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Magnetic order in the archetypical 2D van der Waals magnet CrI3

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Two-dimensional van der Waals materials host a diverse landscape of optical, electronic and topologically non-trivial states in the monolayer limit and have therefore fuelled a search for the next candidate materials that may enable novel information technologies. The magnetic features can be controlled by different processes including thickness, doping and other tuning parameters. CrI3 specifically shows a changing magnetic order and strong pressure dependence between ferromagnetic and antiferromagnetic in the limit of a few monolayers. However, it has recently become clear that even the bulk material hosts a much richer behaviour than initially assumed caused by the coexistence of different crystal structures.

In a recent muSR study some of us revealed that bulk CrI3 shows several magnetic phases with distinct transitions below the established TC. Remarkably, the traditionally defined TC does not correspond to long range magnetic order in the full volume of the crystal but rather to a partial transition where less than 25% of the crystal is spin-ordered. In total three distinct magnetic phases were identified with specific amount of crystal structures (monoclinic and rhombohedral).

Here we present a small angle neutron scattering study of the magnetic correlations in bulk CrI3. We find a complex behaviour in this inhomogeneous ferromagnet below TC, with several temperature regimes which are dominated by very distinct correlation lengths, including a regime of phase coexistence.

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