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Resolving spin reorientation of CrCl₃ induced by high-pressure with MuSR and neutron diffraction

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Researches on two-dimensional (2D) materials have attracted tremendous attention both from fundamental and applied sciences since accelerated by the discovery of graphene. Among a large number of 2D materials, chromium trihalides CrX₃ (X = Cl, Br, I) van der Waals (vdW) magnets have also raised a large interest due to the existence of many magnetic subtleties that cannot be explained by their magnetic and/or structural transitions.

Numerous studies were performed on CrI₃, but only a few have been reported so far on its analogue CrCl₃. The 2D vdW CrCl₃ compound is stabilized under a rhombohedral symmetry, consisting of 2D Cr layers arranged in a honeycomb web fashion and surrounded by octahedrally coordinated Cl, with weak vdW inter-layer coupling. This makes CrCl₃ an ideal system to study under external stimuli such as pressure or magnetic field, where new intriguing states can be unveiled. Expectantly, studies of CrCl₃ under high pressure and room temperature have been reported. [1] However, its spin dynamics at low-temperature and high-pressure regimes remain unexplored. Motivated by the variability of the spin degree of freedom and spin dynamics under such conditions, we performed muon spin rotation (MuSR) and neutron powder diffractions (NPD) on ambient and hydrostatically pressured CrCl₃ up to 23 kbar down to 2 K. [2,3]

In this study, by incorporating the two techniques and high-pressure, we resolved a suppression of the magnetic ground state and a stronger relaxation rate by MuSR. Within the magnetically ordered states, a spin reorientation was also observed by NPD at high pressure. A linear extrapolation points toward the suppression of magnetism at about $p_c = 30$ kbar indicating the possible existence of a critical point at p_c . [3]

[1] Ahmad, A., et al. *Nanoscale* 12.45 (2020): 22935-22944.

[2] Forslund, O., et al. arXiv:2111.06246 (2021).

[3] Ge, Y., et al., in preparation.

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