Abstract

Transition metal di- and tri-halides (TMH) have recently gathered research attention owing to their intrinsic magnetism all the way down to their two-dimensional limit. 2D magnets, despite being a crucial component for realizing van der Waals heterostructures and devices with various functionalities, were not experimentally proven until very recently in 2017. The findings opened up enormous possibilities for studying new quantum states of matter that can enable potential to design spintronic, magnetic memory, data storage, sensing, and topological devices. However, practical applications in modern technologies demand materials with various physical and chemical properties such as electronic, optical, structural, catalytic, magnetic etc., which cannot be found within single material systems. Considering that compositional modifications in 2D systems lead to significant changes in properties due to the high anisotropy inherent to their crystallographic structure, this work focuses on alloying of TMH compounds to explore the potentials for tuning their properties. In this thesis, the ternary cation alloys of \( \text{Co}(1-x)\text{Ni}x\text{Cl}_2 \) and \( \text{Mo}(1-x)\text{Cr}x\text{Cl}_3 \) were synthesized via chemical vapor transport at a various stoichiometry. Their compositional, structural, and magnetic properties were studied using Energy Dispersive Spectroscopy, Raman Spectroscopy, X-Ray Diffraction, and Vibrating Sample Magnetometry. It was found that completely miscible ternary alloys of \( \text{Co}(1-x)\text{Ni}x\text{Cl}_2 \) show an increasing Néel temperature with nickel concentration. The \( \text{Mo}(1-x)\text{Cr}x\text{Cl}_3 \) alloy shows potential magnetic phase changes induced by the incorporation of molybdenum species within the host CrCl3 lattice. Magnetic measurements give insight into potential antiferromagnetic to ferromagnetic transition with molybdenum incorporation, accompanied by a shift in the magnetic easy-axis from parallel to perpendicular. Phase separation was found in the \( \text{Fe}(1-x)\text{Cr}x\text{Cl}_3 \) ternary alloy indicating that crystallographic structure compatibility plays an essential role in determining the miscibility of two parent compounds. Alloying across two similar (TMH) compounds appears to yield predictable results in properties as in the case of \( \text{Co}(1-x)\text{Ni}x\text{Cl}_2 \), while more exotic transitions, as in the case of \( \text{Mo}(1-x)\text{Cr}x\text{Cl}_3 \), can emerge by alloying dissimilar compounds. When dissimilarity reaches a certain limit, as with \( \text{Fe}(1-x)\text{Cr}x\text{Cl}_3 \), phase separation becomes more favorable. Future studies focusing on magnetic and structural phase transitions will reveal more insight into the effect of alloying in these TMH systems.