Controlling the Magnetic Properties of the van der Waals Multiferroic Crystals $Co_{1-x}Ni_xI_2$

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In the growing family of magnetic van der Waals materials multiferroic compounds CoI_2 and NiI_2 have drawn significant attention lately due to the multiferroicity down to the monolayer and frustrated cycloid ground state caused by Kitaev interactions, respectively [1-2]. Despite prior investigations of these materials, the evolution of magnetic ground states within the solid solution between them remains unresolved.

We have successfully grown crystals of the whole solid solution $Co_{1-x}Ni_xI_2$ by employing the self-selecting vapor growth (SSVG) technique and carefully tuning the synthesis conditions according to the composition. Both the lattice parameters and magnetic properties evolve continuously and smoothly from one end member to the other, showing that they can be finely chemically tuned. We also observe that the Ni substitution for Co affects the metamagnetic transition typical for CoI_2. In particular, we find the existence of this metamagnetic transition of CoI_2 in the NiI_2 structure. Based on magnetic measurements we construct the phase diagram of the $Co_{1-x}Ni_xI_2$ system.

Controlling the magnetic properties by the chemical composition opens new pathways for fabricating electronic devices made of two-dimensional (2D) flakes of multiferroic van der Waals materials.

[1] C. Kim, et al., Bond-dependent anisotropy and magnon decay in cobalt-based Kitaev triangular antiferromagnet. Nature Physics 2023, 19, 1624-1629.

[2] Q. Song, et al., Evidence for a single-layer van der Waals multiferroic. Nature 2022, 602, 601-605.