

# Berezinskii-Kosterlitz-Thouless phase transition in a 2D-XY ferromagnetic monolayer

Jiesu Wang<sup>†</sup>

Beijing Academy of Quantum Information Sciences, Beijing 100193, China

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It has been widely discussed whether long-range magnetic order could exist in low-dimensional ( $d < 3$ ) systems. Early studies, like Hohenberg-Mermin-Wagner theorem based on the Heisenberg model of magnetism, incline to a negative conclusion, as the thermal fluctuation at any finite temperature prevents the formation of long-range magnetic order<sup>[1, 2]</sup>. However, further studies theoretically predicted an unusual long-range magnetic order in the two-dimensional (2D) systems that are described by a so-called 2D-XY model, resulting from the formation of bound pairs of vortices and anti-vortices with opposite chirality<sup>[3, 4]</sup>. The induced ferromagnetism is suppressed as the 2D system is heated over a critical temperature, accompanied with the destruction of those bound pairs and thus the long-range magnetic order. This mechanism of phase transition is known as Berezinskii-Kosterlitz-Thouless (BKT) formalism<sup>[3-5]</sup>.

To determine the mechanism of a phase transition, people usually employ the “critical exponents” which relies only on the underneath mechanism itself, rather than specific material systems. In a ferromagnetic material, the magnetization is a good order parameter, which evolves as  $M = M_0(1-T/T_C)^\beta$  as the temperature rises, where  $\beta$  is the so-called critical exponent that defines the universality class of the phase transition. Fig. 1 displays the comparison of three main mechanisms of ferromagnetic phase transition (Heisenberg model in bulk, 2D-XY model, and 2D-Ising model) and the corresponding Hamiltonians<sup>[6, 7]</sup>, as well as the theoretical  $M-T$  curves around the critical temperature. In 2D systems,  $\beta = 1/8$  for Ising ferromagnetism while  $\beta = 3\pi^2/128 \approx 0.231$  for 2D-XY ferromagnetism<sup>[8]</sup>. Although researchers have created plenty of magnetic metal and oxide thin films through various methods, the strong interaction between films and substrates or between the atomic layers in the film make these systems difficult for verifying the BKT scenario. Recently, the experimental characterization of a series of 2D ferromagnetic materials with van der Waals layered structures, such as  $\text{CrI}_3$ <sup>[9]</sup>,  $\text{CrBr}_3$ <sup>[10]</sup>,  $\text{Cr}_2\text{Ge}_2\text{Te}_6$ <sup>[11]</sup>,  $\text{Fe}_3\text{GeTe}_2$ <sup>[12]</sup>,  $\text{VSe}_2$ <sup>[13]</sup>, etc. have been reported. However, none of these materials has a magnetic easy plane anisotropy.

Writing in Science (<https://www.science.org/doi/10.1126/science.abd5146>), Amilcar Bedoya-Pinto, Kai Chang, Stuart S. P. Parkin and their colleagues reported the growth and characterization of a nearly ideal easy-plane system, a single  $\text{CrCl}_3$  monolayer<sup>[8]</sup>. The van der Waals layered structure of this film-

substrate system minimized the out-of-plane interaction. By measuring the intrinsic magnetic properties using element-resolved X-ray magnetic circular dichroism (XMCD) technique, they observed robust ferromagnetic order with critical scaling characteristic of a 2D-XY system in the  $\text{CrCl}_3$  monolayer. Their variant temperature studies proved that the nature of the ferro- to paramagnetic phase transition in  $\text{CrCl}_3$  monolayer is of the BKT-type with a ferromagnetic Curie temperature ( $T_C$ ) of 12.95 K.

In order to create a magnetic system with easy plane, i.e. continuous rotational symmetry  $O(2)$ , the authors used molecular beam epitaxy (MBE) technique to grow  $\text{CrCl}_3$  monolayers on graphitized 6H-SiC(0001) substrates that has very low surface energy. Fig. 2 shows the lattice structure of  $\text{CrCl}_3$  monolayer featuring a Cl-Cr-Cl trilayer structure and a honeycomb Cr sublattice. The scanning tunneling microscopy (STM) results indicate nearly perfect coverage and extremely low defect density in a  $\text{CrCl}_3$  monolayer. Further tunneling spectroscopic studies reveal an intrinsic bandgap of 1.6 eV and negligible charge transfer from the substrate<sup>[14]</sup>. Thus, it is appropriate to take the van der Waals  $\text{CrCl}_3$  monolayer as a quasi-freestanding 2D system.

Having solved the challenge of sample growth, the authors had to handle the other problem of the precise measurement of the magnetic properties, in order to explore the phase transition mechanism of the  $\text{CrCl}_3$  van der Waals monolayer, whose magnetic signal is extremely weak because of its ultralow thickness. Traditional magnetic measurements through techniques like SQUID (superconducting quantum interference device), MOKE (magnetic optical Kerr effect), or electric transport experiment only focus on the overall magnetization, so any subtle magnetic impurities can introduce huge deviation from the intrinsic signal. The authors turned their eyes to XMCD technology<sup>[15]</sup>. Unlike the methods mentioned above, XMCD is based on the difference between the X-ray absorption spectra taken in left and right circularly polarized light, thus can distinguish the magnetic moments that reside in different elements and extract the intrinsic properties of the monolayers by excluding the extra signal from impurities.

The magnetic features of the  $\text{CrCl}_3$  monolayer are obtained by XMCD signal from the  $L_3$  edge of  $\text{Cr}^{3+}$ , because Cr contributes the major part of the magnetization. Field-dependent XMCD hysteresis loops clearly demonstrated the magnetic anisotropy with an easy plane. When an in-plane magnetic field is applied, the authors observed that the hysteresis loops gradually shrink as the system is heated up. The vanishment of both the residual magnetization and coercive field imply a critical temperature of  $\sim 13$  K. The temperature-depend-

Correspondence to: J S Wang, [wangjs@baqis.ac.cn](mailto:wangjs@baqis.ac.cn)

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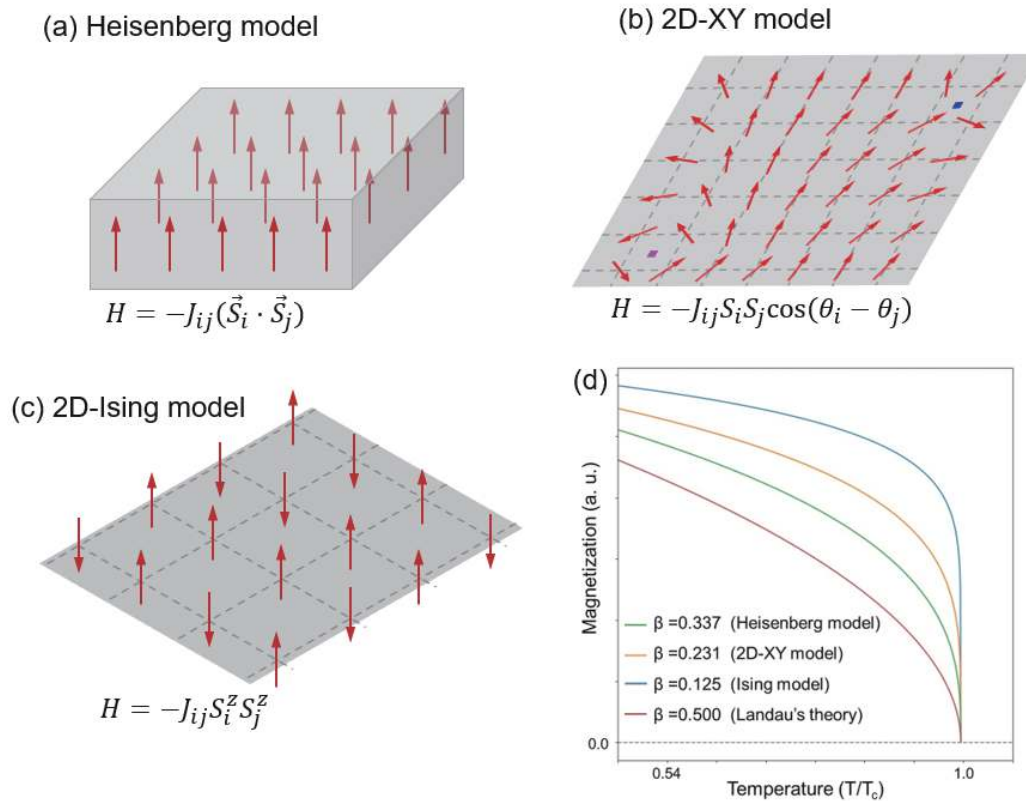


Fig. 1. (Color online) (a–c) Schematic diagrams of different mechanisms of ferroelectric phase transition without applied external magnetic field in (a) Heisenberg model, (b) 2D-XY model, and (c) 2D-Ising model. (d) Theoretical comparison of  $M$ – $T$  curves around  $T_C$  under different mechanisms indicated by corresponding critical exponent  $\beta$ .

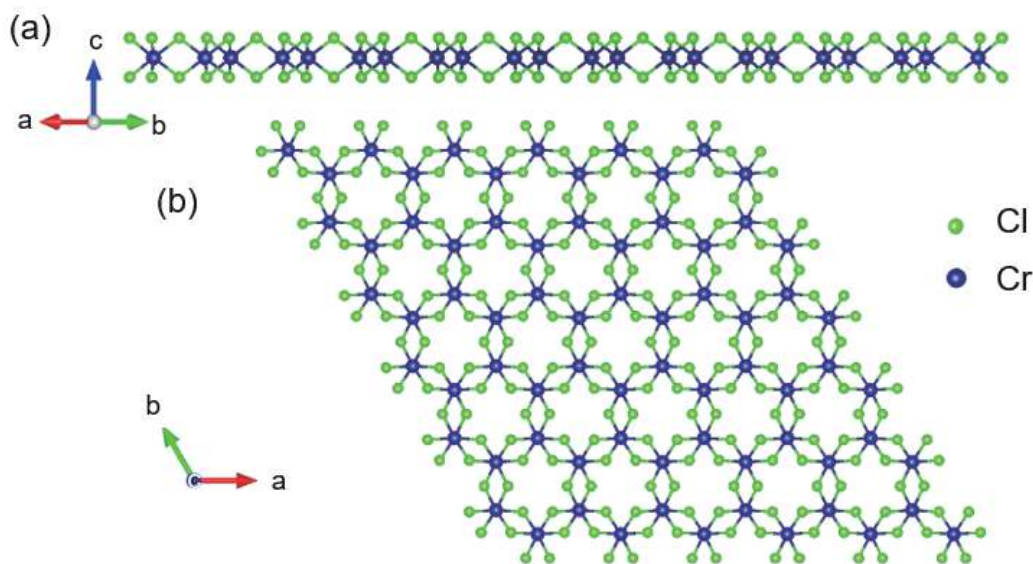


Fig. 2. (Color online) Lattice structure of the  $\text{CrCl}_3$  monolayer in (a) side view and (b) top view. The sublattice constructed by Cr atoms can be clearly seen.

ent XMCD data near  $T_C$  can be well fitted by the formula above with  $\beta = 0.227$ , which is very close to the theoretical value in 2D-XY model. This result, together with the extra analysis reported in the article, support the BKT nature of the related phase transition.

The discovery of intrinsic 2D-XY ferromagnetism as well as the demonstration of a BKT-type ferromagnetic phase transition at the 2D limit are of great importance for researching

the magnetism and spintronics in low-dimensional quantum materials. Because of the van der Waals nature at the interface between  $\text{CrCl}_3$  monolayer and the substrate, the interaction such as hybridization, bonding, substrate-driven crystalline anisotropy, and other interface effects are minimized, resulting in a nearly ideal 2D-XY experimental system that is promising for further studies in spin superfluids<sup>[16, 17]</sup>, 2D topological magnetic orders<sup>[18, 19]</sup>, and other exotic phenomena.

As claimed in their article, further studies on regulating the 2D-XY behavior in the monolayer are expected, such as changing the in-plane anisotropy fields induced by different substrates, and exploring the finite-size effects of BKT phase transition by altering the grain scale during the MBE growth process.

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**Jiesu Wang** earned her bachelor's degree from Minzu University of China in 2013, and her PhD from Institute of Physics, Chinese Academy of Sciences (IOP, CAS) in 2018. Subsequently, Wang worked as a postdoc. in Prof. Kuijuan Jin's group in IOP, CAS. Since June 2020, Wang has started as an Assistant Research Scientist in Beijing Academy of Quantum Information Sciences.