Response in Three Ultrathin Chromium Trihalides*, Nano Lett. 19 85739-5745 (2019)

[KT 1973] Kosterlitz, J.M. & Thouless, D.J. Ordering, *Metastability and phase transitions in two-dimensional systems*, Journal of Physics C: Solid State Physics 6, 1181 (1973)

[de Jong 1974] de Jongh, L.J. & Miedema, A.R. *Experiments on simple magnetic model systems*. Advances in Physics 23, 1-260 (1974).

[Mondal 2011] Mondal, M. et al. *Role of the Vortex-Core Energy on the Berezinskii-Kosterlitz-Thouless Transition in Thin Films of NbN*, Physical Review Letters 107, 217003 (2011).

[Deng 2018] Y. Deng, Y. Yu, Y. Song, J. Zhang, N. Z. Wang, Z. Sun, Y. Yi, Y. Z. Wu, S. Wu, J. Zhu, J. Wang, X. H. Chen, Y. Zhang, *Gate-tunable room-temperature ferromagnetism in two-dimensional Fe₃GeTe₂*, Nature 563, 94 (2018).

[Bonilla 2018] M. Bonilla, S. Kolekar, Y. Ma, H. C. Diaz, V. Kalappattil, R. Das, T. Eggers, H. R. Gutierrez, M. Phan, M. Batzill, *Strong room-temperature ferromagnetism in VSe₂ monolayers on van der Waals substrates*, Nature Nanotechnology, 13, 289 (2018).