Crystal growth of new van der Waals materials solid solutions (*TM´*, *TM´´*)I_2 (*TM* = Co, Fe, Ni) with tuned magnetic properties

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Magnetic van der Waals materials have emerged as a class of materials that offer exciting possibilities due to their distinctive interplay of magnetic interactions and van der Waals forces, leading to a rich and diverse range of functionalities. In the growing family of novel magnetic van der Waals materials, transition metal di-iodides (TMI_2, *TM* = Co, Fe, Ni) represent an exciting group of compounds in which different magnetic ground states are stabilized by subtle differences in their crystal structure [1-3]. Competing AFM and FM magnetic orders are the origin of magnetic frustrations at low temperatures. Despite prior investigations of these frustrated states in some of these compounds, the elusive interconversion between magnetic ground states within solid solutions of these materials remains unresolved.

Crystal growth of these compounds requires careful adjustment of precursor composition, growth temperature, and thermal gradient all along the solid solution. The choice and control of synthesis conditions are crucial to avoid phase separation in a reactor. We have successfully grown crystals of the solid solutions (*TM´*, *TM´´*)I_2 (*TM* = Co, Fe, Ni) with the aim of tuning the magnetic transitions and searching for new magnetic ground states. Plate-like crystals up to 6x10 mm^2 in size could be obtained by chemical vapor transport. Interestingly, with this technique, TMI_2 crystals were found to grow both from the vapor at the cold end of the reactor and from the melt at the hot end. All these crystals are extremely air sensitive and require handling inside a glovebox under an inert atmosphere.

Bulk crystals of (*TM´*, *TM´´*)I_2 can be exfoliated into 2D thin flakes expected to have thickness-dependent magnetic behavior down to a monolayer, thus opening the possibility of fabricating new 2D magnetic devices with novel magnetic properties.

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[2] Qian Song et al, *Nature*, **2022**, 602, 601-605.

[3] T. Kurumaji et al, *Physical Review B*, **2013**, 87(1), 014429(9).