

Controlling the Magnetic Properties of the Layered van der Waals Crystals



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Layered van der Waals materials have gained an ever-increasing interest due to their ability to provide a versatile playground to explore the emergence of new quantum phenomena. Among these, the trihalides CrI_3 and VI_3 stand out due to their unique magnetic properties and the interplay with their structural transitions,^[1,2] with recent studies focussing on elucidating the effect of layer control in CrI_3 ,^[3] and on the importance of the spin and orbital moments in VI_3 .^[4] To date, only one experimental study exists on their elemental doping,^[5] and none on the solid solution due to its significant synthetic challenge.

Our work showcases the successful growth of single crystals across the entire range of the solid solution of $\text{V}_{1-x}\text{Cr}_x\text{I}_3$, *i.e.*, $x = 0 - 1$, by employing the self-selecting vapour-growth (SSVG) technique.^[6] In the resulting magnetic measurements, we identified three regimes. In the $0.1 \leq x \leq 0.3$ regime, we observed complex ferri- and ferromagnetic behaviour parallel and perpendicular to the crystallographic *c*-axis, and spin-glass-like behaviour. For $0.4 \leq x \leq 0.6$, we observed a transition from one to two ferri- and ferromagnetic transitions in both orientations, while for $0.7 \leq x \leq 0.9$, we observed two ferri- and ferromagnetic transitions parallel to the crystallographic *c*-axis.

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[4] A. De Vita et al., *Nano Letters*, 24, 1487-1493 (2024).

[5] S. Pan et al., *Journal of Alloys and Compounds*, 908, 164573 (2022).

[6] A. Lukovkina et al., *Chemistry of Materials*, 36, 6237-6245 (2024).