The emergence of magnetism in two dimensions

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2. **Giant tunneling magnetoresistance in spin-filter van der Waals heterostructures**
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3. **Electric-field switching of two-dimensional van der Waals magnets**
   Authors: S. Jiang, J. Shan and K. F. Mak
   Nat. Mater. **17**, 406410 (2018); doi: 10.1038/s41563-018-0040-6.

Recommended with a Commentary by Luis Balicas, National High Magnetic Field Lab and Florida State University

Magnetism in two-dimensions has the potential to open a number of technological opportunities, ranging from magnetic memories, to sensing, to spintronics to novel functionalities based on heterostructures composed of two-dimensional materials. However, according to the MerminWagner theorem, magnetic order would not occur in the isotropic two-dimensional Heisenberg models at finite temperatures [1].

A recent publication [2] on the monoclinic CrI_3 semiconducting compound have shown for the first time that magnetism, or more precisely ferromagnetism (FM), does not only survive down to the two-dimensional limit [2] but also that the nature of the magnetic order, FM or antiferromagnetic (AFM) can depend upon the number of layers. CrI_3 is a layered compound characterized by planar covalent bonds and weak inter-planar coupling which allows one to mechanically exfoliate it down to the monolayer limit. The bulk crystal orders at 61 K while displaying a pronounced magnetic anisotropy with its magnetic moments pointing perpendicularly to the layers [2, 3, 4] ($\sim 3\ \mu_B$ per Cr^{+3} ions with these forming a honeycomb

lattice). In contrast to the monolayer, two adjacent layers order antiferromagnetically with their inter-planar magnetic moments pointing in opposite directions.

Ref. [2] carefully analyzed the magnetic response of CrI_3 as a function of both the number of layers and of the magnetic field applied perpendicularly to them via polar magneto-optical Kerr effect (MOKE) measurements (Faraday geometry). The authors find pronounced hysteresis or coercive fields associated with the polarization of the moments in monolayers and in tri-layers (both ferromagnetic) but the near absence of hysteresis in bilayers, which in turn display a metamagnetic transition towards the spin-polarized state.

Subsequently, the same University of Washington group, built magnetic tunnel junctions (MTJ) based on the spin-filter effect after stacking CrI_3 bilayers with graphene (acting as electrodes) and hexagonal boron nitride (acting as encapsulating layers) [3]. The authors observe a dramatic increase in inter-layer tunneling current whenever the magnetic field surpasses the critical field required to polarize the magnetic moments in bilayers, trilayers and also in four-layer stacks. Their inter-layer resistance changes between several hundred percent in the bi-layers to values exceeding 10^4 percent in four-layers when they are driven from the spin unpolarized to the polarized state with an external magnetic field. According to the authors of Ref. [3] four-layer stacks exceed, by one order of magnitude, values extracted from conventional MTJs under similar experimental conditions.

In field-effect transistors (FETs) based on CrI_3, Ref. [4] claims that it is possible to control the magnetic state of bilayers through the application of a small gate-voltage when the FETs are brought into the vicinity of the metamagnetic transition that leads to the spin-polarized state. When the external magnetic field barely exceeds the spin-flip critical-field, the application of a gate-voltage can suppress the spin-polarized state re-stabilizing the AF-ordered state. This observation would correspond to a novel type of magnetoelectric-effect although a microscopic mechanism remains to be developed. The authors have also applied a periodic gate electric field to drive the system forth and back between both magnetic states finding that this switching survives an undefined number of cycles. Ref. [4] suggests that this magnetoelectric-effect would originate from the electric-field dependence of the interlayer exchange bias.

Beyond the potential for applications implied by these observations, several fundamental questions remain to be addressed: i) why the interaction with the substrate does not affect the magnetic order in the monolayers? ii) Why does the ferromagnetism in the monolayer remain completely oblivious to the absence of an adjacent layer? iii) What is the origin of the magnetic anisotropy in this compound? iv) What is the precise role played by the electric-field concerning the claimed magnetoelectric-effect in a compound that displays inversion symmetry?

Notice that there are a number of other rare-earth based trihalides for which there is virtually no magnetic data in the literature. Examples include $CeIn_3$, DyI_3, TbI_3, HoI_3, etc., and their Cl and Br variants. For instance, TbI_3 would seem to display the same crystallographic structure as CrI_3 at lower temperatures, that is $R\bar{3}h$ (148), suggesting that it might also be exfoliable. The fact that Tb displays a large free moment $\sim 9\ \mu_B$ and orders ferromagnetically at relatively high temperatures, i.e. $T_c \sim 230$ K, suggests that TbI_3 might display magnetic order at much higher temperatures when compared to CrI_3, opening perhaps a real potential for applications based on magnetic two-dimensional materials. Nevertheless, its magnetic properties could be considerably more complex than those of CrI_3

given its large spin orbit coupling and the role played by its crystalline electric field scheme. Notice also that none of these compounds are stable under ambient conditions thus their handling requires protocols to avoid air exposure.

Here, the question is if we are observing the dawn of a new era corresponding to the emergence of magnetism in truly two-dimensional insulating systems and to its control via an electric field.

References

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[3] *Giant tunneling magnetoresistance in spin-filter van der Waals heterostructures*, T. Song, X. Cai, M. W.-Y. Tu, X. Zhang, B. Huang, N. P. Wilson, K. L. Seyler, L. Zhu, Takashi Taniguchi, K. Watanabe, M. A. McGuire, D. H. Cobden, D. Xiao, W. Yao, and X. Xu, Science **360**, 1214 (2018); doi:10.1126/science.aar4851.

[4] *Electric-field switching of two-dimensional van der Waals magnets*, S. Jiang, J. Shan and K. F. Mak, Nat. Mater. **17**, 406410 (2018); doi: 10.1038/s41563-018-0040-6.